A global fixed point proof for time-dependent density functional theory

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Density-potential mapping

The development of new functionals in TDDFT requires more insight into what the space-time non-localities in the time-dependent xc-potential actually look like, both in theory and numerically.

We are therefore studying:

1) A mathematically explicit and precise construction of the density to potential mapping
(M.Ruggenthaler, M.Penz)

2) Numerical solutions of exact time-dependent xc-potentials
(S.Nielsen, M.Ruggenthaler, M.Penz)

We here give a general outline of the proof.

Many technical points are discussed in detail on the poster:

Michael Ruggenthaler, RvL 'Global fixed point proof for TDDFT'

and in the papers:

M.Ruggenthaler, RvL, Europhysics Lett. 95, 13001 (2011)
Ruggenthaler, Penz, Bauer J.Phys.A. Math. Theor. 42, 425207 (2009)
Penz, Ruggenthaler, J. Phys.A. Math. Theor. 44, 335208 (2011)

From potentials to densities

Hamiltonian

$$\hat{H}(t) = \hat{T} + \hat{V}(t) + \hat{W}$$

Schrödinger equation

$$i\partial_t |\Psi[v](t)\rangle = \hat{H}(t)|\Psi[v](t)\rangle \qquad |\Psi[v](t_0)\rangle = |\Psi_0\rangle$$

Density

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

This maps from a certain domain of potentials to a certain domain of densities

Local force equation

equations of motion for the density and current operators (RG 1984)

$$\partial_t n(\mathbf{r}t) = -\nabla \cdot j(\mathbf{r}t)$$

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

local force

Combination of both then gives

$$\boxed{-\nabla \cdot (n([v], \mathbf{r}t)\nabla v(\mathbf{r}t)) = q([v], \mathbf{r}t) - \partial_t^2 n([v], \mathbf{r}t)}$$

where

$$q([v], \mathbf{r}t) = -i\nabla \cdot \langle\Psi(t)|\left[\hat{j}(\mathbf{r}), \hat{T} + \hat{W}\right]|\Psi(t)\rangle$$

Let us now replace n([v],rt) by a given density n(rt) subject to the conditions

$$n(rt_0) = \langle \Psi_0 | \hat{n}(\mathbf{r}) | \Psi_0 \rangle \qquad \partial_t n(rt_0) = -\langle \Psi_0 | \nabla \cdot \hat{j}(\mathbf{r}) | \Psi_0 \rangle$$

This is a nonlinear equation for v(rt)

$$\boxed{-\nabla \cdot (n(\mathbf{r}t)\nabla v(\mathbf{r}t)) = q([v], \mathbf{r}t) - \partial_t^2 n(\mathbf{r}t)}$$

If we propagate the TDSE with the solution v(rt) also have

$$-\nabla \cdot (n([v], \mathbf{r}t)\nabla v(\mathbf{r}t)) = q([v], \mathbf{r}t) - \partial_t^2 n([v], \mathbf{r}t)$$

Subtracting both equations we have

$$\boxed{\partial_t^2 \rho(\mathbf{r}, t) - \nabla \cdot (\rho(\mathbf{r}t)\nabla v(\mathbf{r}t)) \qquad \rho(\mathbf{r}t) = n([v], \mathbf{r}t) - n(\mathbf{r}t)}$$

with initial conditions

$$\rho(\mathbf{r}t_0) = 0 \qquad \partial_t \rho(\mathbf{r}t_0) = 0$$

The unique solution satisfying the initial conditions is $\rho(\mathbf{r}t) = 0$

$$n(\mathbf{r}t) = n([v], \mathbf{r}t)$$

If we now choose

$$n(\mathbf{r}t) = n_{W'}([u, \Phi_0], \mathbf{r}t)$$

to be the density obtained from the TDSE in a system with different interactions W' , external potential u(rt) and a different initial state Φ_0 then the existence of a solution to

$$\boxed{-\nabla \cdot (n(\mathbf{r}t)\nabla v(\mathbf{r}t)) = q([v], \mathbf{r}t) - \partial_t^2 n(\mathbf{r}t)}$$

implies

1) v-representability of n(rt) in our system

2) uniqueness for W=W' and $\Phi_0 = \Psi_0$ implies the Runge-Gross theorem

The main question therefore is:

Does a solution to

$$\boxed{-\nabla \cdot (n(\mathbf{r}t)\nabla v(\mathbf{r}t)) = q([v],\mathbf{r}t) - \partial_t^2 n(\mathbf{r}t)}$$

for v(rt) exist and is it unique ?

