

Basic concepts of TDDFT

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Overview

- The Runge-Gross theorem
- The extended Runge-Gross theorem
- Invertibility of the density response function
- The action principle
- Conserving approximations
- Conclusions and outlook

The Runge-Gross Theorem

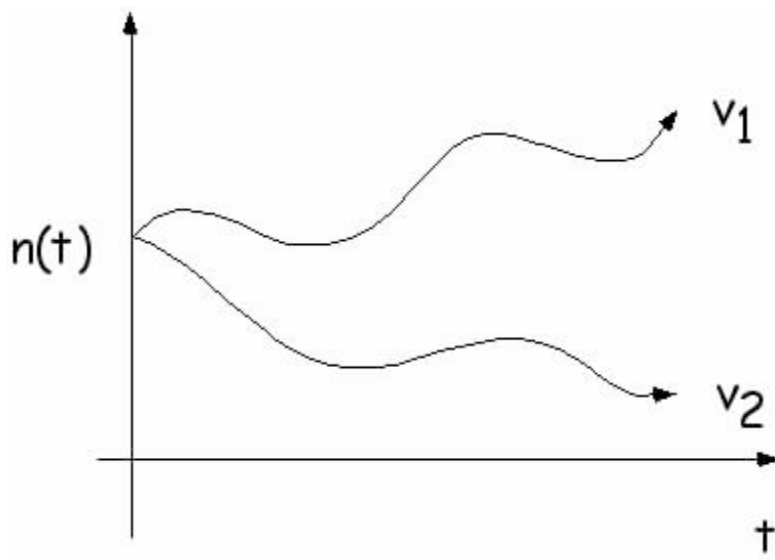
E.Runge and E.K.U.Gross
(Phys.Rev.Lett.52,997 (1984))

$$(i\partial_t - (\hat{T} + \hat{V}_1(t) + \hat{W}))\Psi_1(t) = 0$$

$$(i\partial_t - (\hat{T} + \hat{V}_2(t) + \hat{W}))\Psi_2(t) = 0$$

$$\Psi_1(t_0) = \Psi_2(t_0) = \Psi_0$$

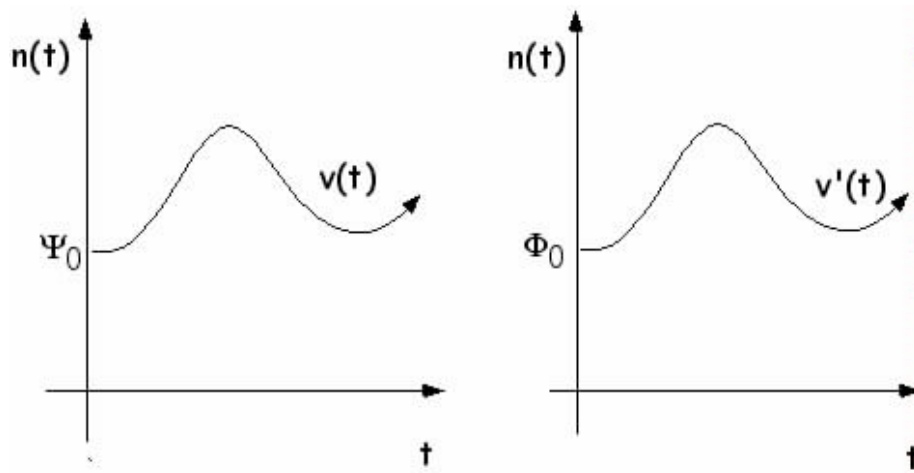
$$\hat{V}_1(t) \neq \hat{V}_2(t) + C(t) \implies n_1(t) \neq n_2(t)$$



Any expectation value is a
functional of the
density and the initial state

$$O[n, \Psi_0](t) = \langle \Psi[n](t) | \hat{O} | \Psi[n](t) \rangle$$

Different interactions and initial states



True system

Kohn-Sham system

The Kohn-Sham system has the same density as the true system for a different two-particle interaction and a different initial state

Suppose we have found the density $n(\mathbf{r},t)$ in an interacting system with potential $v(\mathbf{r},t)$

Can exactly the same density $n(\mathbf{r},t)$ be reproduced by an external potential $v'(\mathbf{r},t)$ in a system with different initial state and a different two-particle interaction?

