Time-dependent Internal DFT formalism and Kohn-Sham scheme.

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We generalize to the time-dependent case the stationary Internal DFT / Kohn-Sham formalism presented in Ref. [14]. We prove that, in the time-dependent case, the internal properties of a self-bound system (as an atomic nuclei or a Helium droplet) are all defined by the internal one-body density and the initial state. We set-up a time-dependent Internal Kohn-Sham scheme as a practical way to compute the internal density. The main difference with the traditional DFT / Kohn-Sham formalism is the inclusion of the center-of-mass correlations in the functional.

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I. INTRODUCTION.

Traditional Density Functional Theory (DFT) [1, 2, 3] and its time-dependent generalization [4, 5] have evolved into standard tools for the description of electronic properties in condensed-matter physics and quantum chemistry through the simple local density instead of the less tractable N-body wave function. Stationary DFT is based on the Hohenberg-Kohn (HK) theorem [6], which proves that, for any non-degenerate system of N Fermions or Bosons [7], put into a local external potential, the N-body ground-state wave function can be written as a functional of the local ground-state density. A similar theorem exists for the time-dependent case [4, 5], where a dependence on the initial state appears. The Kohn-Sham (KS) scheme [6] and its time-dependent generalization [4, 5] provide a straightforward method to compute self-consistently the density in a quantum framework, defining the non-interacting system (i.e. the local single-particle potential) which reproduces the exact density.

Traditional DFT is particularly well suited to study the electronic properties in molecules [8]. As a molecule is a self-bound system, the corresponding Hamiltonian is translationally invariant (which ensures Galilean invariance of the wave function [8]), and one can apply the Jacobi coordinates method. This permits to decouple the center-of-mass (c.m.) properties from the internal ones, and to treat correctly the redundant coordinate problem (i.e. the fact that one coordinate is redundant for the description of the internal properties [10]) and the c.m. correlations. But as the nuclei are much heavier than the electrons, we can apply the Jacobi coordinates method to the nuclei only, so that only the nuclei will carry the c.m. correlations, and use the clamped nuclei approximation. Then, one recovers the “external” potential of traditional DFT, of the form \( \sum_{i=1}^{N} v_{\text{ext}}(r_{i}) \), which accounts for the nuclear background as seen by the electrons in the frame attached to the c.m. of the nuclei. Thus, traditional DFT is particularly adapted to the study of the electronic properties in molecules [8]. It is implicitly formulated in the nuclear c.m. frame [11] and the energy functional does not contain any c.m. correlations. Of course, contrary to the whole molecule, the pure electronic system is not a self-bound system: the \( v_{\text{ext}} \) potential breaks translational invariance and is compulsory in order to reach bound states in the stationary case.

For other self-bound systems, as isolated atomic nuclei or He droplets, the situation is intrinsically different because the masses of all the particles (Fermions or Bosons) are of the same order of magnitude. As a consequence, to decouple the c.m. properties from the internal ones, one has to apply the Jacobi coordinates method to all the particles. The redundant coordinate problem (thus the c.m. correlations) will now concern all the particles and should be treated properly. If a DFT exists, the c.m. correlations should be taken into account in the functional.

Moreover, no ”external” potential of the form \( \sum_{i=1}^{N} v_{\text{ext}}(r_{i}) \) can be justified in the corresponding self-bound Hamiltonians (we denote \( r_{i} \) the \( N \) particles coordinates related to any inertial frame as the laboratory). One may be tempted to formulate a DFT using the traditional DFT conclusions in the limit \( v_{\text{ext}} \rightarrow 0 \), but this would lead to false and incoherent results because:

- in the stationary case, the Hohenberg-Kohn theorem is valid only for external potentials that lead to bound many-body states [13], which is not the case anymore at the limit \( v_{\text{ext}} \rightarrow 0 \) for translational invariant particle-particle interactions [4]:

- the form of \( v_{\text{ext}} \) is not translationally invariant, but translational invariance is a key feature of self-bound systems [10, 14, 15];

- traditional DFT concepts as formulated so far are not applicable in terms of a well-defined internal density \( \rho_{\text{int}} \), i.e. the density relative to the system’s c.m. , which is of experimental interest [8, 14, 16] (it is for example measured in nuclear scattering experiments).

Instead of the traditional DFT potential \( \sum_{i=1}^{N} v_{\text{ext}}(r_{i}) \), one might be tempted to introduce an arbitrary translational invariant potential of the form \( \sum_{i=1}^{N} v_{\text{int}}(r_{i} - R) \), where \( R = \frac{1}{N} \sum_{j=1}^{N} r_{j} \) is the total c.m. of the particles.
This potential is an "internal" potential, i.e. seen in the c.m. frame, and in [14] we underlined that it is the only form which satisfies all the key formal properties. However, \( v_{\text{int}} \) should be zero in the purely isolated self-bound case. This is why in [14] we presented it as a mathematical "auxiliary" to reach our goal and showed that it can be dropped properly at the end, conserving all the conclusions. Through it (and using the Jacobi coordinates), we proved, by a different way than those found in [16, 17], the stationary "Internal DFT" theorem: the internal many-body state can be written as a functional of \( \rho_{\text{int}} \). Then we formulated rigorously the corresponding "Internal" KS scheme (in the c.m. frame).