This can indeed be established when v(rt) and n(rt) can be expanded in a Taylor series around the initial time t_0 . However, there are indications that it is valid more generally. For instance, RG can be established without this assumption for

1) Linear response from the ground state (Laplace transformable v(rt))
2) External fields of dipole form (Ruggenthaler et al. , PRA (2010))
3) Lattice systems (Tokatly, PRB 83, 035127 (2011))

This suggests the possibility of a more general proof

Moreover there are examples of time-analytic potentials leading to non-time -analytic densities. (Maitra, Todorov, Woodward, Burke, PRA81, 042525 (2010))

It is therefore also desirable to find a proof that avoids the Taylor-expansion.

General idea:

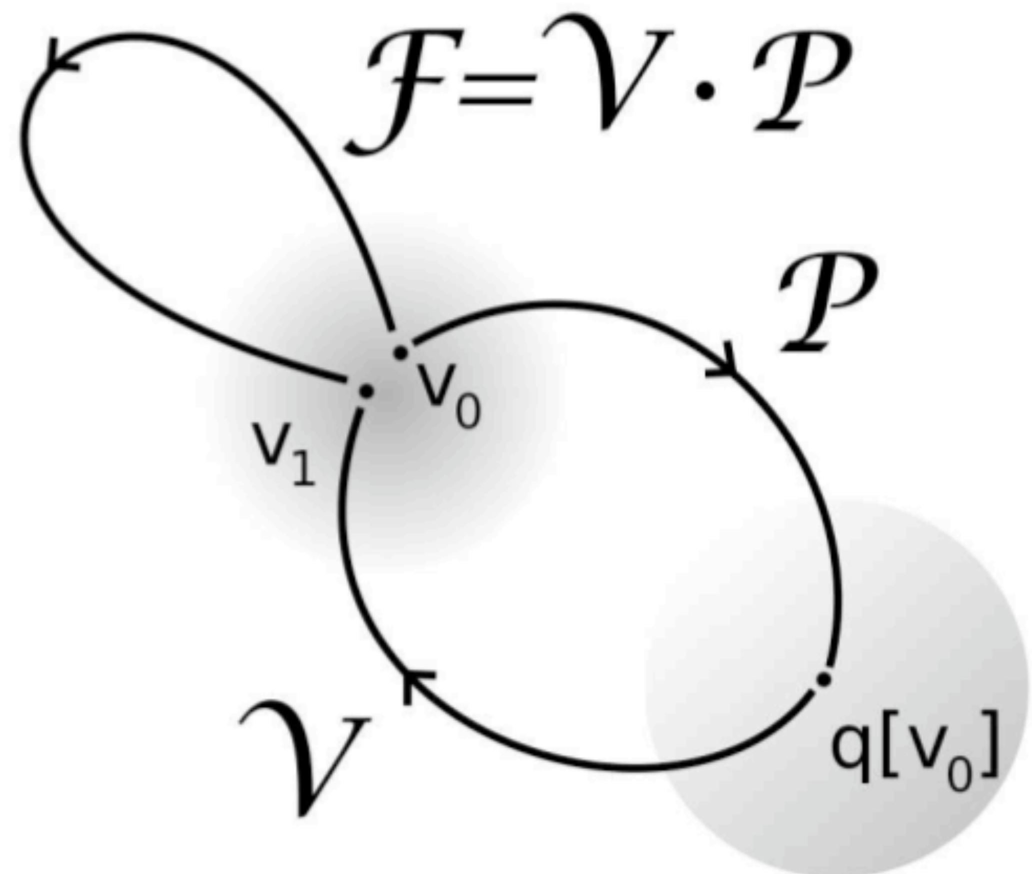
Rather than Taylor-expanding we consider a finite time-interval $[t_0, T]$ on which we find a global convergence scheme.
We then need to prove its convergence and uniqueness

(in the same spirit as the solution of the TDOEP equations by Wijewardane and Ullrich, PRL100, 056404 (2008))

