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THE KELDYSH FORMALISM APPLIED TO TIME-DEPENDENT CURRENT-DENSITY-FUNCTIONAL THEORY

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Abstract   In this work we demonstrate how to derive the Kohn-Sham equations of time-dependent current-density functional theory from a generating action functional defined on a Keldysh time contour. These Kohn-Sham equations contain an exchange-correlation contribution to the vector potential. For this quantity we derive an integral equation. We further derive an integral equation for its functional derivative, the exchange-correlation kernel, which plays an essential role in response theory. The exchange-only limits of the latter equation is studied in detail for the electron gas and future applications are discussed.

1.   Introduction

Time-dependent density-functional theory (TDDFT) [1, 2] is a method for calculating properties of many-electron systems in time-dependent external fields. The theory has originally been formulated for systems in longitudinal electric fields, which can be described by a time-dependent scalar potential $v(\mathbf{r}t)$ (for reviews see [3, 4, 5]). The basis of this theory is the Runge-Gross theorem [2] which states that, for a given initial state, the external potential $v(\mathbf{r}t)$ (modulo an arbitrary purely time-dependent function $C(t)$) is a functional of the time-dependent density $n(\mathbf{r}t)$. This implies that every observable is a functional of the density and the initial state. The next step in TDDFT is then to introduce 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 $v_s(\mathbf{r}t)$. If one assumes that the initial state exists, then an explicit construction procedure for $v_s(\mathbf{r}t)$ can be given [6, 5, 7]. Once an approximation for $v_s(\mathbf{r}t)$ as a func-

tional of the density is given, the Kohn-Sham equations can be solved self-consistently. This time-dependent Kohn-Sham theory has had many successful applications and is widely used.

Nevertheless, the theory has some limitations and drawbacks. First of all, the theory only applies to systems for which the external field can be described by a scalar potential, i.e. longitudinal fields. This excludes, for instance, the important case of external light fields, at least if we want to go beyond the dipole approximation. Secondly, the theory can not deal with infinite systems with periodic boundary conditions as one does in standard band structure methods [8]. The reason is that we try to describe the properties of a finite sample of material (macroscopic on a atomic scale) with the idealized infinite system. As a consequence the long range effects which result from the boundary charges must be externally reintroduced in a theory in which the basic variable is the ground state density of the bulk. This therefore requires an extension of TDDFT. Thirdly, it was shown by Vignale and Kohn [9, 10] that if one wants to go beyond the simple adiabatic local density approximation and include memory effects into the density functionals, one consequently needs functionals that are very nonlocal in space. Although this is not a problem in principle, it can be a practical problem for constructing reliable approximations. A theory that does not suffer from these three drawbacks is time-dependent current-density-functional theory (TDCDFT). In this theory the time-dependent current $\mathbf{j}(\mathbf{r}t)$, rather than the density, is the basic variable.In this theory the pair of scalar and vector potentials $(v(\mathbf{r}t), \mathbf{A}(\mathbf{r}t))$ rather than the scalar potential $v(\mathbf{r}t)$ alone plays a crucial role. As in the case of TDDFT we can derive a theorem of the Runge-Gross type [11, 4]. For a given initial state the time-dependent current density $\mathbf{j}(\mathbf{r}t)$ uniquely determines the applied potentials $(v(\mathbf{r}t), \mathbf{A}(\mathbf{r}t))$ up to a gauge transformation. This implies that all physical observables are functionals of the current density and the initial state. On the basis of this theorem we can guarantee the uniqueness of a Kohn-Sham system. This Kohn-Sham system is defined to be that system which has the same current as the true system. On the basis of the continuity equation

$$\partial_t n(\mathbf{r}t) + \nabla \cdot \mathbf{j}(\mathbf{r}t) = 0 \tag{1}$$

and knowledge of the initial state it will also have the same density as the true system. The Kohn-Sham system will now have an external pair of potentials $(v_s(\mathbf{r}t), \mathbf{A}_s(\mathbf{r}t))$ which are functionals of the current-density. Once an approximation of these potentials as a functional of the current-density is given the Kohn-Sham equations can be solved self-consistently, and the current-density can be calculated. Such an ap-

proach has several advantages. First of all, the theory can be applied to transverse perturbing fields described by an vector potential $\mathbf{A}(\mathbf{r}t)$ and therefore presents an extension of the TDDFT formulation which only dealt with longitudinal fields. Secondly, the theory can be applied to calculate response properties of infinite systems, in which the changes of the boundary charges are measured by the current and from which the polarizability can be directly calculated [12, 13, 14, 15] Thirdly, it has been shown by Vignale and Kohn [9] that TDCDFT allows for functionals that are nonlocal in time, but local in space. This is related to the fact that a gradient expansion exists for the current-current response function, whereas it does not for the density-density response function. An approximate current functional derived by Vignale and Kohn was shown to be very successful for the calculation of plasmon linewidths in quantum wells [16, 17] and the polarizabilities of polymers [18].

The aim of this paper is to provide a systematic method to calculate current functionals to be used in the time-dependent Kohn-Sham equations. We will also fill in some details on the Keldysh formalism that were left out in earlier work [19, 20, 5]. The paper is divided as follows. We first introduce the Keldysh action functional and show how this functional can be used to generate the current-current response functions. We then show how the Keldysh functional can be used to derive the Kohn-Sham and corresponding linear response equations. By expanding this functional in terms of the two-particle interaction we obtain a diagrammatic perturbation expansion in terms of Keldysh Green functions for the effective vector potential and its functional derivative, to be used in the Kohn-Sham equations or their linearized form. The procedure is illustrated for the electron gas. We finally present an outlook and conclusions.

2. Keldysh action

We consider a system of interacting electrons in a time-dependent external field. A typical example would for instance be an atom or molecule in a laser field. The Hamiltonian which describes this system is given by

$$\hat{H}(t) = \hat{T} + \hat{W} + \hat{U}(t) \tag{2}$$

where the constituent term are given as follows. The kinetic energy $\hat{T}$ and the two-particle interaction $\hat{W}$ are given by

$$\hat{T} = -\sum_{\sigma} \frac{1}{2} \int d^3 r \hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) \nabla^2 \hat{\psi}_{\sigma}(\mathbf{r}) \tag{3}$$

4

and

$$\hat{W} = \sum_{\sigma,\sigma'} \frac{1}{2} \int d^3 r d^3 r' \hat{\psi}_\sigma^\dagger(\mathbf{r}) \hat{\psi}_{\sigma'}^\dagger(\mathbf{r}') w(\mathbf{r},\mathbf{r}') \hat{\psi}_{\sigma'}(\mathbf{r}') \hat{\psi}_\sigma(\mathbf{r}) \qquad (4)$$

where w represent the usual Coulomb repulsion between the electrons. The term $\hat{U}(t)$ represents the time-dependent external field

$$\hat{U}(t) = \int d^3 r (\hat{n}(\mathbf{r}) v_0(\mathbf{r}) + \hat{\mathbf{j}}_p(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}t) + \frac{1}{2} \hat{n}(\mathbf{r}) \mathbf{A}^2(\mathbf{r}t)) \qquad (5)$$

where

$$\hat{n}(\mathbf{r}) = \sum_\sigma \hat{\psi}_\sigma^\dagger(\mathbf{r}) \hat{\psi}_\sigma(\mathbf{r}) \qquad (6)$$

$$\hat{\mathbf{j}}_p(\mathbf{r}) = \sum_\sigma \frac{1}{2i} (\hat{\psi}_\sigma^\dagger(\mathbf{r}) \nabla \hat{\psi}_\sigma(\mathbf{r}) - [\nabla \hat{\psi}_\sigma^\dagger(\mathbf{r})] \hat{\psi}_\sigma(\mathbf{r})) \qquad (7)$$

are the density and the paramagnetic current operator. In the following, where we will mostly consider the linear response case, $v_0(\mathbf{r})$ will be the external potential in the ground state and all time-dependence will be incorporated in $\mathbf{A}(\mathbf{r}t)$. We have now completely defined our physical system. This leaves us with the task of calculating the physical quantities of interest. We are mostly interested in calculating the response of the system due to external perturbations. We therefore want to introduce a generating functional for the response functions. This approach is analogous to the situation in statistical mechanics where all thermodynamic quantities can be calculated as derivatives with respect to parameters in the partition function. The basic idea is therefore to start with a generating functional resembling the partition function of statistical mechanics. This functional should be constructed in such a way that its first order derivative with respect to the external field yields the physical current density and its higher order derivatives yield the desired higher order response functions.

In earlier work we showed how such a functional can be constructed. This functional was defined on using the so-called Keldysh time contour. This time contour technique was introduced by Schwinger [21] and used by Keldysh [22] to obtain a diagrammatic perturbation expansion for nonequilibrium systems. In this technique the real time $t(\tau)$ is parametrized by an underlying pseudotime parameter τ [23, 24, 25, 26]. The parametrization is such that, if the pseudotime time runs from a certain initial time τ_i to a final time τ_f, then the real time runs from t_0 to t_1 and back to t_0. This trick has the great virtue that, when evolution operators are used that are τ-ordered rather than t-ordered, then in perturbation theory Wick's theorem can be used, without invoking

any adiabatic switch-off of the two-particle interactions as is done in the Gellman-Low theorem [27, 28]. It is exactly this feature that makes the theory applicable to nonstationary systems. Within time-dependent density functional theory it has already been shown that the formalism can also be used to resolve a paradox [20, 5] involving the symmetry and causality properties of response functions. Here we make an extension of that work to the case of current functionals.