If so, would this potential be unique (modulo a purely time-dependent function)?

Extended Runge-Gross theorem

R.van Leeuwen

(Phys.Rev.Lett.82,3863 (1999),

Int.J.Mod.Phys.B15, 1969 (2001))

Continuity equation for the density

$$n(\mathbf{r}t) = \langle \Psi(t) | \hat{n}(\mathbf{r}) | \Psi(t) \rangle$$

$$\partial_t n(\mathbf{r}t) = -i \langle \Psi(t) | [\hat{n}(\mathbf{r}), \hat{H}(t)] | \Psi(t) \rangle = -\nabla \cdot \mathbf{j}(\mathbf{r}t)$$

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

$$\mathbf{j}(\mathbf{r}t) = \langle \Psi(t) | \hat{\mathbf{j}}(\mathbf{r}) | \Psi(t) \rangle$$

Continuity equation for the current density

$$\partial_t \mathbf{j}(\mathbf{r}t) = -i \langle \Psi(t) | [\hat{\mathbf{j}}(\mathbf{r}), \hat{H}(t)] | \Psi(t) \rangle$$

$$\partial_t j_k(\mathbf{r}t) = -n(\mathbf{r}t) \partial_k v(\mathbf{r}t) - \sum_i \partial_i T_{ik}(\mathbf{r}t) - W_k(\mathbf{r}t)$$

$$\hat{T}_{ik}(\mathbf{r}) = \frac{1}{2} \sum_{\sigma} [\partial_i \hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) \partial_k \hat{\psi}_{\sigma}(\mathbf{r}) + \partial_k \hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) \partial_i \hat{\psi}_{\sigma}(\mathbf{r}) - \frac{1}{2} \partial_i \partial_k (\hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) \hat{\psi}_{\sigma}(\mathbf{r}))]$$

$$\hat{W}_k(\mathbf{r}) = \sum_{\sigma, \sigma'} \int d^3 r' \hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) \hat{\psi}_{\sigma'}^{\dagger}(\mathbf{r}') \partial_k w(|\mathbf{r} - \mathbf{r}'|) \hat{\psi}_{\sigma'}(\mathbf{r}') \hat{\psi}_{\sigma}(\mathbf{r})$$

G.Vignale has recently extended this proof to current-density functional theory

Combining the continuity equations for the density and the current density gives

$$\partial_t^2 n(\mathbf{r}t) = \nabla \cdot (n(\mathbf{r}t) \nabla v(\mathbf{r}t)) + q(\mathbf{r}t)$$

$$q(\mathbf{r}t) = \langle \Psi(t) | \hat{q}(\mathbf{r}) | \Psi(t) \rangle$$

$$\hat{q}(\mathbf{r}) = \sum_{i,k} \partial_i \partial_k \hat{T}_{ik}(\mathbf{r}) + \sum_k \partial_k \hat{W}_k(\mathbf{r})$$

Now we consider two systems with different external potentials and two-particle interactions, but with identical electron densities:

$$\begin{aligned} \hat{H}(t) &= \hat{T} + \hat{V}(t) + \hat{W} & (i\partial - \hat{H}(t))\Psi(t) &= 0 & \Psi(t_0) &= \Psi_0 \\ \hat{H}'(t) &= \hat{T} + \hat{V}'(t) + \hat{W}' & (i\partial - \hat{H}'(t))\Phi(t) &= 0 & \Phi(t_0) &= \Phi_0 \end{aligned}$$

We further demand that at the initial time the density and its first time-derivative are the same:

$$\begin{aligned} \langle \Psi_0 | \hat{n}(\mathbf{r}) | \Psi_0 \rangle &= \langle \Phi_0 | \hat{n}(\mathbf{r}) | \Phi_0 \rangle \\ \langle \Psi_0 | \nabla \cdot \hat{\mathbf{j}}(\mathbf{r}) | \Psi_0 \rangle &= \langle \Phi_0 | \nabla \cdot \hat{\mathbf{j}}(\mathbf{r}) | \Phi_0 \rangle \end{aligned}$$