M.Ruggenthaler, RvL,
Europhysics Lett. 95, 13001 (2011)

From time-propagation we have

$$\mathcal{P} : v_0 \mapsto q[v_0]$$



We then solve

$$-\nabla \cdot (n(\mathbf{r}t)\nabla v_1(\mathbf{r}t)) = q([v_0], \mathbf{r}t) - \partial_t^2 n(\mathbf{r}t)$$

for $v_1(rt)$. This yields a mapping

$$\mathcal{V} : q[v_0] \mapsto v_1$$

The combined mapping

$$\boxed{\mathcal{F}[v_0] = (\mathcal{V} \circ \mathcal{P})[v_0] = v_1}$$

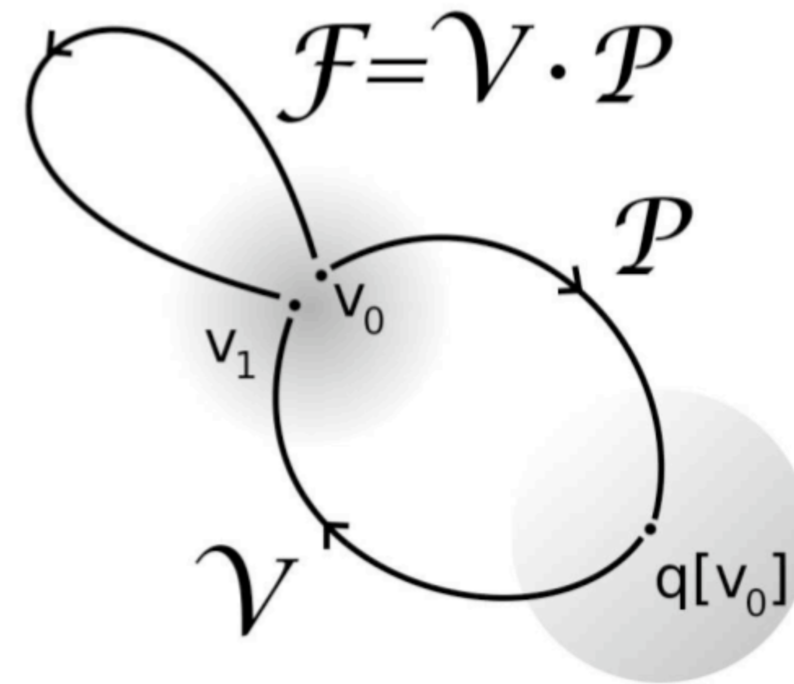
maps potentials to potentials

Whenever we have

$$\mathcal{F}[v] = v$$

then we are solving

$$\boxed{-\nabla \cdot (n(\mathbf{r}t)\nabla v(\mathbf{r}t)) = q([v], \mathbf{r}t) - \partial_t^2 n(\mathbf{r}t)}$$



The question whether a solution to this equation exists and is unique is thus equivalent to the question whether a unique fixed point of the mapping F exists.

The main existence and uniqueness question of TDDFT is in this way reformulated as a fixed point question

What we will show is the following :

$$\boxed{\|\mathcal{F}[v_1] - \mathcal{F}[v_0]\|_\alpha \le a\|v_1 - v_0\|_\alpha \qquad a < 1}$$

for some parameter-dependent norm on the space of potentials.
This follows from the two inequalities

$$\|q[v_1] - q[v_0]\|_\alpha \le \frac{C}{\sqrt{\alpha}}\|v_1 - v_0\|_\alpha$$

$$\|\mathcal{F}[v_1] - \mathcal{F}[v_0]\|_\alpha \le D\|q[v_1] - q[v_0]\|_\alpha$$

where

$$a = \frac{CD}{\sqrt{\alpha}}$$

and we can choose $\sqrt{\alpha} > CD$

Outline of the proof

From

$$O[v_1] - O[v_0] = \int_0^1 d\lambda\, \frac{dO}{d\xi}[v_0 + \xi(v_1 - v_0)]|_{\xi=\lambda}$$

with $\hat{O} = \hat{q}(\mathbf{r})$ we have

$$q([v_1], \mathbf{r}t) - q([v_0], \mathbf{r}t) = \int_{t_0}^{t} dt' \int_\Omega d\mathbf{r}' \chi(\mathbf{r}t, \mathbf{r}'t')(v_1(\mathbf{r}'t') - v_0(\mathbf{r}'t'))$$

$$\chi(\mathbf{r}t, \mathbf{r}'t') = -i \int_0^1 d\lambda \langle \Psi_0 | [\hat{q}_{H_\lambda}(\mathbf{r}t), \hat{n}_{H_\lambda}(\mathbf{r}'t')] | \Psi_0 \rangle$$