We define a functional of the external field $\mathbf{A}$ by

$$\tilde{F}[\mathbf{A}] = i \ln\langle\Psi_0|V(\tau_f, \tau_i)|\Psi_0\rangle \tag{8}$$

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

$$V(\tau_2, \tau_1) = T_C \exp\left[-i \int_{\tau_1}^{\tau_2} d\tau\, t'(\tau)\hat{H}(\tau)\right] \tag{9}$$

and where T_C denotes ordering in τ and the state Ψ_0 is the initial state. Here we extend the definition of the Hamiltonian and allow $\mathbf{A}(\mathbf{r}\tau)$ to take different values of the forward and backward parts of the contour. It is clear from this equation that if the external field $\mathbf{A}$ is equal on the forward and backward parts of the contour, i.e. depending on τ only through $t(\tau)$, then this evolution will become unity and $\tilde{F}$ will become zero. Vector potentials of this type will in the following be denoted as physical vector potentials. The functional derivatives with respect to $\mathbf{A}$ taken at a physical vector potential will, however, be nonzero in general. When taking functional derivatives we use the basic equation

$$\frac{\delta}{\delta\mathbf{A}(\mathbf{r}\tau)}V(\tau_2, \tau_1) = -iV(\tau_2, \tau)(\hat{\mathbf{j}}_p(\mathbf{r}) + \hat{n}(\mathbf{r})\mathbf{A}(\mathbf{r}\tau))V(\tau, \tau_1) \tag{10}$$

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

$$\begin{aligned}
\frac{\delta\tilde{F}}{\delta\mathbf{A}(\mathbf{r}\tau)} &= \frac{\langle\Psi_0|V(\tau_f, \tau)(\hat{\mathbf{j}}_p(\mathbf{r}) + \hat{n}(\mathbf{r})\mathbf{A}(\mathbf{r}\tau))V(\tau, \tau_i)|\Psi_0\rangle}{\langle\Psi_0|V(\tau_f, \tau_i)|\Psi_0\rangle} \\
&= \langle\hat{\mathbf{j}}_{p,H}(\mathbf{r}\tau)\rangle + \langle\hat{n}_H(\mathbf{r}\tau)\rangle\mathbf{A}(\mathbf{r}\tau) = \mathbf{j}(\mathbf{r}\tau) \tag{11}
\end{aligned}$$

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

$$\langle\hat{O}_H(\tau)\rangle = \frac{\langle\Psi_0|T_C[V(\tau_f, \tau_i)\hat{O}_H(\tau)]|\Psi_0\rangle}{\langle\Psi_0|V(\tau_f, \tau_i)|\Psi_0\rangle} \tag{12}$$

If we evaluate the derivative $\delta\tilde{F}/\delta\mathbf{A}$ at a physical potential $\mathbf{A}(\mathbf{r}t(\tau))$ we obtain

$$\frac{\delta\tilde{F}}{\delta\mathbf{A}(\mathbf{r}\tau)}\Big|_{\mathbf{A}=\mathbf{A}(\mathbf{r}t_1)} = \langle\Psi_0|U(t_0, t_1)(\hat{\mathbf{j}}_p(\mathbf{r}) + \hat{n}(\mathbf{r})\mathbf{A}(\mathbf{r}t_1))U(t_1, t_0)|\Psi_0\rangle$$

$$= \mathbf{j}(\mathbf{r}t_1) \tag{13}$$

where the evolution operator in real time is defined as usual by

$$U(t_1, t_0) = T \exp\left[-i \int_{t_0}^{t_1} dt \hat{H}(t)\right] \tag{14}$$

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. At this point it will be convenient to introduce a shortened notation. We will write $\bar{k} = \mathbf{r}_k\tau_k$ and $k = \mathbf{r}_k t_k$ and will also drop the subindex H from the operators. The current-current response function $\chi_{\mu\nu}$ is then given by

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

$$= \delta_{\mu\nu} n(\bar{1})\delta_C(1-2) - i\langle T_C[\Delta\hat{\mathbf{j}}_p(\bar{1})\Delta\hat{\mathbf{j}}_p(\bar{2})]\rangle \tag{15}$$

where $\delta_C(t_1 - t_2) = \delta(\tau_1 - \tau_2)/t'(\tau_1)$ is the contour delta function 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.(11). 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. In order to see this we calculate the current response $\delta\mathbf{j}(\mathbf{r}t)$ due to a potential variation $\delta\mathbf{A}(\mathbf{r}t)$. The density response function $\chi_{\mu\nu}$ evaluated at a physical current density $\mathbf{j}(\mathbf{r}t)$ is given by

$$\begin{aligned}
\chi_{\mu\nu}(\bar{1}, \bar{2}) &= \delta_{\mu\nu} n(1)\delta_C(1-2) \\
&\quad - i\theta(\tau_1 - \tau_2)\langle\Delta\hat{\mathbf{j}}_{p,\mu}(1)\Delta\hat{\mathbf{j}}_{p,\nu}(2)\rangle \\
&\quad - i\theta(\tau_2 - \tau_1)\langle\Delta\hat{\mathbf{j}}_{p,\nu}(2)\Delta\hat{\mathbf{j}}_{p,\mu}(1)\rangle
\end{aligned} \tag{16}$$

Hence we have

$$\begin{aligned}
\delta j_\mu(\mathbf{r}_1 t_1) &= \sum_\nu \int_{\tau_i}^{\tau_f} d\tau_2 t'(\tau_2) d^3 r_2 \chi_{\mu\nu}(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2)\delta A_\nu(\mathbf{r}_2 t_2) \\
&= n_0(\mathbf{r}_1)\delta A_\nu(\mathbf{r}, t_1) \\
&\quad - \sum_\nu i \int_{\tau_i}^{\tau_1} d\tau_2 t'(\tau_2) d^3 r_2 \langle\Delta\hat{\mathbf{j}}_{p,\mu}(\mathbf{r}_1 t_1)\Delta\hat{\mathbf{j}}_{p,\nu}(\mathbf{r}_2 t_2)\rangle\delta A_\nu(\mathbf{r}_2 t_2) \\
&\quad - \sum_\nu i \int_{\tau_1}^{\tau_f} d\tau_2 t'(\tau_2) d^3 r_2 \langle\Delta\hat{\mathbf{j}}_{p,\nu}(\mathbf{r}_2 t_2)\Delta\hat{\mathbf{j}}_{p,\mu}(\mathbf{r}_1 t_1)\rangle\delta A_\nu(\mathbf{r}_2 t_2)
\end{aligned}$$

$$= \sum_\nu \int_{t_0}^{+\infty} dt_2 d^3 r_2 \chi_{R,\mu\nu}(\mathbf{r}_1 t_1, \mathbf{r}_2 t_2) \delta A_\nu(\mathbf{r}_2 t_2) \tag{17}$$

where

$$
\begin{aligned}
\chi_{R,\mu\nu}(1,2) &= n_0(\mathbf{r}_1)\delta_{\mu\nu}\delta(t_1 - t_2)\delta(\mathbf{r}_1 - \mathbf{r}_2) \\
&- i\theta(t_1 - t_2)\langle\Psi_0|[\hat{\mathbf{j}}_{p,\mu}(1), \hat{\mathbf{j}}_{p,\nu}(2)]|\Psi_0\rangle
\end{aligned}
\tag{18}
$$

In the last step we used that the expectation value of the commutator of the current fluctuation operators is equal to the expectation value of the commutator of the current operators themselves. The function $\chi_{R,\mu\nu}$ is the usual retarded response function 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 (see Appendix D for an explicit example).

3.   Kohn-Sham equations and linear response

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

$$F[\mathbf{j}] = -\tilde{F}[\mathbf{A}] + \int_C dt d^3 r \mathbf{j}(\mathbf{r}\tau) \cdot \mathbf{A}(\mathbf{r}\tau) \tag{19}$$

so that

$$\delta F/\delta\mathbf{j}(\mathbf{r}\tau) = \mathbf{A}(\mathbf{r}\tau) \tag{20}$$

For convenience we introduced the short notation $\int_C dt$ for $\int d\tau t'(\tau)$. The Legendre transformation assumes that there is a one-to-one relation between $\mathbf{j}(\mathbf{r}\tau)$ and $\mathbf{A}(\mathbf{r}\tau)$ such that Eq. (11) is invertible (up to gauge). One can prove, when considering a system initially in its ground state, that the Keldysh current-current reponse function is invertible for switch-on processes. This can be done by a direct generalization of the proof given for the density-density response function in ref.[5]. We now define an action functional for a noninteracting system with the Hamiltonian

$$\hat{H}_s(\tau) = \hat{T} + \hat{U}_s(\tau) \tag{21}$$

where U_s has a form analogous to Eq.(5), and the action

$$\tilde{F}_s[\mathbf{A}_s] = i \ln\langle\Phi_0|V_s(\tau_f, \tau_i)|\Phi_0\rangle \tag{22}$$

The evolution operator $V_s(\tau_f, \tau_i)$ is defined similarly as in Eq.(9) 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 a similar Legendre transform and define

$$F_s[\mathbf{j}] = -\tilde{F}_s[\mathbf{A}_s] + \int_C dt d^3r \mathbf{j}(\mathbf{r}\tau) \cdot \mathbf{A}_s(\mathbf{r}\tau) \tag{23}$$

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

$$F[\mathbf{j}] = F_s[\mathbf{j}] - F_{xc}[\mathbf{j}] - \frac{1}{2} \int_C dt d^3r_1 d^3r_2 \frac{n(\mathbf{r}_1\tau)n(\mathbf{r}_2\tau)}{|\mathbf{r}_1 - \mathbf{r}_2|} \tag{24}$$

where the Keldysh density $n(\mathbf{r}\tau)$ is a functional of the initial state and the current $\mathbf{j}$ through the continuity equation

$$\partial_t n(\mathbf{r}\tau) + \nabla \cdot \mathbf{j}(\mathbf{r}\tau) = 0 \tag{25}$$