We now want to determine $v'(\mathbf{r}t)$ such that also the higher derivatives of the density are the same

For the second system we have the equation

$$\partial_t^2 n(\mathbf{r}t) = \nabla \cdot (n(\mathbf{r}t) \nabla v'(\mathbf{r}t)) + q'(\mathbf{r}t)$$

Explicit construction of the potential

$$\begin{aligned}0 &= \nabla \cdot (n(\mathbf{r}t) \nabla \Delta v(\mathbf{r}t)) + \Delta q(\mathbf{r}t) \\ \Delta v(\mathbf{r}t) &= v(\mathbf{r}t) - v'(\mathbf{r}t) \\ \Delta q(\mathbf{r}t) &= q(\mathbf{r}t) - q'(\mathbf{r}t)\end{aligned}$$

k-fold differentiation at $t = t_0$ gives

$$0 = \Delta q_k(\mathbf{r}) + \sum_{l=0}^k \binom{k}{l} \nabla \cdot (n_{k-l}(\mathbf{r}) \nabla \Delta v_l(\mathbf{r}))$$

where $f_k(\mathbf{r}) = \partial_t^k f(\mathbf{r}t)|_{t=t_0}$

This can be rewritten as

$$\nabla \cdot (n_0(\mathbf{r}) \nabla \Delta v_k(\mathbf{r})) = -\Delta q_k(\mathbf{r}) - \sum_{l=0}^{k-1} \binom{k}{l} \nabla \cdot (n_{k-l}(\mathbf{r}) \nabla \Delta v_l(\mathbf{r}))$$

This equation provides an iterative scheme to construct the potential $v'(\mathbf{r}t)$

$$k=0 \quad \nabla \cdot (n_0(\mathbf{r}) \nabla \Delta v_0(\mathbf{r})) = -\Delta q_0(\mathbf{r}) = \langle \Phi_0 | \hat{q}'(\mathbf{r}) | \Phi_0 \rangle - \langle \Psi_0 | \hat{q}(\mathbf{r}) | \Psi_0 \rangle$$

This equation must be solved with the boundary condition $\Delta v_0(\mathbf{r}) \rightarrow 0 \quad (r \rightarrow \infty)$

$$\begin{aligned} k=1 \quad \nabla \cdot (n_0(\mathbf{r}) \nabla \Delta v_1(\mathbf{r})) &= -\Delta q_1(\mathbf{r}) - \nabla \cdot (n_1(\mathbf{r}) \nabla \Delta v_0(\mathbf{r})) \\ &= i \langle \Psi_0 | [\hat{q}(\mathbf{r}), \hat{H}(t_0)] | \Psi_0 \rangle - i \langle \Phi_0 | [\hat{q}'(\mathbf{r}), \hat{H}'(t_0)] | \Phi_0 \rangle - \nabla \cdot (n_1(\mathbf{r}) \nabla \Delta v_0(\mathbf{r})) \end{aligned}$$

And so on

$$v'_0(\mathbf{r}) = v_0(\mathbf{r}) - \Delta v_0(\mathbf{r})$$

In this way we can construct

$$v'_1(\mathbf{r}) = v_1(\mathbf{r}) - \Delta v_1(\mathbf{r})$$

...

$$v'_k(\mathbf{r}) = v_k(\mathbf{r}) - \Delta v_k(\mathbf{r})$$

and we obtain

$$v'(\mathbf{r}t) = \sum_k \frac{1}{k!} v'_k(\mathbf{r}) (t - t_0)^k$$

Special cases

$$A) \quad (\Psi_0 = \Phi_0, \quad \hat{W} = \hat{W}') \implies \Delta v_k(\mathbf{r}) = 0 \quad \text{Runge-Gross theorem}$$

$$B) \quad \hat{W}' = 0 \quad \text{Construction procedure for the time-dependent Kohn-Sham potential}$$

Invertibility of the density response function

R.van Leeuwen
(Int.J.Mod.Phys.B15, 1969 (2001))

