

Can quantum dynamics be described by the density alone?

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Density Functional Theory proposes methods to calculate, or approximate, the one-particle density of an N -particle quantum system directly – without first calculating the full N -body wave-function. Such methods are popular computational tools, due to their moderate cost. This leads to the question how much information about the full system is contained in the one-body density. More specifically, one may ask which quantities in the full system may be reconstructed from the density, given some a priori information, such as what kind of particles constitute the system. We will address this question here for a time-dependent problem.

We assume that the quantum system in question consists of N fermions or bosons, and that its time-dependent Hamiltonian, defined on $L^2_{\text{a}}(\mathbb{R}^{dN})$ or $L^2_{\text{s}}(\mathbb{R}^{dN})$, is of the form

$$H_V(t) := H_0 + \sum_{i=1}^N V(x_i, t),$$

with

$$H_0 = \sum_{i=1}^N -\Delta_{x_i} + V_0(x_i) + \sum_{1 \leq i < j \leq N} W(x_i - x_j).$$

The conditions on the potentials V , V_0 , W on \mathbb{R}^d will be discussed in detail later. We consider V_0 and W as quantities which are a priori known. If, for example, the particles in our system are electrons they will interact via Coulomb forces $W(x) = 1/|x|$.

In the time-independent case of ground-states, it is known that for $V, V_0, W \in L^{d/2}(\mathbb{R}^d) + L^\infty(\mathbb{R}^d)$ (for $d \geq 3$) the potential V is uniquely determined by the ground state density

$$\rho(x) = N \int |\psi(x, y_1, \dots, y_{N-1})|^2 dy_1 \cdots dy_{N-1}.$$

That is, if $V_1, V_2 \in L^{d/2}(\mathbb{R}^d) + L^\infty(\mathbb{R}^d)$ differ by more than a constant and have ground states ψ_1, ψ_2 , then the corresponding one-particle densities ρ_1 and ρ_2 are different. Equivalently, if $\rho_1 = \rho_2$, then $V_1 = V_2 + \text{const.}$. This statement is known as the Hohenberg-Kohn Theorem [HK64], see Lieb [Li83] for a proof.

Runge and Gross [RG84] have argued that a similar property should hold for time dependent systems. That is, the time-dependent density $\rho(x, t)$ should determine the external potential $V(x, t)$ up to a constant $C(t)$ if the system starts with a given initial wave-function ψ_0 . The argument is based on an order-by order analysis of the Taylor series in time of the density $\rho(x, t)$ obtained from the solution $\psi(t)$ of the Schrödinger equation with initial condition ψ_0 and time-dependent Hamiltonian H_V . Such an expansion clearly relies on smoothness of ρ and V w.r.t. the time-variable, which cannot always be guaranteed. This has recently led

to a discussion in the physical-chemistry literature regarding the validity of the argument, see e.g. [YB13].

The first mathematical work concerning this question is our article [FLLS16], where we discuss the possibility of choosing a set \mathcal{I} of admissible initial conditions and \mathcal{V} of external potentials for which the statement above can be proven rigorously, using an argument similar to that of Runge and Gross. In order to avoid pathologies, these sets should satisfy the following conditions:

- The set \mathcal{I} is invariant under the dynamics generated by H_V for any $V \in \mathcal{V}$.
- $0 \in \mathcal{V}$ and if $V(x, t)$, $t \in [0, T)$ is an admissible potential, then so is the time-independent potential $V(x, t_0)$ for any $t_0 \in [0, T)$.
- If the time-independent potential $V(x) \in \mathcal{V}$, then any eigenvectors of H_V are admissible initial conditions.

In order to have smooth solutions, we also need to restrict the set of initial conditions

$$\mathcal{I} \subset \bigcap_{V(x) \in \mathcal{V}} C^\infty(H_V),$$

where the intersection is over all time-independent potentials and $C^\infty(H_V) := \bigcap_{k \in \mathbb{N}} D(H_V^k)$ denotes the set of H_V -smooth vectors. Now, depending on the set \mathcal{V} , the intersection above may be very small – and will in general not be invariant under the dynamics of the operators H_V . In order to avoid this we have to restrict \mathcal{V} so that

$$(1) \quad C^\infty(H_V) = C^\infty(H_0)$$

for every $V \in \mathcal{V}$. This condition clearly shows that the possible choice of \mathcal{V} depends strongly on H_0 – and thus on V_0 and W .