Nonequilibrium response function

where $\hat{H}_\lambda$ is the hamiltonian with potential

$$v_\lambda = v_0 + \lambda(v_1 - v_0)$$

For a general integral equation of the form

$$f(\mathbf{r}t) = \int_{t_0}^{t} dt' \int_{\Omega} d\mathbf{r}' \chi(\mathbf{r}t, \mathbf{r}'t') g(\mathbf{r}'t')$$

we have

$$\|f(t)\|^2 \leq C^2(t) \int_{t_0}^{t} dt' \|g(t')\|^2$$

where

$$\|f(t)\|^2 = \int_{\Omega} d\mathbf{r}\, f(\mathbf{r}t)^2$$

$$C^2(t) = \sup_{g \neq 0} \frac{\|(\chi g)(t)\|^2}{\int_{t_0}^{t} dt' \|g(t')\|^2}$$

Then it is readily shown that:

$$\|f\|_\alpha \leq \frac{C}{\sqrt{\alpha}} \|g\|_\alpha$$

where

$$\|f\|_\alpha^2 = \sup_{t\in[t_0,T]} \left(\|f(t)\|^2 e^{-\alpha(t-t_0)} \right)$$

$$C^2 = \sup_{t\in[t_0,T]} C^2(t)$$

α-norm, where α can be chosen freely

Applying this to our case gives

$$\boxed{\|q[v_1] - q[v_0]\|_\alpha \leq \frac{C}{\sqrt{\alpha}} \|v_1 - v_0\|_\alpha}$$

Let us now consider the second inequality

$$\|\mathcal{F}[v_1] - \mathcal{F}[v_0]\|_\alpha \leq D\|q[v_1] - q[v_0]\|_\alpha$$

If we define $v_1 = \mathcal{F}[v_0]$ and $v_2 = \mathcal{F}[v_1]$ then

$$\boxed{-\nabla \cdot [n(\mathbf{r}t)\nabla(v_2(\mathbf{r}t) - v_1(\mathbf{r}t))] = q([v_1], \mathbf{r}t) - q([v_0], \mathbf{r}t)}$$

This is an equation of the Sturm-Liouville form

$$Qv(\mathbf{r}t) = \zeta(\mathbf{r}t)$$

$$Q = -\nabla \cdot [n(\mathbf{r}t)\nabla]$$

The mathematical properties of this equation have been studied rigorously in Ruggenthaler, Penz, Bauer J.Phys.A. Math. Theor. 42, 425207 (2009)
Penz, Ruggenthaler, J. Phys.A. Math. Theor. 44, 335208 (2011)

Since

$$\langle v_0|Qv_1\rangle - \langle Qv_0|v_1\rangle = \int_{\partial\Omega} dS \cdot n(\mathbf{r}t)\,(v_1(\mathbf{r}t)\nabla v_0(\mathbf{r}t) - v_0(\mathbf{r}t)\nabla v_1(\mathbf{r}t))$$

the Sturm-Liouville operator is self-adjoint whenever either the density or the potential vanishes at the boundaries. In that case there is an orthonormal eigenbasis

$$Q\phi_i(\mathbf{r}t) = \lambda_i(t)\phi_i(\mathbf{r}t)$$

where

$$\lambda_i(t) = \langle \phi_i|Q\phi_i\rangle = \int_{\Omega} d\mathbf{r}\, n(\mathbf{r}t)|\nabla\phi_i(\mathbf{r}t)|^2 \geq 0$$

and the only zero eigenvalue corresponds to the constant function

$$\phi_0(\mathbf{r}t) = c(t) \qquad\qquad 0 = \lambda_0(t) < \lambda_1(t) \leq \lambda_2(t) \leq \ldots$$