We look at a density response $\delta n(\mathbf{r}_1 t_1) = -i \int_0^{t_1} dt_2 d^3 r_2 \langle \Psi_0 | [\Delta \hat{n}_H(\mathbf{r}_1 t_1), \Delta \hat{n}_H(\mathbf{r}_2 t_2)] | \Psi_0 \rangle \phi(\mathbf{r}_2 t_2)$

Using the Lehmann representation this can be rewritten as

$$\delta n(\mathbf{r}_1 t_1) = i \sum_n f_n^*(\mathbf{r}_1) \int_0^{t_1} dt_2 a_n(t_2) \exp i\Omega_n(t_1 - t_2) + c.c.$$

$$a_n(t) = \int d^3 r f_n(\mathbf{r}) \phi(\mathbf{r} t)$$

$$f_n(\mathbf{r}) = \langle \Psi_0 | \Delta \hat{n}(\mathbf{r}) | \Psi_n \rangle$$

$$\Omega_n = E_n - E_0 > 0 \quad (\text{excitation energies})$$

We now use the convolution property of Laplace transforms

$$Lf(s) = \int_0^\infty dt e^{-st} f(t)$$

$$(f * g)(t) = \int_0^t d\tau f(\tau) g(t - \tau)$$

$$L(f * g)(s) = Lf(s) Lg(s)$$

To obtain

$$\begin{aligned} \int d^3 r_1 L\phi(\mathbf{r}_1 s) L(\delta n)(\mathbf{r}_1 s) &= i \sum_n \frac{1}{s - i\Omega_n} |La_n(s)|^2 + c.c. \\ &= -2 \sum_n \frac{\Omega_n}{s^2 + \Omega_n^2} |La_n(s)|^2 \end{aligned}$$

Let us now assume that $\delta n = 0$

Then

$$0 = \sum_n \frac{\Omega_n}{s^2 + \Omega_n^2} |La_n(s)|^2 \implies La_n = 0 \implies a_n(t) = 0$$

From this we obtain

$$\int d^3r \Delta \hat{n}(r) \phi(\mathbf{r}t) |\Psi_0\rangle = \sum_n |\Psi_n\rangle \int d^3r \langle \Psi_n | \Delta \hat{n}(\mathbf{r}) | \Psi_0 \rangle \phi(\mathbf{r}t) = \sum_n a_n(t) |\Psi_n\rangle = 0$$

And therefore

$$\sum_{k=1}^N \Delta \phi(\mathbf{r}_k t) |\Psi_0\rangle = 0$$
$$\Delta \phi(\mathbf{r}t) = \phi(\mathbf{r}t) - \frac{1}{N} \int d^3r n_0(\mathbf{r}) \phi(\mathbf{r}t)$$

We find that $\Delta \phi = 0$ and consequently

$$\phi(\mathbf{r}t) = C(t)$$

We have proven the Runge-Gross theorem for Laplace-transformable switch-on potentials. This is an extension of the set of potentials allowed in the Runge-Gross proof. However, we needed the extra assumption that the system is initially in the ground state.

The action principle

$$A[\Psi] = \int_{t_0}^{t_1} dt \langle \Psi | i\partial_t - \hat{H}(t) | \Psi \rangle$$

Requiring the action to be stationary leads under certain conditions to the time-dependent Schrödinger equation (TDSE)

A) $\delta\Psi(t_0) = \delta\Psi(t_1) = 0$

$$\begin{aligned} \delta A &= \int_{t_0}^{t_1} dt \langle \delta\Psi | i\partial_t - \hat{H}(t) | \Psi \rangle + c.c. + [i\langle \Psi | \delta\Psi \rangle]_{t_0}^{t_1} \\ &= 2\text{Re} \int_{t_0}^{t_1} dt \langle \delta\Psi | i\partial_t - \hat{H}(t) | \Psi \rangle \\ &= 2\text{Re} \int_{t_0}^{t_1} dt \langle \delta\Psi_1 | i\partial_t - \hat{H}(t) | \Psi \rangle - 2\text{Im} \int_{t_0}^{t_1} dt \langle \delta\Psi_2 | i\partial_t - \hat{H}(t) | \Psi \rangle \end{aligned}$$