(note that $\partial_t = 1/t'(\tau)\partial_\tau$) We implicitly assume that the functionals F and F_s are defined on the same domain, i.e., that there exists a noninteracting system described by the Hamiltonian $\hat{H}_s$ with the same current density as the interacting system described by the Hamiltonian $\hat{H}$. A necessary requirement in order for this to be true is that the initial states Ψ_0 and Φ_0 must yield the same current density. For most applications, Ψ_0 will be the ground state of the system before the time-dependent field is switched on and Φ_0 will be the corresponding Kohn-Sham determinant of stationary density-functional theory. Functional differentiation of Eq. (24) with respect to $\mathbf{j}(\mathbf{r}\tau)$ yields

$$\mathbf{A}(\mathbf{r}\tau) = \mathbf{A}_s(\mathbf{r}\tau) - \mathbf{A}_{xc}(\mathbf{r}\tau) - \mathbf{A}_H(\mathbf{r}\tau) \tag{26}$$

where

$$\partial_t \mathbf{A}_H(\mathbf{r}\tau) = -\nabla \int d^3r' \frac{n(\mathbf{r}'\tau)}{|\mathbf{r} - \mathbf{r}'|} \tag{27}$$

is the Hartree part of the vector potential and

$$\mathbf{A}_{xc}(\mathbf{r}\tau) = \delta F_{xc}/\delta \mathbf{j}(\mathbf{r}\tau) \tag{28}$$

is the exchange-correlation 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. If we take the functional derivatives at the physical time-dependent current density $\mathbf{j}(\mathbf{r}t)$ corresponding to the vector potential $\mathbf{A}(\mathbf{r}\tau) = \mathbf{A}(\mathbf{r}t(\tau))$ of the interacting system, we can transform to physical time and the Kohn-Sham system is then given by the equations

$$(-\frac{1}{2}(\nabla + i(\mathbf{A} + \mathbf{A}_H + \mathbf{A}_{xc}))^2 + v_0(\mathbf{r}))\phi_i(\mathbf{r}t) = i\partial_t\phi_i(\mathbf{r}t)$$

$$\mathbf{A}_{xc}(\mathbf{r}t) = \frac{\delta F_{xc}}{\delta \mathbf{j}(\mathbf{r}\tau)}\Big|_{\mathbf{j}=\mathbf{j}(\mathbf{r}t)} \tag{29}$$

where the density $\mathbf{j}(\mathbf{r}t)$ can be calculated from the orbitals according to

$$\mathbf{j}(\mathbf{r}t) = n(\mathbf{r}t)\mathbf{A}_s(\mathbf{r}t) + \frac{1}{2i}\sum_i^N (\phi_i^*(\mathbf{r}t)\nabla\phi_i(\mathbf{r}t) - (\nabla\phi_i^*(\mathbf{r}t))\phi_i(\mathbf{r}t)) \tag{30}$$

Let us now see how the current-current response function can be obtained from this formalism. With the chain rule for differentiation we can write

$$
\begin{aligned}
\frac{\delta j_\mu(\bar{1})}{\delta A_\nu(\bar{2})} &= \sum_\lambda \int_C d3 \frac{\delta j_\mu(\bar{1})}{\delta A_{s,\lambda}(\bar{3})} \frac{\delta A_{s,\lambda}(\bar{3})}{\delta A_\nu(\bar{2})} = \frac{\delta j_\mu(\bar{1})}{\delta A_{s,\nu}(\bar{2})} \\
&+ \sum_{\lambda\kappa} \int_C d3d4 \frac{\delta j_\mu(\bar{1})}{\delta A_{s,\lambda}(\bar{3})} \frac{\delta(A_{H,\lambda}(\bar{3}) + A_{xc,\lambda}(\bar{3}))}{\delta j_\kappa(\bar{4})} \frac{\delta j_\kappa(\bar{4})}{\delta A_\nu(\bar{2})} \tag{31}
\end{aligned}
$$

Hence we obtain

$$
\begin{aligned}
\chi_{\mu\nu}(\bar{1},\bar{2}) &= \chi_{s,\mu\nu}(\bar{1},\bar{2}) \\
&+ \sum_{\lambda\kappa} \int_C d3d4 \chi_{s,\mu\lambda}(\bar{1},\bar{3}) f_{Hxc,\lambda\kappa}(\bar{3},\bar{4}) \chi_{\kappa\nu}(\bar{4},\bar{2}) \tag{32}
\end{aligned}
$$

where we defined

$$f_{H,\mu\nu}(\bar{1},\bar{2}) = \frac{\delta A_{H,\mu}(\bar{1})}{\delta j_\nu(\bar{2})} \tag{33}$$

$$f_{xc,\mu\nu}(\bar{1},\bar{2}) = \frac{\delta A_{xc,\mu}(\bar{1})}{\delta j_\nu(\bar{2})} \tag{34}$$

$$f_{Hxc,\mu\nu}(\bar{1},\bar{2}) = f_{H,\mu\nu}(\bar{1},\bar{2}) + f_{xc,\mu\nu}(\bar{1},\bar{2}) \tag{35}$$

We have therefore found a relation between the full and the Kohn-Sham current-current response function. A similar equation has been derived before within the context of pure density functional theory [29]. Equation (32) can now be transformed to physical time (for details see Appendix C) giving

$$
\begin{aligned}
\chi_{R,\mu\nu}(1,2) &= \chi_{R,s,\mu\nu}(1,2) \\
&+ \sum_{\lambda\kappa} \int d3d4 \chi_{R,s,\mu\lambda}(1,3) f_{R,Hxc,\lambda\kappa}(3,4) \chi_{R,\kappa\nu}(4,2) \tag{36}
\end{aligned}
$$

where the Keldysh response functions are now replaced with the retarded causal ones. If we consider the response of a system initially in its

ground state then the response function depends on the difference of the time coordinates. In that case it is convenient to Fourier transform to frequency space and we can write

$$\chi_{R,\mu\nu}(\mathbf{r}_1,\mathbf{r}_2,\omega) = \chi_{R,s,\mu\nu}(\mathbf{r}_1,\mathbf{r}_2,\omega) + \sum_{\lambda\kappa}\int d^3r_3 d^3r_4 \chi_{R,s,\mu\lambda}(\mathbf{r}_1,\mathbf{r}_3,\omega)$$
$$\times (f_{R,xc,\lambda\kappa}(\mathbf{r}_3,\mathbf{r}_4,\omega) - \frac{1}{\omega^2}\partial_\lambda^3\frac{1}{|\mathbf{r}_3-\mathbf{r}_4|}\partial_\kappa^4)\chi_{R,\kappa\nu}(\mathbf{r}_4,\mathbf{r}_2,\omega)(37)$$

Here we used the continuity equation in

$$\partial_t^2\delta A_H(\mathbf{r}\tau) = \nabla\int d^3r'\frac{\nabla'\cdot\delta\mathbf{j}(\mathbf{r}'\tau)}{|\mathbf{r}-\mathbf{r}'|} \tag{38}$$

to calculate $f_{R,H}(\mathbf{r}_3,\mathbf{r}_4,\omega)$ explicitly in frequency space. Eq.(37) is the basic equation of time-dependent current response theory. It can, in fact, be used to defined $f_{xc,\mu\nu}$, as has been done by Vignale and Kohn [9, 10]. This equation, together with the time-dependent Kohn-Sham equations, are the main results of this section. The next step will be to obtain approximations for $\mathbf{A}_{xc}$ and for the xc-kernel $f_{R,xc,\mu\nu}$. How to obtain such a approximations will be the subject of the following sections.

4. TDOPM equations

We will now use the new formalism to derive the current extension of the time-dependent optimized potential method (TDOPM) [30].
The exchange-correlation part F_{xc} of the action functional can be expanded in terms of Keldysh Green functions [19] where the perturbing Hamiltonian is given by $\hat{H} - \hat{H}_s$. The Keldysh Green function is defined as

$$
\begin{aligned}
G_{\sigma\sigma'}(\bar{1},\bar{2}) &= -i\langle\Psi_0|T_C\hat{\psi}_{\sigma,H}(\bar{1})\hat{\psi}_{\sigma'}^\dagger(\bar{2})|\Psi_0\rangle \\
&= \theta(\tau_1-\tau_2)G_{\sigma,\sigma'}^>(\bar{1},\bar{2}) + \theta(\tau_2-\tau_1)G_{\sigma,\sigma'}^<(\bar{1},\bar{2}) \quad (39)
\end{aligned}
$$

where we defined

$$G_{\sigma\sigma'}^>(\bar{1},\bar{2}) = -i\langle\Psi_0|\hat{\psi}_{\sigma,H}(\bar{1})\hat{\psi}_{\sigma',H}^\dagger(\bar{2})|\Psi_0\rangle \tag{40}$$
$$G_{\sigma\sigma'}^<(\bar{1},\bar{2}) = i\langle\Psi_0|\hat{\psi}_{\sigma',H}^\dagger(\bar{2})\hat{\psi}_{\sigma,H}(\bar{1})|\Psi_0\rangle \tag{41}$$

The expansion of the logarithm of the evolution operator yields the set of closed connected diagrams. Perturbation theory also requires an adiabatic switching-on of $\hat{H} - \hat{H}_s$ in the physical time interval $(-\infty, t_0)$ in order to connect the states Ψ_0 and Φ_0. This is however readily achieved

by extending the Keldysh contour to $-\infty$ [19]. The expansion of the action is then given in terms of the Kohn-Sham Green function

$$G_s(\bar{1}, \bar{2}) = \theta(\tau_1 - \tau_2)G_s^>(\bar{1}, \bar{2}) + \theta(\tau_2 - \tau_1)G_s^<(\bar{1}, \bar{2}) \qquad (42)$$

where

$$G_s^<(\bar{1}, \bar{2}) = i\sum_{i=1}^{N/2} \phi_i^*(\bar{1})\phi_i(\bar{2}) \qquad (43)$$

$$G_s^>(\bar{1}, \bar{2}) = -i\sum_{i>N/2} \phi_i^*(\bar{1})\phi_i(\bar{2}) \qquad (44)$$

where we consider the spin restricted case. If we restrict ourselves to the first order terms we find that the Hartree term and the term with $\mathbf{A} - \mathbf{A}_s$ cancel and obtain the exchange-only expression in terms of the Kohn-Sham Green function

$$
\begin{aligned}
F_x[\mathbf{j}] &= \int_C d1 \int_C d2 G_s(\bar{1}, \bar{2})v(1,2)G_s(\bar{2}, \bar{1}) \\
&= \int_C dt d^3 r_1 d^3 r_2 \frac{G_s^<(\mathbf{r}_1\tau, \mathbf{r}_2\tau)G_s^<(\mathbf{r}_2\tau, \mathbf{r}_1\tau)}{|\mathbf{r}_1 - \mathbf{r}_2|} \\
&= -\sum_{ij}^{N/2} \int_C dt d^3 r_1 d^3 r_2 \frac{\phi_i^*(\mathbf{r}_1\tau)\phi_i(\mathbf{r}_2\tau)\phi_j(\mathbf{r}_1\tau)\phi_j^*(\mathbf{r}_2\tau)}{|\mathbf{r}_1 - \mathbf{r}_2|} \qquad (45)
\end{aligned}
$$

where $v(1,2) = \delta_C(t_1 - t_2)/|\mathbf{r}_1 - \mathbf{r}_2|$ and we use spinintegrated Green functions. One sees that this functional is an implicit functional of $n(\mathbf{r}\tau)$ but an explicit functional of the orbitals. Going to higher order in $\hat{H} - \hat{H}_s$, the Keldysh perturbation expansion, in a similar way, leads to orbital dependent expressions for the correlation part F_c of the action (although there will also be implicit orbital dependence since higher order diagrams also contain terms that involve $\mathbf{A}_{xc}$). In the general case one may obtain $\mathbf{A}_{xc}$ from

$$A_{xc,\mu}(\bar{2}) = \int_C dt d^3 r_1 \frac{\delta F_{xc}}{\delta A_{s,\nu}(\bar{1})} \frac{\delta A_{s,\nu}(\bar{1})}{\delta j_\mu(\bar{2})} \qquad (46)$$