Expanding

$$(v_1 - v_2)(\mathbf{r}t) = \sum_{i=1}^{\infty} u_i(t)\phi_i(\mathbf{r}t)$$

$$(q[v_1] - q[v_0])(\mathbf{r}t) = \sum_{i=1}^{\infty} \zeta_i(t)\phi_i(\mathbf{r}t)$$

we have

$$\|v_1(t) - v_2(t)\|^2 = \sum_{i=1}^{\infty} |\frac{\zeta_i(t)}{\lambda_i(t)}|^2 \leq \frac{1}{\lambda_1(t)^2} \sum_{i=1}^{\infty} |\zeta_i(t)|^2$$

$$= \frac{1}{\lambda_1(t)^2} \|q[v_1](t) - q[v_0](t)\|^2$$

It follows that

$$\|\mathcal{F}[v_1] - \mathcal{F}[v_0]\|_\alpha = \|v_2 - v_1\|_\alpha \leq D\|q[v_1] - q[v_0]\|_\alpha$$

$$D^2 = \sup_{t\in[t_0,T]} \frac{1}{\lambda_1(t)^2}$$

as we wanted to prove

Consequences

$$\|\mathcal{F}[u] - \mathcal{F}[v]\|_\alpha \le a\|u - v\|_\alpha \qquad\qquad a = \frac{CD}{\sqrt{\alpha}}$$

Suppose now that there are 2 fixed points

$$\mathcal{F}[u] = u \qquad\qquad \mathcal{F}[v] = v$$

then choosing $\sqrt{\alpha} = 2CD$ we find

$$\|u - v\|_\alpha = \|\mathcal{F}[u] - \mathcal{F}[v]\|_\alpha \le \frac{1}{2}\|u - v\|_\alpha$$

and therefore

$$\|u - v\|_\alpha = 0 \quad \Longrightarrow \quad u = v$$

This amounts to the RG theorem: There is a unique potential producing a given density for a given initial state

We established uniqueness, but what about existence (v-representability) ?

If $C_{\text{sup}} = \sup_v C[v, \mathcal{F}[v]]$ exists then by choosing $\sqrt{\alpha} > C_{\text{sup}} D$ we have

$$\|v_{k+1} - v_k\|_\alpha \leq a^k \|v_1 - v_0\| \qquad a = \frac{C_{\text{sup}} D}{\sqrt{\alpha}}$$

$$v_k = \mathcal{F}^k[v_0]$$

We have a Cauchy sequence in a complete space and therefore

$$v_k \longrightarrow v$$

Existence therefore requires a condition on the operator norm of the response function

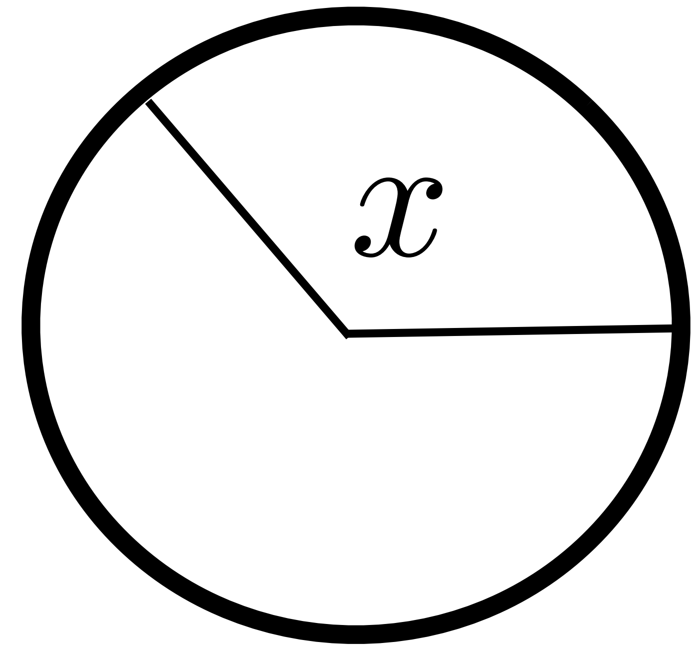
Summary of mathematical details

The potentials that we consider are in $L^2(\Omega)$ which for example includes the Coulomb potential

The local forces that we consider are in the dual to a certain weighted Sobolev space $H_0^1(\Omega, n)$