where $\delta\Psi = \delta\Psi_1 + i\delta\Psi_2$

If the real and imaginary part of $\delta\Psi$ can be varied independently then

$$\delta A = 0 \implies (i\partial_t - \hat{H}(t))\Psi(t) = 0$$

B) More elegantly. Assume that $\delta\Psi = \delta\Phi$ and $\delta\Psi = i\delta\Phi$ are allowed variations for arbitrary $\delta\Phi$
 Then

$$0 = \delta A = \int_{t_0}^{t_1} dt \langle \delta\Phi | i\partial_t - \hat{H}(t) | \Psi \rangle + \int_{t_0}^{t_1} dt \langle \Psi | i\partial_t - \hat{H}(t) | \delta\Phi \rangle$$

$$0 = \delta A = -i \int_{t_0}^{t_1} dt \langle \delta\Phi | i\partial_t - \hat{H}(t) | \Psi \rangle + i \int_{t_0}^{t_1} dt \langle \Psi | i\partial_t - \hat{H}(t) | \delta\Phi \rangle$$

Therefore

$$0 = \int_{t_0}^{t_1} dt \langle \delta\Phi | i\partial_t - \hat{H}(t) | \Psi \rangle \implies (i\partial_t - \hat{H}(t))\Psi(t) = 0$$

TDDFT action

First guess

$$A[n] = \int_{t_0}^{t_1} dt \langle \Psi[n] | i\partial_t - \hat{H}(t) | \Psi[n] \rangle$$

$$(n, \Psi_0) \rightarrow \Psi[n, \Psi_0](t) \quad \text{up to a phase factor}$$

We can fix the gauge, for instance by requiring that $\Psi[n, \Psi_0](t)$ evolves in a potential that vanishes at infinity.

The TDDFT action $A[n]$ is defined on a restricted set of wavefunctions, namely all v -representable wavefunctions that evolve from a given initial state.

Can on this restricted set of wavefunctions the variations required to derive the TDSE still be carried out?

A) The TDSE is first order in time. The variation of the wavefunction can only be fixed at one endpoint, for instance $\delta\Psi(t_0) = 0$

The real and imaginary parts of the variation of the wavefunction are not independent

B) If $\delta\Psi$ is a variation generated by some δv then

$$(i\partial_t - \hat{H}_v(t))|\delta\Psi\rangle = \delta v|\Psi\rangle$$

Variation $i\delta\Psi$ is then generated by $i\delta v$

The variations required to derive the TDSE can not be carried out!!

Can we find a proper action functional for TDDFT?

We want the Kohn-Sham potential to be the functional derivative of some action with respect to the density. Let us therefore assume that there are functionals such that:

$$\frac{\delta A}{\delta n(\mathbf{r}t)} = v(\mathbf{r}t) \qquad \frac{\delta A_s}{\delta n(\mathbf{r}t)} = v_s(\mathbf{r}t)$$

We can then define

$$A[n] = A_s[n] - \frac{1}{2} \int d^3r d^3r' dt n(\mathbf{r}t) n(\mathbf{r}'t) w(|\mathbf{r} - \mathbf{r}'|) - A_{xc}[n]$$

Such that

$$v_s(\mathbf{r}t) = v(\mathbf{r}t) + \int d^3r' n(\mathbf{r}'t) w(|\mathbf{r} - \mathbf{r}'|) + v_{xc}(\mathbf{r}t)$$

$$v_{xc}(\mathbf{r}t) = \frac{\delta A_{xc}}{\delta n(\mathbf{r}t)}$$

and derive time-dependent Kohn-Sham equations

Can we construct a functional $A[n]$ with the property: $\frac{\delta A}{\delta n(\mathbf{r}t)} = v(\mathbf{r}t) \quad ?$

If there were such a functional then the Legendre transformed functional

$$\tilde{A}[v] = -A[n] + \int d^3\mathbf{r}dt n(\mathbf{r}t)v(\mathbf{r}t)$$

would have the property

$$\frac{\delta \tilde{A}}{\delta v(\mathbf{r}t)} = - \int \frac{\delta A}{\delta n} \frac{\delta n}{\delta v} + \int \frac{\delta n}{\delta v} v + n = n(\mathbf{r}t)$$