1. SMOOTH POTENTIALS

If the potentials V_0 and W are smooth, we have natural choices for \mathcal{I} and \mathcal{V} and the Runge-Gross argument becomes a rigorous theorem. To be more precise, let $V_0, W \in C_b^\infty(\mathbb{R}^d, \mathbb{R})$, W even, and set

$$\begin{aligned} \mathcal{I} &= \bigcap_{k \in \mathbb{N}} H^{2k}(\mathbb{R}^{dN}) \cap L_{a/s}^2(\mathbb{R}^{dN}) \\ \mathcal{V} &= C_b^\infty([0, T) \times \mathbb{R}^d, \mathbb{R}) \end{aligned}$$

for some $T > 0$. We then have:

Theorem 1. *Let $V_1, V_2 \in \mathcal{V}$ and $\psi_0 \in \mathcal{I}$ with one-particle density ρ_0 . Denote by $\psi_k(t)$, $k \in 1, 2$ the solution at time $t \in [0, T)$ of the Schrödinger equation with Hamiltonian H_{V_k} and initial condition $\psi_k(0) = \psi_0$. Denote by ρ_k the corresponding one-particle density. If $\rho_1 = \rho_2$, then for all $\ell \in \mathbb{N}$*

$$(2) \quad \int_{\mathbb{R}^d} \rho_0(x) |\nabla \partial_t^\ell (V_1 - V_2)|^2(x, 0) dx = 0.$$

If additionally the set $\rho_0^{-1}(0)$ has zero Lebesgue measure, then $\partial_t^\ell V_1(x) = \partial_t^\ell V_2(x) + c_\ell$ for some constant c_ℓ . If furthermore $(V_1 - V_2)(x, t)$ is real-analytic in t for every x we also have $V_1(x, t) = V_2(x, t) + C(t)$, with $C(t) = \sum_{\ell \in \mathbb{N}} \frac{c_\ell}{\ell!} t^\ell$.

For the proof of this theorem, one first applies a result of Kato [Ka53] to show that $\psi_k(t) \in \mathcal{I}$ depends smoothly on time. Then Equation (2) is obtained recursively by calculating weak time-derivatives of $\rho_1 - \rho_2$ at $t = 0$, which must equal zero since $\rho_1 = \rho_2$. For instance, the weak second derivative yields

$$0 = \frac{d^2}{dt^2} \Big|_{t=0} \int_{\mathbb{R}^d} \varphi(x) (\rho_1 - \rho_2)(x, t) dx = 2N \int (\nabla \varphi)(x) \nabla (V_2 - V_1)(x, 0) dx,$$

so choosing the test-function $\varphi = V_2 - V_1$ gives (2) for $\ell = 0$. The additional statements follow easily from (2).

2. SINGULAR POTENTIALS

If V_0 or W are not smooth, the condition (1) will lead to strong restrictions on the set \mathcal{V} . For example, it is easy to see that if H_0 is the one-dimensional Schrödinger operator with a delta-potential at $x = 0$, any potential satisfying (1) must be smooth on \mathbb{R} and vanish to infinite order at $x = 0$. We expect this statement to hold generically for non-smooth potentials V_0 . The restrictions on \mathcal{V} are far more severe for singular interactions. We have the following:

Proposition 1. *Let $d = 3$, $N = 2$, $V_0 = 0$, $W(x) = \frac{1}{|x|}$ be the Coulomb-interaction and H_0 the operator acting on symmetric functions given by these choices. If $V \in C_b^6(\mathbb{R}^3)$ satisfies $D(H_V^4) = D(H_0^4)$ then V is constant.*

To prove this, one separates the relative and centre-of-mass coordinates and then applies similar arguments as in the case of a singular one-body potential V_0 . This yields $\Delta V = 0$ and thus proves that V is constant.

We thus see that for the Coulomb-interaction Equation (1) already implies that V is constant, and there is nothing of interest to prove afterwards. Equation (2) can still be shown to hold for $\ell \leq 3$ ($\ell \leq 4$ for fermions), under reasonable assumptions. In order to obtain more information, a new approach that avoids Taylor expansions of high order is clearly necessary.

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