Numerical example

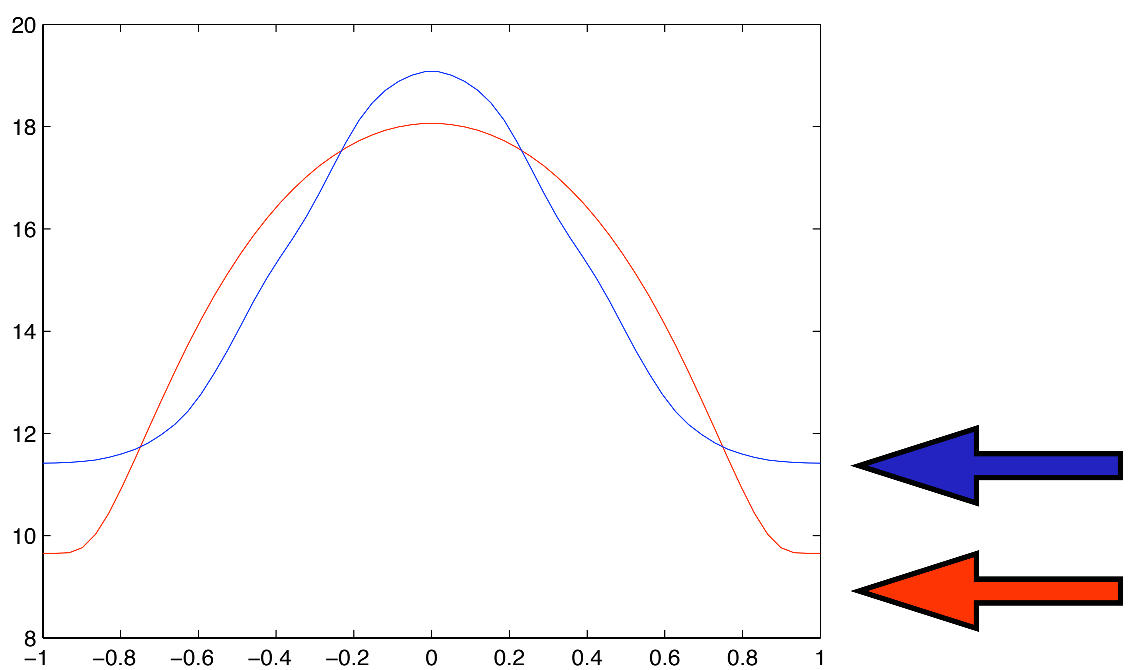
Particle on a ring of circumference 1.
We apply a potential