Then for the second derivative we would have

$$\frac{\delta^2 \tilde{A}}{\delta v(\mathbf{r}t) \delta v(\mathbf{r}'t')} = \frac{\delta n(\mathbf{r}t)}{\delta v(\mathbf{r}'t')}$$

Symmetric

Causal

Zero for $t' > t$

The symmetry and causality conditions contradict each other:

There is no functional such that:

$$\frac{\delta A}{\delta n(\mathbf{r}t)} = v(\mathbf{r}t)$$

The Kohn-Sham potential is not the functional derivative of some action functional defined in real time!!

The construction of an action functional

Consider the evolution operator corresponding to the TDSE $|\Psi(t)\rangle = \hat{U}(t, t')|\Psi(t')\rangle$

The evolution operator satisfies the equations of motion

$$\begin{aligned} i\partial_t \hat{U}(t, t') &= \hat{H}(t) \hat{U}(t, t') \\ i\partial_{t'} \hat{U}(t, t') &= -\hat{U}(t, t') \hat{H}(t') \end{aligned} \quad \text{with boundary condition} \quad \hat{U}(t, t) = 1$$

By integration of these equations one obtains the formal expression $\hat{U}(t, t') = T \exp(-i \int_{t'}^t ds \hat{H}(s))$

Let us now make a change in the potential. The equations for the evolution operator then become:

$$\begin{aligned} i\partial_t \delta \hat{U}(t, t') &= \delta \hat{V}(t) \hat{U}(t, t') + \hat{H}(t) \delta \hat{U}(t, t') \\ i\partial_{t'} \delta \hat{U}(t, t') &= -\hat{U}(t, t') \delta \hat{V}(t') - \delta \hat{U}(t, t') \hat{H}(t') \end{aligned} \quad \text{with boundary condition} \quad \delta \hat{U}(t, t) = 0$$

The solution to these equations is

$$\delta \hat{U}(t, t') = -i \int_{t'}^t ds \hat{U}(t, s) \delta \hat{V}(s) \hat{U}(s, t')$$

We know take the perturbation of the form

$$\delta\hat{V}(t) = \int d^3r \hat{n}(\mathbf{r}) \delta v(\mathbf{r}t)$$

Then

$$\langle \Psi_0 | \delta\hat{U}(t_1, t_0) | \Psi_0 \rangle = -i \int_{t_0}^{t_1} dt \int d^3r \langle \Psi_0 | \hat{U}(t_1, t) \hat{n}(\mathbf{r}) \hat{U}(t, t_0) | \Psi_0 \rangle \delta v(\mathbf{r}t)$$

In other words, if we define

$$\tilde{A}[v] = i \langle \Psi_0 | \hat{U}(t_1, t_0) | \Psi_0 \rangle$$

Then

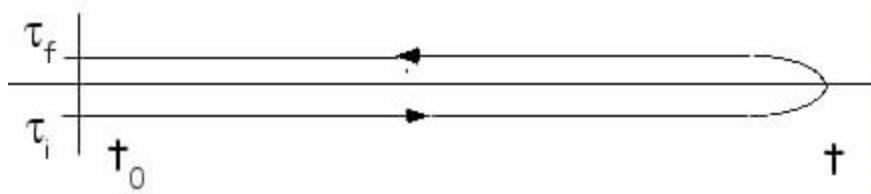
$$\frac{\delta \tilde{A}}{\delta v(\mathbf{r}t)} = \langle \Psi_0 | \hat{U}(t_1, t) \hat{n}(\mathbf{r}) \hat{U}(t, t_0) | \Psi_0 \rangle$$

