Runge-Gross action-integral functional re-examined

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Abstract

The density-based action-integral functional introduced by Runge and Gross [Phys. Rev. Lett. 52, 997(1984)] in their foundation of time-dependent density-functional theory (TDDFT) is re-examined. Based on an obvious expansion of the original definition, it becomes apparent that the action-integral functional is both trivial and non-stationary. It cannot be used to establish equations of motion for the time-evolution of quantum systems at the density-function level.
In the following, we briefly address the action-integral functional (AIF) introduced by Runge and Gross (RG) in their attempt to establish time-dependent density-functional theory (TDDFT) [1]. The RG-AIF has been criticized as lacking definitness due to a purely time-dependent phase function entering the definition of the density-based AIF [2, 3]. However, the phase problem may not be the main issue here. The AIF can readily be written in a more explicit form showing that it is not stationary and does not establish an equation of motion at the density level. While the finding discussed below is entirely obvious, it seems to have escaped due attention previously.

Let us consider an \( N \)-electron system subject to a time-dependent one-particle potential, the Hamiltonian being of the form

\[
\hat{H}(t) = \hat{T} + \hat{V} + \hat{U}(t)
\]  

where \( \hat{T} \) and \( \hat{V} \) are the kinetic energy operator and Coulomb repulsion operator, respectively, while

\[
\hat{U}(t) = \sum_i^N u(r_i, t)
\]

denotes the time-dependent local one-body potential operator, comprising a static and a time-dependent part according to \( \hat{U}(t) = \hat{W} + \hat{F}(t) \). For definitness, we will suppose that the time-dependent “external” potential sets in at \( t = 0 \), that is, \( \hat{F}(t) = 0 \) for \( t < 0 \), and the system is in the ground state \( \Psi_0 \) of the time-independent Hamiltonian \( \hat{H} = \hat{T} + \hat{V} + \hat{W} \) at \( t = 0 \). The solution \( \Psi(t) \) of the time-dependent Schrödinger equation (TDSE) for the initial value \( \Psi(0) = \Psi_0 \) gives rise to the time-dependent density function \( n_0(r, t) \). TDDFT claims that it is possible to determine \( n_0(r, t) \) without recourse to the TDSE.

The basic entity in the original RG foundation of TDDFT is the density-based AIF for the system under consideration, given by

\[
A[n] = \int_{t_1}^{t_2} dt \langle \Psi[n](t)|i \frac{\partial}{\partial t} - \hat{H}(t)|\Psi[n](t)\rangle
\]  

Here \( \Psi[n](t) \) is the wave function associated with the time-dependent density function \( n(r, t) \) according to the first Runge-Gross (RG1) theorem [1]. Let us recall that the RG1 theorem establishes a mapping between time-dependent densities, \( n(r, t) \), and time-dependent “external” potentials, \( v_{\text{ext}}[n](r, t) \),

\[
n(r, t) \rightarrow v_{\text{ext}}[n](r, t) + c(t)
\]
such that the solution $\Psi[n](t)$ of the $N$-electron TDSE

$$i \frac{\partial}{\partial t} \Psi[n](t) = \{ \hat{T} + \hat{V} + \hat{V}_{ext}[n](t) + C(t) \} \Psi[n](t)$$

(5)

reproduces the respective density $n(r, t)$. Here $\hat{V}_{ext}[n](t)$ is the $N$-electron form of the external potential, that is,

$$\hat{V}_{ext}[n](t) = \sum_{i} v_{ext}[n](r_i, t)$$

(6)

Note that $v_{ext}[n](r, t)$ is determined by the density only up to a time-dependent function $c(t)$, that is, $C(t) = Nc(t)$ in Eq. (5). To specify the initial value problem, we may consider densities where $n(r, 0) = n_0(r)$, and, moreover, suppose $\Psi[n](0) = \Psi_0$.