$$v(x,t) = \sin^2(\pi x)\sin(\omega t)$$

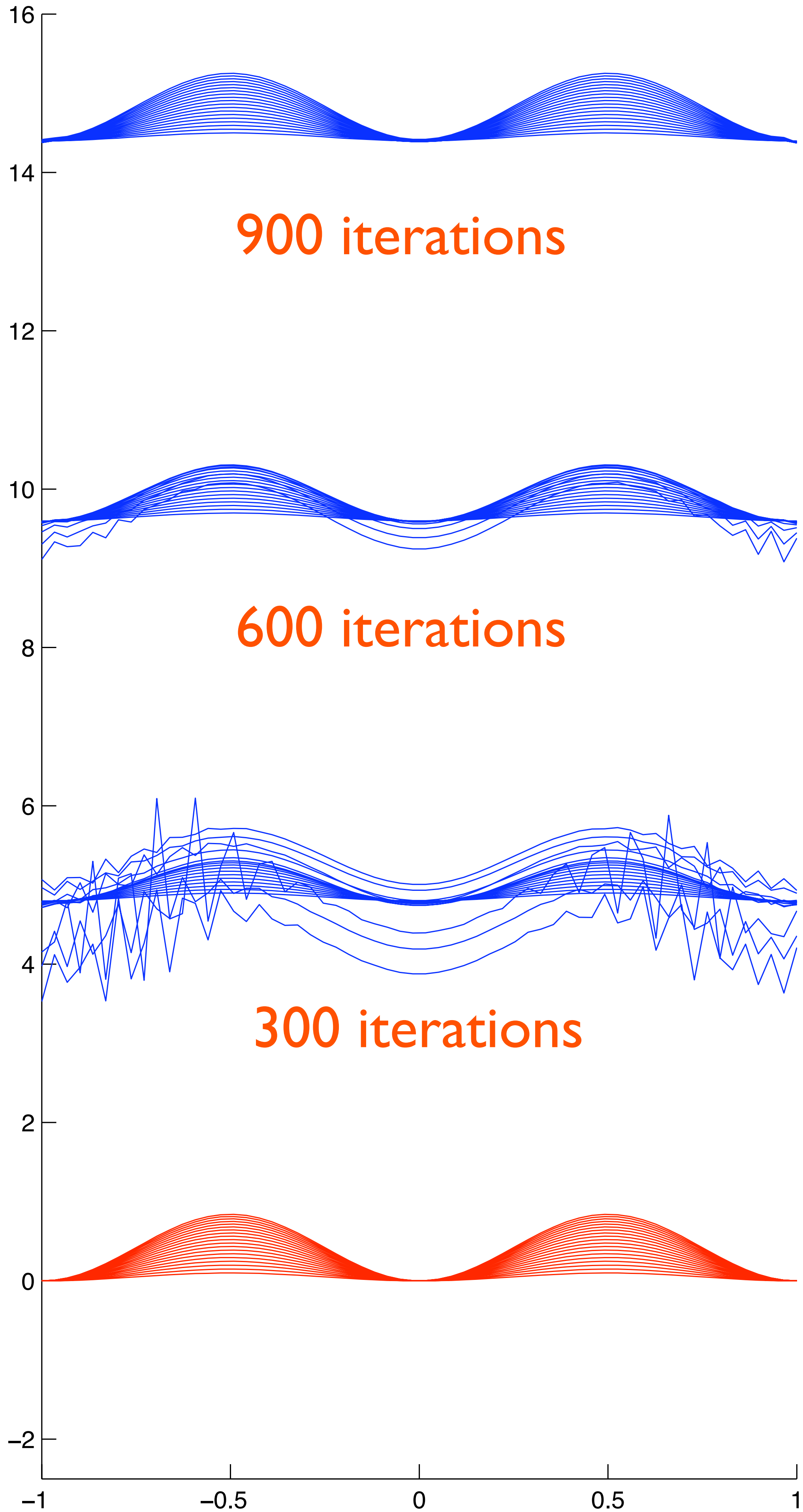
with initial state

$$\Psi_0(x) = A\left(e^{-\frac{1}{(1-x^2)}} + 1\right) \qquad x \in [-1, 1]$$



final density

initial density



Then we feed the density n(x,t) in the fixed point procedure and recover the potential v(x,t) (red one in the picture)

We start from the initial guess v(x,t)=0

The potential converges nicely to the exact v(x,t)

Exact v(x,t) (snapshots)

Further numerics

In collaboration with Søren Nielsen, Jeppe Olsen from Århus, Denmark

We are presently investigate the dynamics of xc-potentials for 2-particle systems (soft-Coulomb) with various initial states to study:

- Memory effects in the xc-potential

- Initial state dependence
  (e.g. correlated initial states in the KS system)

- Quantum control of interacting system
  (find v(x,t) for prescribed n(x,t))

Conclusions

- The existence and uniqueness question of TDDFT are reformulated as a fixed point problem

- The Taylor expansion in time-derivatives is avoided. Instead we demand the existence of the operator norm of a certain response function

- The Volterra structure of the response function is exploited by use of the α-norm

- Numerical work (Søren Nielsen, Michael Ruggenthaler) on 1-d correlated systems has already shown that the fixed point scheme converges very well (work in progress)

Now we can do the following trick

$$\|f(t)\|^2 \leq C^2(t) \int_{t_0}^{t} dt' e^{\alpha(t'-t_0)} e^{-\alpha(t'-t_0)} \|g(t')\|^2$$

$$\leq C^2(t) \sup_{t' \in [t_0, t]} \|g(t')\|^2 e^{-\alpha(t'-t_0)} \underbrace{\int_{t_0}^{t} dt' e^{\alpha(t'-t_0)}}_{\leq \frac{e^{\alpha(t-t_0)}}{\alpha}}$$

and find

$$\boxed{e^{-\alpha(t-t_0)} \|f(t)\|^2 \leq \frac{C^2(t)}{\alpha} \sup_{t' \in [t_0, t]} e^{-\alpha(t-t_0)} \|g(t')\|^2}$$