This is not quite what we wanted, since the density is given by

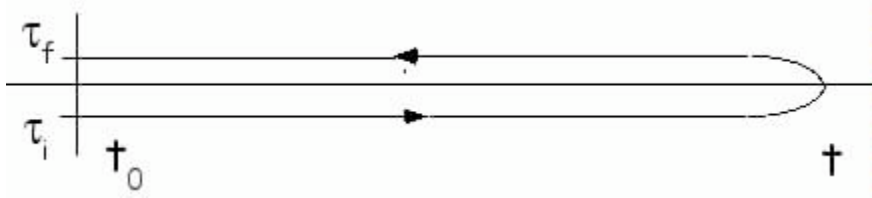
$$n(\mathbf{r}t) = \langle \Psi(t) | \hat{n}(\mathbf{r}) | \Psi(t) \rangle = \langle \Psi_0 | \hat{U}(t_0, t) \hat{n}(\mathbf{r}) \hat{U}(t, t_0) | \Psi_0 \rangle$$

$$t_0 \xleftarrow{\quad} t \quad t \xleftarrow{\quad} t_0$$

We define a time contour parametrized by an underlying pseudotime $t(\tau)$



$$t_0 = t(\tau_i) = t(\tau_f)$$



$$t_0 = t(\tau_i) = t(\tau_f)$$

We further define the contour-ordered evolution operator as $\hat{U}(\tau_f, \tau_i) = T_C \exp(-i \int_C dt \hat{H}(t))$

We further define the action as

$$\tilde{A}[v] = i \ln \langle \Psi_0 | \hat{U}(\tau_f, \tau_i) | \Psi_0 \rangle$$

then its derivative evaluated at a physical potential (equal on forward and backward part of the contour) is

$$\frac{\delta \tilde{A}[v]}{\delta v(\mathbf{r}\tau)} = \frac{\langle \Psi_0 | \hat{U}(\tau_f, \tau) \hat{n}(\mathbf{r}) \hat{U}(\tau, \tau_i) | \Psi_0 \rangle}{\langle \Psi_0 | \hat{U}(\tau_f, \tau_i) | \Psi_0 \rangle} = \langle \Psi_0 | \hat{U}(t_0, t) \hat{n}(\mathbf{r}) \hat{U}(t, t_0) | \Psi_0 \rangle$$

This functional can be used to derive the time-dependent Kohn-Sham equations.

For its second derivative we obtain:

$$\frac{\delta^2 \tilde{A}}{\delta v(\mathbf{r}_1 t_1) \delta v(\mathbf{r}_2 t_2)} = -i \langle \Psi_0 | T_C \Delta \hat{n}_H(\mathbf{r}_1 t_1) \Delta \hat{n}_H(\mathbf{r}_2 t_2) | \Psi_0 \rangle = \chi(\mathbf{r}_1 t_1, \mathbf{r}_2 t_2)$$

What about the causality-symmetry problem?

The contour-ordered response function has the form

$$\chi(1, 2) = \theta_C(t_1, t_2)\chi^>(1, 2) + \theta_C(t_2, t_1)\chi^<(1, 2)$$

We calculate the density change due to a physical perturbation

$$\begin{aligned}\delta n(1) &= \int_C d2 \chi(1, 2) \delta v(2) \\ &= \int_{t_0}^{t_1} d2 \chi^>(1, 2) \delta v(2) + \int_{t_1}^{t_0} d2 \chi^<(1, 2) \delta v(2) \\ &= \int_{t_0}^{t_1} d2 (\chi^>(1, 2) - \chi^<(1, 2)) \delta v(2) \\ &= \int d2 \chi_R(1, 2) \delta v(2)\end{aligned}$$

where the retarded response function is given by

$$\chi_R(1, 2) = \theta(t_1 - t_2)(\chi^>(1, 2) - \chi^<(1, 2)) = -i\theta(t_1 - t_2)\langle\Psi_0|[\hat{n}_H(1), \hat{n}_H(2)]|\Psi_0\rangle$$

The response functions are symmetric in the contour time variables but become retarded functions when acting in physical time

Conserving approximations

R.van Leeuwen and N.E.Dahlen
(vol.157, "Enrico Fermi" School of Physics, 2004)