Matrix multiplication by $\chi_{s,\mu\nu}$ and using the chain rule for differentiation yields (from now on we drop subindex s from the Green functions)

$$
\begin{aligned}
&\sum_\nu \int_C dt_2 d^3 r_2 \chi_{s,\mu\nu}(\bar{1}, \bar{2})A_{xc,\nu}(\bar{2}) \\
&= \int_C d2 \int_C d3 \frac{\delta F_{xc}}{\delta G(\bar{2}, \bar{3})} \frac{\delta G(\bar{2}, \bar{3})}{\delta A_{s,\mu}(\bar{1})} \\
&= \int_C d2 \int_C d3 \frac{\delta F_{xc}}{\delta G(\bar{2}, \bar{3})} G(\bar{2}, \bar{1})\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G(\bar{1}, \bar{3}) \qquad (47)
\end{aligned}
$$

where we defined the one-particle current operator $\mathbf{j_{op,\nu}} = 1/2i(\overrightarrow{\partial}_\mu - \overleftarrow{\partial}_\mu)$, where the second derivarive $\overleftarrow{\partial}_\mu)$ acts on the function to the left of the operator. We further used the relation (see Appendix B)

$$\frac{\delta G(\bar{1}, \bar{2})}{\delta A_\nu(\bar{3})} = G(\bar{1}, \bar{3})\mathbf{j_{op,\nu}}(\mathbf{r}_3)G(\bar{3}, \bar{2}) \qquad (48)$$

Let us now restrict ourselves to the exchange-only case. Then we obtain the integral equation

$$\sum_\nu \int_C dt_2 d^3 r_2 \chi_{s,\mu\nu}(\bar{1}, \bar{2}) A_{x,\nu}(\bar{2})$$

$$= 2 \int_C d2 \int_C d3 v(3, 2) G(\bar{3}, \bar{2}) G(\bar{2}, \bar{1}) \mathbf{j_{op,\mu}}(\mathbf{r}_1) G(\bar{1}, \bar{3}) \qquad (49)$$

An explicit expression for the Kohn-Sham response function in terms of the orbitals is given in Appendix A. If we transform to physical time we obtain the following equation for the x-only vector potential

$$\sum_\nu \int dt_2 d^3 r_2 \chi_{R,s,\mu\nu}(1, 2) A_{x,\nu}(2) = \Pi_\mu(1) \qquad (50)$$

where

$$\begin{aligned}
\Pi_\mu(1) &= 2 \int d^3 r_2 \int d^3 r_2 \int_{-\infty}^{t_1} dt \frac{G^<(\mathbf{r}_2 t, \mathbf{r}_3 t)}{|\mathbf{r}_2 - \mathbf{r}_3|} \\
&\times \left[G^<(\mathbf{r}_2 t, \mathbf{r}_1 t)\mathbf{j_{op,\mu}}(\mathbf{r}_1)G^>(\mathbf{r}_1 t, \mathbf{r}_3 t) - (> \leftrightarrow <) \right] \qquad (51)
\end{aligned}$$

where the last term in brackets denotes that this term is obtain by interchanging $>$ and $<$ in the previous term. Note that the time integration in this expression runs to $-\infty$. This is due to the adiabatic connection between Kohn-Sham and true initial state before time t_0. The time-integration in the interval $[-\infty, t_0]$ can be done explicitly since the time-dependence of the Kohn-Sham orbitals is known in this time interval. For a further discussion of similar terms see ref.[31, 32]

Eqn.(50) together with (51) is the main result of this section. They are the current-density generalization of the well-known equations of the time-dependent optimized potential method (TDOPM). In case the external fields are longitudinal these equations can (using gauge transformation and the continuity equation) be shown to be equivalent to the TDOPM equations.

5.   Integral equation for the xc-kernel

We will now study the integral equation for the xc-kernel $f_{xc,\mu\nu}$. Being a two-point function it gives much more information than just $\mathbf{A}_{xc}$. The

determining equation for $f_{xc,\mu\nu}$ is consequently more complicated. It can be obtained by differentiation of Eq.(47) with respect to $A_{s,\kappa}$. This yields

$$\sum_\nu \int_C dt_2 d^3 r_2 \frac{\delta\chi_{s,\mu\nu}(\bar{1},\bar{2})}{\delta A_{s,\kappa}(\bar{3})} A_{xc,\nu}(\bar{2})$$

$$+ \quad \sum_\nu \int_C dt_2 d^3 r_2 \chi_{s,\mu\nu}(\bar{1},\bar{2}) \frac{\delta A_{xc,\nu}(\bar{2})}{\delta A_{s,\kappa}(\bar{3})} = Q_{\mu\kappa}(\bar{1},\bar{3}) \qquad (52)$$

where we defined

$$Q_{\mu\kappa}(\bar{1},\bar{3}) = \frac{\delta^2 F_{xc}}{\delta A_{s,\kappa}(\bar{3})\delta A_{s,\mu}(\bar{1})} \qquad (53)$$

With the chain rule for differentiation this can be written as

$$\sum_\nu \int_C dt_2 d^3 r_2 \chi^{(2)}_{s,\mu\nu\kappa}(\bar{1},\bar{2},\bar{3}) A_{xc,\nu}(\bar{2})$$

$$+ \quad \sum_{\nu\lambda} \int_C dt_2 d^3 r_2 \int_C dt_4 d^3 r_4 \chi_{s,\mu\nu}(\bar{1},\bar{2}) f_{xc,\nu\lambda}(\bar{2},\bar{4}) \chi_{s,\lambda\kappa}(\bar{4},\bar{3})$$

$$= \quad Q_{\mu\kappa}(\bar{1},\bar{3}) \qquad (54)$$

where we defined the second order Keldysh response function of the Kohn-Sham system

$$\chi^{(2)}_{s,\mu\nu\kappa}(\bar{1},\bar{2},\bar{3}) = (-i)^2 \langle T_C \Delta j_{p,H,\mu}(\bar{1}) \Delta j_{p,H,\nu}(\bar{2}) \Delta j_{p,H,\kappa}(\bar{3}) \rangle \qquad (55)$$

Eq.(54) is an integral equation for the xc-kernel. In these equations the first and second order Kohn-Sham response functions $\chi_{s,\mu\nu}$ and $\chi^{(2)}_{s,\mu\nu\kappa}$ are explicitly known in terms of the Kohn-Sham orbitals (see Appendix A for the explicit form of $\chi_{s,\mu\nu}$). The equation can now be transformed to physical time. The key steps to do this are explained in Appendix C and D. The result is

$$\sum_\nu \int dt_2 d^3 r_2 \chi^{(2)}_{R,s,\mu\nu\kappa}(1,2,3) A_{xc,\nu}(2)$$

$$+ \quad \sum_{\nu\lambda} \int dt_2 d^3 r_2 \int dt_4 d^3 r_4 \chi_{R,s,\mu\nu}(1,2) f_{R,xc,\nu\lambda}(2,4) \chi_{R,s,\lambda\kappa}(4,3)$$

$$= \quad Q_{R,\mu\kappa}(1,3) \qquad (56)$$

where

$$\chi^{(2)}_{s,\mu\nu\kappa}(1,2,3) = (-i)^2 \theta(t_1 - t_2)\theta(t_2 - t_3)$$

$$\times \quad \langle \Psi_0 | [[j_{p,H,\mu}(1), j_{p,H,\nu}(2)], j_{p,H,\kappa}(3)] | \Psi_0 \rangle \qquad (57)$$
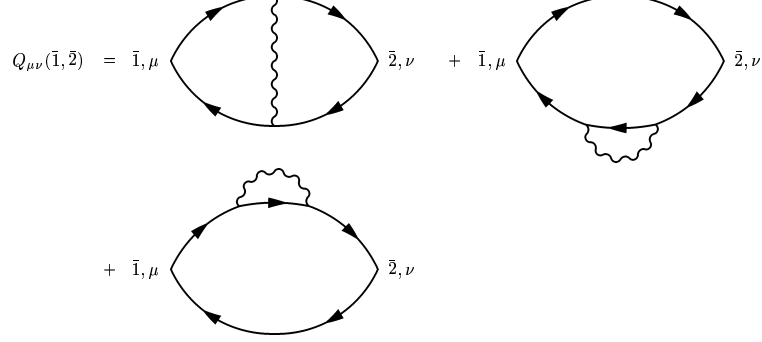
Figure 1. The three first order polarization diagrams. The labels μ and ν denote insertion of the operator $j_{\mu,\mathbf{op}}$ and $j_{\nu,\mathbf{op}}$. The first diagram contains a vertex while the other two contain self-energy insertions.