Using that $\Psi[n](t)$ fulfills the TDSE (5), the RG-AIF according to Eq. (3) can be written in a simple form with an explicit Lagrange-type function,

$$A[n] = \int_{t_1}^{t_2} dt \int n(r, t) d\mathbf{r} \left\{ v_{ext}[n](r, t) - u(r, t) \right\} + \int_{t_1}^{t_2} C(t) dt$$

(7)

Here, the kinetic and Coulomb energy contributions have cancelled along with the time derivative, and the remaining potential energy expectation values can be expressed entirely in terms of the density functions $n(r, t)$, since both $\hat{V}_{ext}[n](t)$ and $\hat{U}(t)$ are local one-particle operators. This shows that the AIF can be defined directly at the level of the density functions, and its relation to the solution of TDSE, as implied by Eqs. (3, 5), is an illusion.

Let us note that the indefiniteness of the AIF due to the $\int C(t) dt$ term has been addressed previously (see Refs. [2–4]), and we shall ignore it in the present context by supposing $C(t) = 0$ in Eq. (7).

Obviously, $A[n]$ vanishes for $n(t) = n_0(t)$ since

$$v_{ext}[n_0](r, t) = u(r, t)$$

(8)

which simply reflects the construction underlying $v_{ext}[n](r, t)$. Eq. (8) is not an equation of motion, nor can it be seen as a realistic means to determine $n_0(t)$. An eventual solution would require guessing the potential-functional $v_{ext}[n](r, t)$ (or an approximation to it) and solving the implicit equation (8), possibly by adopting a fixed-point iteration scheme.

Even more disturbing is the observation that $A[n]$ is not stationary at $n_0(t)$. This can be seen by evaluating the variation according to Eq. (7) and using Eq. (8):

$$\delta A[n]|_{n_0} = \int_{t_1}^{t_2} dt \int d\mathbf{r} n_0(r, t) \delta v_{ext}[n](r, t)|_{n_0}$$

(9)
Here one cannot expect that $\delta v_{\text{ext}}[n](\mathbf{r}, t)|_{n_0}$ vanishes, since $n_0(\mathbf{r}, t)$ is just an ordinary density-function argument for the external potential-functional, not distinguished from other densities.

Assuming for simplicity that $v_{\text{ext}}[n](\mathbf{r}, t)$ will depend only on $n(\mathbf{r}, t)$ (and not on the first and higher time derivatives of $n(\mathbf{r}, t)$), the functional derivative of $A[n]$ takes on the form

$$\frac{\delta A[n]}{\delta n(\mathbf{r}, t)} = v_{\text{ext}}[n](\mathbf{r}, t) - u(\mathbf{r}, t) + n(\mathbf{r}, t) \frac{\partial v_{\text{ext}}[n]}{\partial n(\mathbf{r}, t)}$$

(10)

which shows, according to

$$\left. \frac{\delta A[n]}{\delta n(\mathbf{r}, t)} \right|_{n_0} = n_0(\mathbf{r}, t) \left. \frac{\partial v_{\text{ext}}[n]}{\partial n(\mathbf{r}, t)} \right|_{n_0} \neq 0$$

(11)

that the functional derivative does not vanish at the desired density $n_0(t)$. This means that Eq. (11) in Ref. [1] is patently wrong.

In conclusion, the RG-AIF can readily be written in a more explicit form, which makes apparent that

(i) it is an essentially trivial construct that does not establish an equation of motion for the time-evolution of a quantum system at the density level;

(ii) it is not stationary for the density of the system under consideration.

It should be noted that the original RG foundation of TDDFT, based on the stationarity of the RG-AIF, was abandoned by its principal architects some time ago, notwithstanding occasional attempts at a rehabilitation (see, e.g. Ref. [5]). The alternative offered to establish time-dependent Kohn-Sham (TDKS) equations is based entirely on the RG1 mapping theorem (see Ref. [6]). Unfortunately, the mapping foundation has not been fully disclosed in the TDDFT literature so far. For a discussion of the problems arising here, the reader is referred to Refs. [3, 7].

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