The nonequilibrium Green function is defined as

$$G(1, 2) = -i \langle T_C \hat{\psi}(1) \hat{\psi}^\dagger(2) \rangle$$

and satisfies the equations of motion

$$(i\partial_{t_1} - h_0(1) - v_H(1))G(1, 2) = \delta(1, 2) + \int_C d3 \Sigma(1, 3)G(3, 2)$$

$$(-i\partial_{t_2} - h_0(1) - v_H(1))G(1, 2) = \delta(1, 2) + \int_C d3 G(1, 3)\Sigma(3, 2)$$

with the Kubo-Martin-Schwinger boundary conditions

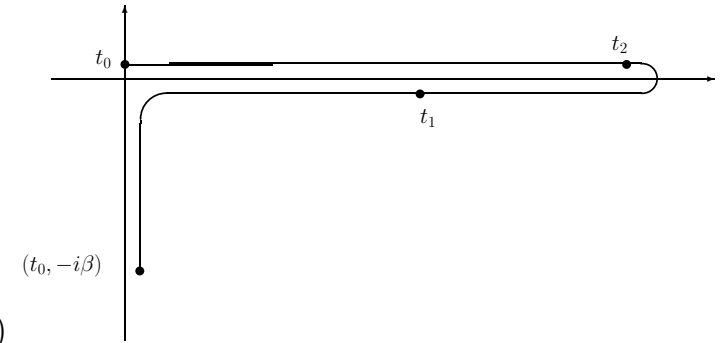
$$G(\mathbf{x}_1 t_0 - i\beta, 2) = -G(\mathbf{x}_1 t_0, 2)$$

$$G(1, \mathbf{x}_2 t_0 - i\beta) = -G(1, \mathbf{x}_2 t_0)$$

G.Baym, Phys.Rev.127, 1391 (1962)

The Green function satisfies the macroscopic conservation laws of energy, number and (angular) momentum whenever the self-energy is Phi-derivable:

$$\Sigma(1, 2) = \frac{\delta \Phi}{\delta G(2, 1)}$$



well known Phi-derivable schemes:
Hartree-Fock, GW, T-matrix

The Sham-Schlüter equation

R.van Leeuwen
(Phys.Rev.Lett.76, 3610 (1996))

The Kohn-Sham Green function satisfies the equations of motion

$$\begin{aligned}(i\partial_{t_1} - h_s(1))G_s(1, 2) &= \delta(1, 2) & G_s(\mathbf{x}_1 t_0 - i\beta, 2) &= -G_s(\mathbf{x}_1 t_0, 2) \\ (-i\partial_{t_2} - h_s(1))G_s(1, 2) &= \delta(1, 2) & G_s(1, \mathbf{x}_2 t_0 - i\beta) &= -G_s(1, \mathbf{x}_2 t_0)\end{aligned}$$

We can then derive

$$G(1, 2) = G_s(1, 2) + \int_C d3d4 G_s(1, 3)(\Sigma(3, 4) - v_{xc}(3)\delta(3, 4))G(4, 2)$$

Since the true and Kohn-Sham Green function give the same density we have

$$n(1) = -iG(1, 1^+) = -iG_s(1, 1^+)$$

and therefore

$$\int_C d2 G_s(1, 2)G(2, 1)v_{xc}(2) = \int_C d2d3 G_s(1, 2)\Sigma(2, 3)G(3, 1)$$

If the selfenergy in this expression is Φ -derivable then the xc-potential satisfies the zero-force theorem.

$$\int d^3r n(\mathbf{r}t)\nabla v_{xc}(\mathbf{r}t) = 0$$

Conclusions and outlook

- The Runge-Gross theorem can be extended and an explicit construction procedure for the time-dependent xc-potential can be given.
- Invertibility of the density response for all Laplace-transformable switch-on potentials can be proven.
- Subtle difficulties with the TDDFT action can be avoided by the introduction of a Keldysh or contour action.
- Using nonequilibrium Green functions conserving approximations for the xc-potential can be constructed.

New developments:

- Combination of nonequilibrium Green function and time-dependent density functional theory for transport
(see also G.Stefanucci, C.-O.Almbladh, Phys.Rev.B69,195318,(2004))
- Development of a multicomponent density-functional theory for coupled dynamics of electrons and nuclei (Berlin collaboration)