is the retarded second order current response function. Eq.(56) can be solved selfconsistently once the $Q_{\mu\kappa}$ is given. In the following we will make the exchange-only approximation for this term, which amounts to an expansion to first order in the two-particle interaction. In that case the inhomogeneity $Q_{\mu\kappa}$ is given as

$$
\begin{aligned}
Q_{\mu\kappa}&(\bar{1},\bar{3}) \\
&= \frac{\delta}{\delta A_{s,\kappa}(\bar{3})}\left(2\int_C d2\int_C d4 v(2,4)G(\bar{2},\bar{4})G(\bar{4},\bar{1})\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G(\bar{1},\bar{2})\right) \\
&= 2\int_C d2\int_C d4 G(\bar{2},\bar{3})\mathbf{j}_{\mathbf{op},\kappa}(\mathbf{r}_3)G(\bar{3},\bar{4})v(2,4)G(\bar{4},\bar{1})\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G(\bar{1},\bar{2}) \\
&+ 2\int_C d2\int_C d4 G(\bar{2},\bar{4})v(2,4)G(\bar{4},\bar{1})\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G(\bar{1},\bar{3})\mathbf{j}_{\mathbf{op},\kappa}(\mathbf{r}_3)G(\bar{3},\bar{2}) \\
&+ 2\int_C d2\int_C d4 G(\bar{2},\bar{4})v(2,4)G(\bar{4},\bar{3})\mathbf{j}_{\mathbf{op},\kappa}(\mathbf{r}_3)G(\bar{3},\bar{1})\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G(\bar{1},\bar{2})
\end{aligned}
$$

Those three terms represent the three first order diagrams of the polarization bubble. They are presented graphically as Feynman diagrams in Figure 1. Each black line denotes a Green function and the wiggly line denotes the two-particle interaction. Let us now transform to physical time and work out the terms in $Q_{R,\mu\kappa}$ which consists of the three Feynman diagrams transformed to physical time. We write these three terms as

$$
Q_{R,\mu\kappa} = D_{1,\mu\kappa} + D_{2,\mu\kappa} + D_{3,\mu\kappa} \tag{58}
$$

The first diagram contains a vertex and yields in real time the expression

$$
D_{1,\mu\kappa}(\mathbf{r}_1 t_1, \mathbf{r}_3 t_3) =
$$

$$-2 \int_{t_0}^{\infty} dt \int d^3r_2 d^3r_4 \theta(t_1 - t)\theta(t - t_3)v(\mathbf{r}_2, \mathbf{r}_4)$$
$$\times \quad [G^{>}(\mathbf{r}_4 t, \mathbf{r}_1 t_1)\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G^{<}(\mathbf{r}_1 t_1, \mathbf{r}_2 t) - (> \leftrightarrow <)]$$
$$\times \quad [G^{>}(\mathbf{r}_2 t, \mathbf{r}_3 t_3)\mathbf{j}_{\mathbf{op},\kappa}(\mathbf{r}_3)G^{<}(\mathbf{r}_3 t_3, \mathbf{r}_4 t) - (> \leftrightarrow <)] \qquad (59)$$

The remaining two diagrams contain self-energy insertions and are complex conjugates of each other, i.e. $D_{3,\mu\kappa} = D_{2,\mu\kappa}^{*}$, and therefore knowledge of one of them suffices to know the other. For $D_{3,\mu\kappa}$ we find the following explicit expression

$$D_{3,\mu\kappa} = A_{3,\mu\kappa} + B_{3,\mu\kappa} + C_{3,\mu\kappa} \qquad (60)$$

where the three terms A, B and C are given by

$$A_{3,\mu\kappa}(\mathbf{r}_1 t_1, \mathbf{r}_3 t_3) =$$
$$2 \int_{t_0}^{\infty} dt \int d^3r_2 d^3r_4 \theta(t_1 - t)\theta(t - t_3)v(\mathbf{r}_2, \mathbf{r}_4)G^{<}(\mathbf{r}_2 t, \mathbf{r}_4 t)$$
$$\times \quad [G^{>}(\mathbf{r}_4 t, \mathbf{r}_3 t_3)\mathbf{j}_{\mathbf{op},\kappa}(\mathbf{r}_3)G^{<}(\mathbf{r}_3 t_3, \mathbf{r}_1 t_1)\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G^{>}(\mathbf{r}_1 t_1, \mathbf{r}_2 t)$$
$$+ \quad (> \leftrightarrow <)] \qquad (61)$$

and

$$B_{3,\mu\kappa}(\mathbf{r}_1 t_1, \mathbf{r}_3 t_3) =$$
$$2 \int_{t_0}^{\infty} dt \int d^3r_2 d^3r_4 \theta(t_1 - t_3)\theta(t_3 - t)v(\mathbf{r}_2, \mathbf{r}_4)G^{<}(\mathbf{r}_2 t, \mathbf{r}_4 t)$$
$$\times \quad [G^{>}(\mathbf{r}_4 t, \mathbf{r}_3 t_3)\mathbf{j}_{\mathbf{op},\kappa}(\mathbf{r}_3)G^{>}(\mathbf{r}_3 t_3, \mathbf{r}_1 t_1)\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G^{<}(\mathbf{r}_1 t_1, \mathbf{r}_2 t)$$
$$+ \quad (> \leftrightarrow <)] \qquad (62)$$

and

$$C_{3,\mu\kappa}(\mathbf{r}_1 t_1, \mathbf{r}_3 t_3) =$$
$$-2 \int_{t_0}^{\infty} dt \int d^3r_2 d^3r_4 \theta(t_1 - t_3)\theta(t_1 - t)v(\mathbf{r}_2, \mathbf{r}_4)G^{<}(\mathbf{r}_2 t, \mathbf{r}_4 t)$$
$$\times \quad [G^{>}(\mathbf{r}_4 t, \mathbf{r}_3 t_3)\mathbf{j}_{\mathbf{op},\kappa}(\mathbf{r}_3)G^{<}(\mathbf{r}_3 t_3, \mathbf{r}_1 t_1)\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)G^{<}(\mathbf{r}_1 t_1, \mathbf{r}_2 t)$$
$$+ \quad (> \leftrightarrow <)] \qquad (63)$$

With these explict equations the x-only kernel is completely defined. The explicit solution for the kernel for the case of the electron gas will discussed in the next section.

6. The exchange-only kernel for the electron gas

The general equations for the xc-kernel for arbitrary inhomogeneous systems is rather involved. The equations become, however, much more

tractable for the homogeneous case for which they can be studied in more detail. Also the investigation of inhomogeneous systems benefits from such a study. The reason is that such kernels can in linear response theory be used in a local density approximation for the induced xc-vector potential $\delta \mathbf{A}_{xc}$ of the form

$$\delta A_{xc,\mu}(\mathbf{r}, \omega) = \sum_\nu \int d^3 r f^h_{xc,R,\mu\nu}(n_0(\mathbf{r}); |\mathbf{r} - \mathbf{r}'|, \omega) \delta j_\nu(\mathbf{r}'\omega) \qquad (64)$$

where we Fourier transformed from time to frequency space. Here $f^h_{xc,R,\mu\nu}$ is the xc-kernel of the homogeneous electron gas, n_0 is the ground state density and $\delta \mathbf{j}$ the induced current. In order to apply this local density approximation we need an explicit form for $f^h_{xc,R,\mu\nu}$. Since for the electron gas $f_{xc,\mu\nu}$ (from now on we will drop supindex h and subindex R, as we will only deal with the retarded electron gas kernel) depends on the difference between coordinates $\mathbf{r}$ and $\mathbf{r}'$ we can Fourier transform with respect to $\mathbf{r} - \mathbf{r}'$ and obtain the xc-kernel $f_{xc,\mu\nu}(\mathbf{q}, \omega)$ in Fourier space. This function has been investigated by several researchers in the limit $|\mathbf{q}| \to 0$ for fixed finite ω (see [33] and references therein). In the present work we consider a different limit. We will study $f_{xc,\mu\nu}$ in the whole $q - \omega$ plane but restrict ourselves to the exchange-only limit. The corresponding function is denoted as $f_{x,\mu\nu}(\mathbf{q}, \omega)$. From the previous section we see that it satisfies the following equation

$$\sum_{\nu\lambda} \chi_{R,s,\mu\nu}(\mathbf{q}, \omega) f_{x,\nu\lambda}(\mathbf{q}, \omega) \chi_{R,s,\lambda\kappa}(\mathbf{q}, \omega) = Q_{\mu\kappa}(\mathbf{q}, \omega) \qquad (65)$$

The Kohn-Sham response function $\chi_{s,\mu\nu}$ and the true response function $\chi_{\mu\nu}$ have the following structure [34] (we also drop subindex R from response functions from now on)

$$\chi_{s,\mu\nu}(\mathbf{q}, \omega) = \chi_{s,L}(\mathbf{q}, \omega) \frac{q_\mu q_\nu}{q^2} + \chi_{s,T}(\mathbf{q}, \omega)(\delta_{\mu\nu} - \frac{q_\mu q_\nu}{q^2}) \qquad (66)$$

which defines the longitudinal and transverse response functions χ_L and χ_T. This structure follows from isotropy of the electron gas.The xc-kernel has a similar structure. We can therefore define longitudinal and transverse exchange response kernels by

$$f_{x,\mu\nu}(\mathbf{q}, \omega) = \frac{1}{\omega^2} \left[f_{x,L}(\mathbf{q}, \omega) q_\mu q_\nu + f_{x,T}(q, \omega)(q^2 \delta_{\mu\nu} - q_\mu q_\nu) \right] \qquad (67)$$

If we insert this in Eq.(65) we obtain

$$
\begin{aligned}
Q_{\mu\kappa}(\mathbf{q}, \omega) &= \frac{q_\mu q_\kappa}{q^2} \chi_{s,L}(\mathbf{q}, \omega) \frac{q^2}{\omega^2} f_{x,L}(\mathbf{q}, \omega) \chi_{s,L}(\mathbf{q}, \omega) \\
&+ (\delta_{\mu\kappa} - \frac{q_\mu q_\kappa}{q^2}) \chi_{s,T}(\mathbf{q}, \omega) \frac{q^2}{\omega^2} f_{x,T}(\mathbf{q}, \omega) \chi_{s,T}(\mathbf{q}, \omega) \quad (68)
\end{aligned}
$$

If we define longitudinal and transverse parts of $Q_{\mu\kappa}$ in the usual way, i.e.

$$Q_{\mu\nu}(\mathbf{q}, \omega) = Q_L(\mathbf{q}, \omega)\frac{q_\mu q_\nu}{q^2} + Q_T(\mathbf{q}, \omega)(\delta_{\mu\nu} - \frac{q_\mu q_\nu}{q^2}) \tag{69}$$

then we obtain the equations

$$f_{x,L}(\mathbf{q}, \omega) = \frac{\omega^2}{q^2}\frac{Q_L(\mathbf{q}, \omega)}{\chi^2_{s,L}(\mathbf{q}, \omega)} \tag{70}$$

$$f_{x,T}(\mathbf{q}, \omega) = \frac{\omega^2}{q^2}\frac{Q_T(\mathbf{q}, \omega)}{\chi^2_{s,T}(\mathbf{q}, \omega)} \tag{71}$$

$$\tag{72}$$

We see that the equations for the longitudinal and transverse part completely decouple for the electron gas, as we would expect for an isotropic system. Since both $\chi_{s,L}$ and $\chi_{s,T}$ are known analytically (they were already calculated by Lindhard [35]) it remains to determine Q_L and Q_T. This can be done be working out the explicit formulas for the diagrams of the previous section. The longitudinal function Q_L has been worked out in detail before [36, 37, 38]. We therefore only present the expression for the transverse term:

$$\begin{aligned} Q_T(\mathbf{q}, \omega) &= \int \frac{d^3k}{(2\pi)^3} \int \frac{d^3k'}{(2\pi)^3} v(\mathbf{k} - \mathbf{k}')(n_\mathbf{k} - n_\mathbf{k+q})(n_{\mathbf{k}'} - n_{\mathbf{k}'+\mathbf{q}}) \\ &\times \left(\frac{(k^2 - (\mathbf{k} \cdot \mathbf{q})^2/q^2)}{(\omega - \epsilon_\mathbf{k+q} + \epsilon_\mathbf{k} + i\eta)^2} \right. \\ &\left. - \frac{(\mathbf{k} \cdot \mathbf{k}' - (\mathbf{k} \cdot \mathbf{q})(\mathbf{k}' \cdot \mathbf{q})/q^2)}{(\omega - \epsilon_\mathbf{k+q} + \epsilon_\mathbf{k} + i\eta)(\omega - \epsilon_{\mathbf{k}'+\mathbf{q}} + \epsilon_{\mathbf{k}'} + i\eta)} \right) \end{aligned}$$

where $n_\mathbf{k} = \theta(k_F - |\mathbf{k}|)$ is the Fermi function in which k_F is the Fermi wave vector. The first term in brackets arises from the self-energy diagrams whereas the second term originates from the vertex diagram. The function Q_T can be reduced to a two-dimensional integral that can be evaluated numerically. Its properties are currently being investigated in detail [39], as well as its applicability in local density type approximations for inhomogeneous systems.

7.   Conclusions

We showed how the basic equations of time-dependent current-density functional theory can be derived elegantly using the Keldysh formalism and provided some details that were left out in previous work. We also to derived an extension of the TDOPM equations that can be used

in current-density-functional theory and found the corresponding equations for the xc-kernels, that play an essential role in response theory. We further exploited a key feature of the Keldysh formalism, namely the possibility to perform a systematic diagrammatic expansion for nonequilibrium systems. In this way we were able to derive an explicit expression for the transverse exchange kernel of the electron gas. This expression is currently being investigated in detail in order to obtain new functionals that can be used in calculations of response properties of inhomogeneous systems.

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Appendix: A

In this section we will calculate the response in the orbitals due to a small change in the vector potential of the Kohn-Sham equations in Keldysh form

$$\left(i\partial_t + \frac{1}{2}(\nabla + i\delta\mathbf{A}_s(\mathbf{r}\tau))^2 - v_0(\mathbf{r}) \right)\varphi(\mathbf{r}\tau) = 0 \tag{A.1}$$

We write $\varphi = \phi + \delta\phi$ where ϕ_0 satisfies the Kohn-Sham equation without the external field $\delta\mathbf{A}_s$, i.e.

$$\left(i\partial_t + \frac{1}{2}\nabla^2 - v_0(\mathbf{r}) \right)\phi(\mathbf{r}\tau) = 0 \tag{A.2}$$

With these equations we find that

$$\left(i\partial_t + \frac{1}{2}\nabla^2 - v_0(\mathbf{r}) \right)\delta\phi(\mathbf{r}\tau) = \sum_\nu -\frac{i}{2}\partial_\nu(A_{s,\nu}\phi) - \frac{i}{2}A_{s,\nu}\partial_\nu\phi \tag{A.3}$$

where for the forward solution on the Keldysh contour we have initial value condition condition $\delta\phi(\mathbf{r}\tau_i) = 0$. This equation can be solved by expanding $\delta\phi$ in the unperturbed Kohn-Sham states $\phi_k(\mathbf{r}\tau) = \varphi_k(\mathbf{r})\exp(-i\epsilon_k t(\tau))$ where φ_k is an Kohn-Sham orbital of the unperturbed stationary system with eigenvalue ϵ_k. Thus we have

$$\delta\phi(\mathbf{r}\tau) = \sum_k c_k(\tau)\phi_k(\mathbf{r}\tau) \tag{A.4}$$

with initial value condition $c_k(\tau_i) = 0$. Substituting this into Eq.(A.3) and integrating over spatial coordinates after multiplying by $\phi_k^*(\mathbf{r}\tau)$ yields the solution

$$c_k(\tau_2) = -i\int_{\tau_i}^{\tau_2} d\tau_1 t'(\tau_1)\int d^3r_1 \sum_\nu \frac{1}{2i}(\phi_k^*(\mathbf{r}_1\tau_1)\partial_\nu\phi(\mathbf{r}_1\tau_1) - \partial_\nu\phi_k^*(\mathbf{r}_1\tau_1)\phi(\mathbf{r}_1\tau_1))\delta A_{s,\nu}(\mathbf{r}_1\tau_1) \tag{A.5}$$

We therefore find

$$\delta\phi(\mathbf{r}_2\tau_2) = -i\sum_k \int_{\tau_i}^{\tau_2} d\tau_1 t'(\tau_1)\int d^3r_1 \sum_\nu [\phi_k^*(\mathbf{r}_1\tau_1)\mathbf{j}_{\mathbf{op},\nu}(\mathbf{r}_1)\phi(\mathbf{r}_1\tau_1)]\phi_k(\mathbf{r}_2\tau_2)\delta A_{s,\nu}(\mathbf{r}_1\tau_1) \tag{A.6}$$

where we defined the one-particle current operator $\mathbf{j}_{\mathbf{op},\nu} = 1/2i(\overrightarrow{\partial}_\nu - \overleftarrow{\partial}_\nu)$, where the second derivarive $\overleftarrow{\partial}_\nu$) acts on the orbital to the left of the operator. We thus conclude that

$$\frac{\delta\phi(\mathbf{r}_2\tau_2)}{\delta A_{s,\nu}(\mathbf{r}_1\tau_1)} = -i\theta(\tau_2 - \tau_1)\sum_k \phi_k(\mathbf{r}_2\tau_2)[\phi_k^*(\mathbf{r}_1\tau_1)\mathbf{j}_{\mathbf{op},\nu}(\mathbf{r}_1)\phi(\mathbf{r}_1\tau_1)] \qquad (A.7)$$

We can carry out a similar procedure for the backward solution on the Keldysh contour, which satisfies $\delta\phi(\mathbf{r}\tau_f) = 0$. With this boundary condition we find

$$\delta\phi^*(\mathbf{r}_2\tau_2) = -i\sum_k \int_{\tau_2}^{\tau_f} d\tau_1 t'(\tau_1) \int d^3r_1 \sum_\nu [\phi_k^*(\mathbf{r}_1\tau_1)\mathbf{j}_{\mathbf{op},\nu}(\mathbf{r}_1)\phi(\mathbf{r}_1\tau_1)]^*\phi_k^*(\mathbf{r}_2\tau_2)\delta A_{s,\nu}(\mathbf{r}_1\tau_1)$$
$$(A.8)$$

and therefore

$$\begin{aligned}\frac{\delta\phi^*(\mathbf{r}_2\tau_2)}{\delta A_{s,\nu}(\mathbf{r}_1\tau_1)} &= -i\theta(\tau_1 - \tau_2)\sum_k \phi_k^*(\mathbf{r}_2\tau_2)[\phi_k^*(\mathbf{r}_1\tau_1)\mathbf{j}_{\mathbf{op},\nu}(\mathbf{r}_1)\phi(\mathbf{r}_1\tau_1)]^* \\ &= -i\theta(\tau_1 - \tau_2)\sum_k \phi_k^*(\mathbf{r}_2\tau_2)[\phi^*(\mathbf{r}_1\tau_1)\mathbf{j}_{\mathbf{op},\nu}(\mathbf{r}_1)\phi_k(\mathbf{r}_1\tau_1)] \quad (A.9)\end{aligned}$$

We have therefore obtained the desired equations for the orbital variations on the forward and backward parts of the Keldysh contour. These equations can now easily be used to calculate the Keldysh current response function of the Kohn-Sham system. Since

$$j_\mu(\mathbf{r}_1\tau_1) = n_0(\mathbf{r}_1)A_{s,\mu}(\mathbf{r}_1\tau_1) + \sum_{i=1}^N [\phi_i^*(\mathbf{r}_1\tau_1)\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)\phi_i(\mathbf{r}_1\tau_1)] \qquad (A.10)$$

we obtain using Eqns.(A.7) and (A.9)

$$\begin{aligned}\frac{\delta j_\mu(\mathbf{r}_1\tau_1)}{\delta A_{s,\nu}(\mathbf{r}_1\tau_1)} &= \delta_{\mu\nu}n_0(\mathbf{r}_1)\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta_C(t_1 - t_2) \\ &+ \theta(\tau_1 - \tau_2)\chi_{\mu\nu}^>(\mathbf{r}_1 t_1, \mathbf{r}_2 t_2) + \theta(\tau_2 - \tau_1)\chi_{\mu\nu}^<(\mathbf{r}_1 t_1, \mathbf{r}_2 t_2)(A.11)\end{aligned}$$

where the functional derivative is evaluated at the physical vector potential $\mathbf{A}_s = 0$ and where we defined

$$\chi_{\mu\nu}^>(\mathbf{r}_1 t_1, \mathbf{r}_2 t_2) =$$
$$-i\sum_{i=1}^N \sum_k [\phi_i^*(\mathbf{r}_1 t_1)\mathbf{j}_{\mathbf{op},\mu}(\mathbf{r}_1)\phi_k(\mathbf{r}_1 t_1)][\phi_k^*(\mathbf{r}_2 t_2)\mathbf{j}_{\mathbf{op},\nu}(\mathbf{r}_2)\phi_i(\mathbf{r}_2 t_2)] \quad (A.12)$$
$$\chi_{\mu\nu}^<(\mathbf{r}_1 t_1, \mathbf{r}_2 t_2) = \chi_{\nu\mu}^>(\mathbf{r}_2 t_2, \mathbf{r}_1 t_1) \qquad (A.13)$$

This defines the Kohn-Sham current response function in terms of the unperturbed orbitals.

Appendix: B

In this appendix we calculate the change in a Keldysh Green function due to a change in the vector potential. The zeroth order Green function satisfies

$$\left(i\partial_{t_1} + \frac{1}{2}\nabla_1^2 - v_0(\mathbf{r}_1)\right)G_0(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = \delta_C(t_1 - t_2) \qquad (B.1)$$

We then want to solve

$$\left(i\partial_{t_1} + \frac{1}{2}(\nabla_1 + i\delta A_s(\mathbf{r}_1\tau_1))^2 - v_0(\mathbf{r}_1)\right)(G_0(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) + \delta G(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2)) = \delta_C(t_1 - t_2)$$
(B.2)

for δG. This gives, to first order in δG, the equation

$$\left(i\partial_{t_1} + \frac{1}{2}\nabla_1^2 - v_0(\mathbf{r}_1)\right)\delta G(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2)$$

$$= -\frac{i}{2}\sum_\nu \partial_\nu^1(A_{s,\nu}(\mathbf{r}_1\tau_1)G_0(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2)) + A_{s,\nu}(\mathbf{r}_1\tau_1)\partial_\nu^1 G_0(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) \text{(B.3)}$$

The solution to this equation is

$$\delta G(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) =$$

$$\sum_\nu \int d^3r_3 \int_{\tau_i}^{\tau_f} d\tau_3 t'(\tau_3) G_0(\mathbf{r}_1\tau_1, \mathbf{r}_3\tau_3)$$

$$\times \quad (-\frac{i}{2}\partial_\nu^3(A_{s,\nu}(\mathbf{r}_3\tau_3)G_0(\mathbf{r}_3\tau_3, \mathbf{r}_2\tau_2)) - \frac{i}{2}A_{s,\nu}(\mathbf{r}_3\tau_3)\partial_\nu^3 G_0(\mathbf{r}_3\tau_3, \mathbf{r}_2\tau_2))$$

$$= \quad \sum_\nu \int d^3r_3 \int_{\tau_i}^{\tau_f} d\tau_3 t'(\tau_3) G_0(\mathbf{r}_1\tau_1, \mathbf{r}_3\tau_3)\mathbf{j}_{\mathbf{op},\nu}(\mathbf{r}_3)G_0(\mathbf{r}_3\tau_3, \mathbf{r}_2\tau_2)\delta A_{s,\nu}(\mathbf{r}_3\tau_3)$$

as can readily be checked by insertion into the original equation (B.3) and using the equation of motion for G_0. We therefore obtain

$$\frac{\delta G(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2)}{\delta A_\nu(\mathbf{r}_3, \tau_3)} = G_0(\mathbf{r}_1\tau_1, \mathbf{r}_3\tau_3)\mathbf{j}_{\mathbf{op},\nu}(\mathbf{r}_3)G_0(\mathbf{r}_3\tau_3, \mathbf{r}_2\tau_2) \tag{B.4}$$

Appendix: C

In this section we will describe derive some properties of product of Keldysh response functions and how they are transformed to real time response functions. All response functions are assumed to have the general structure

$$A(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = A^\delta(\mathbf{r}_1, \mathbf{r}_2, t_1)\delta_C(t_1 - t_2) + \theta(\tau_1 - \tau_2)A^>(1, 2) + \theta(\tau_2 - \tau_1)A^<(1, 2)$$
(C.1)

where we use the short notation $1 = \mathbf{r}_1 t_1$ and where δ_C is the contour delta function $\delta_C(t_1 - t_2) = \delta(\tau_1 - \tau_2)/t'(\tau_1)$. For instance, for the current response function we have

$$\chi_{\mu\nu}^\delta = n_0(\mathbf{r}_1)\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta_{\mu\nu} \tag{C.2}$$

$$\chi_{\mu\nu}^> = -i\langle\Psi_0|\Delta\mathbf{j}_{p,H,\mu}(1)\Delta\mathbf{j}_{p,H,\nu}(2)|\Psi_0\rangle \tag{C.3}$$

$$\chi_{\mu\nu}^< = -i\langle\Psi_0|\Delta\mathbf{j}_{p,H,\nu}(2)\Delta\mathbf{j}_{p,H,\mu}(1)|\Psi_0\rangle \tag{C.4}$$

We have already seen that if we let a Keldysh response function of the general form in Eq.(C.1) act on a perturbation $\delta v(\mathbf{r}_2 t(\tau_2))$ that is equal on both sides of the Keldysh contour that the reponse $\delta a(\mathbf{r}_1 t_1)$ is given as

$$\delta a(\mathbf{r}_1 t_1) = \int_C d^3r_2 d\tau_2 t'(\tau_2)A(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2)\delta v(\mathbf{r}_2 t_2)$$

$$= \int d^3r_2 dt_2 A_R(\mathbf{r}_1 t_1, \mathbf{r}_2 t_2)\delta v(\mathbf{r}_2 t_2) \tag{C.5}$$

where the retarded response function A_R is now a function in real time given by

$$A_R(1,2) = A^\delta(\mathbf{r}_1, \mathbf{r}_2, t_1)\delta(t_1 - t_2) + \theta(t_1 - t_2)(A^>(1,2) - A^<(1,2)) \qquad (C.6)$$

Let now A and B be Keldysh response functions of the form given in Eq.(C.1). We then want to show that the function

$$(AB)(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = \int d^3r_3 d\tau_3 t'(\tau_3) A(\mathbf{r}_1\tau_1, \mathbf{r}_3\tau_3) B(\mathbf{r}_3\tau_3, \mathbf{r}_2\tau_2) \qquad (C.7)$$

has the same general form Eq.(C.1) and that

$$(AB)_R(1,2) = \int d3 A_R(1,3) B_R(3,2) \qquad (C.8)$$

To show this we first write $A = a^\delta + a^r$ where a^δ and a^r are the singular and regular parts of A given by

$$a^\delta(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = A^\delta(\mathbf{r}_1, \mathbf{r}_2, t_1)\delta_C(t_1 - t_2) \qquad (C.9)$$
$$a^r(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) = \theta(\tau_1 - \tau_2)A^>(1,2) + \theta(\tau_2 - \tau_1)A^<(1,2) \qquad (C.10)$$

With this notation the integral (C.7) has the form

$$(AB) = \int (a^\delta b^\delta + a^\delta b^r + b^\delta a^r + a^r b^r) \qquad (C.11)$$

Let us start by evaluating the term $a^r b^r$. We have for the integral of $a^r b^r$ the following expression

$$\int_C d^3r_3 d\tau_3 t'(\tau_3) a^r(\mathbf{r}_1\tau_1, \mathbf{r}_3\tau_3) b^r(\mathbf{r}_3\tau_3, \mathbf{r}_2\tau_2)$$
$$= \int_{\tau_i}^{\tau_f} d^3r_3 d\tau_3 t'(\tau_3) \left(\theta(\tau_1 - \tau_3)\theta(\tau_3 - \tau_2)A^>B^> + \theta(\tau_1 - \tau_3)\theta(\tau_2 - \tau_3)A^>B^< \right.$$
$$+ \left. \theta(\tau_4 - \tau_3)\theta(\tau_4 - \tau_2)B^<C^> + \theta(\tau_4 - \tau_3)\theta(\tau_2 - \tau_4)B^<C^< \right)$$
$$= \theta(\tau_1 - \tau_2)(AB)^{r,>} + \theta(\tau_2 - \tau_1)(AB)^{r,<} \qquad (C.12)$$

where

$$(AB)^{r,>} = \int d^3r_3 \{ \int_{\tau_2}^{\tau_1} d\tau_3 t'(\tau_3) A^>B^> + \int_{\tau_i}^{\tau_2} d\tau_3 t'(\tau_3) A^>B^<$$
$$+ \int_{\tau_1}^{\tau_f} d\tau_3 t'(\tau_3) A^<B^> \} \qquad (C.13)$$
$$(AB)^{r,<} = \int d^3r_3 \{ \int_{\tau_1}^{\tau_2} d\tau_3 t'(\tau_3) A^<B^< + \int_{\tau_i}^{\tau_1} d\tau_3 t'(\tau_3) A^>B^<$$
$$+ \int_{\tau_2}^{\tau_f} d\tau_3 t'(\tau_3) A^<B^> \} \qquad (C.14)$$

Since all terms under the integral depend on $t(\tau)$ they can be transformed to real time integrals. This yields

$$(AB)^{r,>} = \int d^3r_3 \{ \int_{t_2}^{t_1} dt_3 A^>B^> + \int_{t_0}^{t_2} dt_3 A^>B^< - \int_{t_0}^{t_1} dt_3 A^<B^> \} \qquad (C.15)$$
$$(AB)^{r,<} = \int d^3r_3 \{ \int_{t_1}^{t_2} dt_3 A^<B^< + \int_{t_0}^{t_1} dt_3 A^>B^< - \int_{t_0}^{t_2} dt_3 A^<B^> \} \qquad (C.16)$$

This is our first intermediate result. We continue to discuss the singular terms in Eq.(C.11). The term containing two equal time deltafunctions is easily evaluated to give

$$\int a^\delta b^\delta \quad = \quad \frac{\delta(\tau_1 - \tau_2)}{t'(\tau_1)} \int d^3 r_3 A^\delta(\mathbf{r}_1, \mathbf{r}_3, t_1) B^\delta(\mathbf{r}_3, \mathbf{r}_2, t_1) \qquad (C.17)$$

This leaves us with the terms which contain one equal time delta function which are readily evaluated to be

$$\int (a^\delta b^r + a^r b^\delta) = \theta(\tau_1 - \tau_2)(AB)^{\delta,>} + \theta(\tau_2 - \tau_1)(AB)^{\delta,<} \qquad (C.18)$$

where

$$(AB)^{\delta,>} =$$
$$\int d^3 r_3 (A^\delta(\mathbf{r}_1, \mathbf{r}_3, t_1) B^>(\mathbf{r}_3 t_1, \mathbf{r}_2 t_2) + A^>(\mathbf{r}_1 t_1, \mathbf{r}_3 t_2) B^\delta(\mathbf{r}_3, \mathbf{r}_2, t_2))$$
$$(C.19)$$

with an analogous equation for $(AB)^{\delta,<}$. If we combine our results we see that (AB) has the form

$$
\begin{aligned}
(AB)(\mathbf{r}_1\tau_1, \mathbf{r}_2\tau_2) &= (AB)^\delta(\mathbf{r}_1, \mathbf{r}_2, t_1)\delta_C(t_1 - t_2) + \theta(\tau_1 - \tau_2)(AB)^>(1,2) \\
&+ \theta(\tau_2 - \tau_1)(AB)^<(1,2)
\end{aligned}
\qquad (C.20)
$$

where

$$(AB)^\delta(\mathbf{r}_1, \mathbf{r}_2, t_1) = \int d^3 r_3 A^\delta(\mathbf{r}_1, \mathbf{r}_3, t_1) B^\delta(\mathbf{r}_3, \mathbf{r}_2, t_1) \qquad (C.21)$$

$$(AB)^> = (AB)^{r,>} + (AB)^{\delta,>} \qquad (C.22)$$

$$(AB)^< = (AB)^{r,<} + (AB)^{\delta,<} \qquad (C.23)$$

We see that the product (AB) has the same structure as the original functions A and B. The retarded product

$$(AB)_R(1,2) = (AB)^\delta(\mathbf{r}_1, \mathbf{r}_2, t_1)\delta(t_1 - t_2) + \theta(t_1 - t_2)((AB)^>(1,2) - (AB)^<(1,2))$$
$$(C.24)$$

can now be evaluated. If we use

$$(AB)^{r,>} - (AB)^{r,<} = \int d^3 r_3 \int_{t_2}^{t_1} dt_3 (A^>(1,3) - A^<(1,3))(B^>(3,2) - B^<(3,2))$$
$$(C.25)$$

we obtain

$$
\begin{aligned}
(AB)_R &= \int d^3 r_3 \int_{t_0}^{\infty} dt_3 (A^\delta \delta(t_1 - t_3) + \theta(t_1 - t_3)[A^>(1,3) - A^<(1,3)]) \\
&\times (B^\delta \delta(t_3 - t_2) + \theta(t_3 - t_2)[B^>(3,2) - B^<(3,2)]) \\
&= \int d3 A_R(1,3) B_R(3,2)
\end{aligned}
\qquad (C.26)
$$

which proves our statement. By induction one can continue to prove the more general result

$$(A_1 A_2 \ldots A_n)_R(1,2) = \int d3 \ldots d(n+1) A_{1,R}(1,3) A_{2,R}(3,4) \ldots A_{n,R}(n+1,2)$$
$$(C.27)$$

Appendix: D

Here we demonstrate how the second order response function on the contour reduces to the causal one when acting on physical perturbations. The induced current has, up to second order, the expansion

$$
\begin{aligned}
\delta j_\mu(\bar{1}) &= \sum_\nu \int_C d2 \frac{\delta j_\mu(\bar{1})}{\delta A_\nu(\bar{2})} \delta A_\nu(\bar{2}) + \frac{1}{2} \sum_{\nu\kappa} \int_C d2d3 \frac{\delta^2 j_\mu(\bar{1})}{\delta A_\nu(\bar{2}) \delta A_\kappa(\bar{3})} \delta A_\nu(\bar{2}) \delta A_\kappa(\bar{3}) \\
&= \sum_\nu \int_C d2 \chi^{(1)}_{\mu\nu}(\bar{1}, \bar{2}) \delta A_\nu(\bar{2}) \\
&\quad + \frac{1}{2} \sum_{\nu\kappa} \int_C d2d3 \chi^{(2)}_{\mu\nu\kappa}(\bar{1}, \bar{2}, \bar{3}) \delta A_\nu(\bar{2}) \delta A_\kappa(\bar{3})
\end{aligned}
\tag{D.1}
$$

The second order response function has the form

$$
\begin{aligned}
\chi^{(2)}_{s,\mu\nu\kappa}(\bar{1}, \bar{2}, \bar{3}) &= (-i)^2 \langle T_C \Delta j_{p,H,\mu}(\bar{1}) \Delta j_{p,H,\nu}(\bar{2}) \Delta j_{p,H,\kappa}(\bar{3}) \rangle \\
&= \theta(\tau_1 - \tau_2)\theta(\tau_2 - \tau_3)\langle 123 \rangle + \theta(\tau_2 - \tau_3)\theta(\tau_3 - \tau_1)\langle 231 \rangle \\
&\quad + \theta(\tau_3 - \tau_1)\theta(\tau_1 - \tau_2)\langle 312 \rangle + \theta(\tau_1 - \tau_3)\theta(\tau_3 - \tau_2)\langle 132 \rangle \\
&\quad + \theta(\tau_3 - \tau_2)\theta(\tau_2 - \tau_1)\langle 321 \rangle + \theta(\tau_2 - \tau_1)\theta(\tau_1 - \tau_3)\langle 213 \rangle
\end{aligned}
\tag{D.2}
$$

where we introduced the short notation

$$
\langle ijk \rangle = (-i)^2 \langle \Delta j_{p,H,\mu}(\bar{i}) \Delta j_{p,H,\nu}(\bar{j}) \Delta j_{p,H,\kappa}(\bar{k}) \rangle
\tag{D.3}
$$

We then have for the second order change in the current (we use the short notation $2 = (\nu, \mathbf{r}_2, t_2(\tau_2))$ and $3 = (\kappa, \mathbf{r}_3, t_3(\tau_3))$ where $d2$ and $d3$ imply integration along the contour and summation over ν and κ) due to a perturbing field A:

$$
\begin{aligned}
2\delta j_\mu^{(2)}(1) = \\
&\int_{\tau_i}^{\tau_1} d2 \int_{\tau_i}^{\tau_2} d3 \langle 123 \rangle A_2 A_3 + \int_{\tau_1}^{\tau_f} d3 \int_{\tau_3}^{\tau_f} d2 \langle 231 \rangle A_2 A_3 \\
+ &\int_{\tau_1}^{\tau_f} d3 \int_{\tau_i}^{\tau_1} d2 \langle 312 \rangle A_2 A_3 + \int_{\tau_i}^{\tau_1} d3 \int_{\tau_i}^{\tau_3} d2 \langle 132 \rangle A_2 A_3 \\
+ &\int_{\tau_1}^{\tau_f} d2 \int_{\tau_2}^{\tau_f} d3 \langle 321 \rangle A_2 A_3 + \int_{\tau_1}^{\tau_f} d2 \int_{\tau_i}^{\tau_1} d3 \langle 213 \rangle A_2 A_3 \\
= &\int_{t_0}^{t_1} d2 \int_{t_0}^{t_2} d3 \langle 123 \rangle A_2 A_3 + \int_{t_0}^{t_1} d2 \int_{t_0}^{t_2} d3 \langle 321 \rangle A_2 A_3 \\
- &\int_{t_0}^{t_1} d2 \int_{t_0}^{t_1} d3 \langle 213 \rangle A_2 A_3 + \int_{t_0}^{t_1} d2 \int_{t_0}^{t_2} d3 \langle 123 \rangle A_2 A_3 \\
+ &\int_{t_0}^{t_1} d2 \int_{t_0}^{t_2} d3 \langle 321 \rangle A_2 A_3 - \int_{t_0}^{t_1} d2 \int_{t_0}^{t_1} d3 \langle 213 \rangle A_2 A_3
\end{aligned}
\tag{D.4}
$$

where in some terms we interchanged the s 2 and 3 and use. If we use the expression

$$
\int_{t_0}^{t_1} d2 \int_{t_0}^{t_1} d3 \langle 213 \rangle A_2 A_3 = \int_{t_0}^{t_1} d2 \int_{t_0}^{t_2} d3 \langle 213 \rangle A_2 A_3 + \int_{t_0}^{t_1} d2 \int_{t_0}^{t_2} d3 \langle 312 \rangle A_2 A_3
\tag{D.5}
$$

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(which follows by inspection of the integration regions and changing integration order and s) we obtain

$$
\begin{aligned}
2\delta j_\mu^{(2)}(1) &= 2\int_{t_0}^{t_1} d2 \int_{t_0}^{t_3} \langle 123 + 321 - 213 - 312 \rangle A_2 A_3 \\
&= 2\int_{t_0}^{t_1} d2 \int_{t_0}^{t_3} \langle [[1,2],3] \rangle A_2 A_3
\end{aligned} \tag{D.6}
$$

This means that in physical time the second order response function is given by

$$
\chi_{s,\mu\nu\kappa}^{(2)}(1,2,3) = (-i)^2 \theta(t_1 - t_2)\theta(t_2 - t_3)\langle \Psi_0 | [[j_{p,H,\mu}(1), j_{p,H,\nu}(2)], j_{p,H,\kappa}(3)] | \Psi_0 \rangle \tag{D.7}
$$

which is the usual retarded second order response function. Note that taken out of the integrand this response function is not uniquely defined. We can, for instance, still symmetrize the function with respect to coordinates 2 and 3.

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