The main interest of this work is to give a first step towards a fundamental justification to the use of internal density functionals for stationary mean-field like calculations of nuclei [18] or He droplets [19] with effective interactions, showing that there exists an ultimate functional which permits to reproduce the exact internal density, which was not clear up to now.

It is to be noted that the stationary Internal DFT / KS formalism gives a more fundamental justification than the Hartree-Fock (HF) framework to the stationary nuclear mean-field like calculations. Indeed, HF does not contain quantum correlations, nor treats correctly the redundant coordinate problem, which introduces a spurious coupling between the internal properties and the c.m. motion [10, 21]. A way to overcome this problem in the stationary case is to perform projected HF (projection before variation on c.m. momentum), which permits to restore Galilean invariance, but at the price of abandoning the independent-particle description [10, 17, 18]. Within the Internal DFT / KS formalism, we proved that the c.m. correlations can be included in the energy functional / the KS potential [14], so that there would be no need for a c.m. projection if the ultimate functional was known.

The interest (experimentalists always deduce those properties) takes a true meaning. This is because self-bound systems are plagued by a c.m. problem. For instance, in the stationary case, the c.m. will be delocalized in the whole space for isolated self-bound systems [8, 14, 16]. This does not occur in experiments because experimentally observed self-bound systems are not isolated anymore (they interact with the piece of matter they are inserted in which localizes the c.m.). In the time domain, the c.m. motion remains uncomparable to the experimental one (this will be discussed in more detail later), so that it would not make sense to introduce a time-dependent potential which would act on the c.m. motion. It are the internal properties which are of true experimental interest (experimentalists always deduce those properties). This justifies the introduction of an explicitly time-dependent potential of the form [14], which would act on the internal properties only, and models the internal effect (only) of time-dependent potentials used in experiments. Such a potential does not appear any more simply as a mathematical auxiliary (as for the stationary Internal DFT / KS) and should not necessarily be dropped at the end.

We thus start from a general translationally invariant N-body Hamiltonian composed of the usual kinetic energy term, a translationally invariant two-body potential \( u \), which describes the particle-particle interaction, and an arbitrary translationally invariant "internal" potential \( v_{\text{int}} \) which contains an explicit time dependence

\[
H = \sum_{i=1}^{N} \frac{\mathbf{p}_{i}^{2}}{2m} + \sum_{i\neq j}^{N} u(\mathbf{r}_{i} - \mathbf{r}_{j}) + \sum_{i=1}^{N} v_{\text{int}}(\mathbf{r}_{i} - \mathbf{R}; t). \tag{2}
\]

For the sake of simplicity we assume a 2-body interaction \( u \) and \( N \) identical Fermions or Bosons. The generalization to 3-body etc interactions is straightforward; the generalization to different types of particles is underway.

We rewrite the Hamiltonian (2) using the \( (N-1) \) Jacobi coordinates \( \{ \xi_{\alpha}; \alpha = 1, \ldots, N-1 \} \) and the c.m. coordinate \( \mathbf{R} \), defined as

\[
\xi_{1} = \mathbf{r}_{2} - \mathbf{r}_{1}, \xi_{2} = \mathbf{r}_{3} - \frac{\mathbf{r}_{2} + \mathbf{r}_{1}}{2}, \ldots
\]
The \( \xi_\alpha \) are relative to the c.m. of the other \( 1, \ldots, \alpha - 1 \) particles and are independent from \( R \). They are to be distinguished from the \( N \) "laboratory coordinates" \( r_i \), and the \( N \) "c.m. frame coordinates" \( (r_i - R) \) relative to the total c.m. \( R \). As the \( \{r_i - r_j\} \) and the \( \{r_i - R\} \) can be rewritten as functions of the \( \xi_\alpha \) (in Appendix A) is given the expression of the \( \{r_i - R\} \) as a function of the \( \{\xi_\alpha\} \) coordinates, the interaction \( u \) and the internal potential \( v_{\text{int}} \) can be rewritten as functions of the \( \xi_\alpha \). We denote \( U \) and \( V \) the interaction potential and the internal potential in the Jacobi coordinates representation:

\[
\sum_{i \neq j}^N u(r_i - r_j) \rightarrow U(\xi_1, \ldots, \xi_{N-1})
\]

\[
\sum_{i=1}^N v_{\text{int}}(r_i - R; t) \rightarrow V(\xi_1, \ldots, \xi_{N-1}; t).
\]  

Of course we have \( U[u] \) and \( V[v_{\text{int}}] \). The \( V[v_{\text{int}}] \) potential is \( (N-1) \) body in the Jacobi coordinates representation and cannot be written in a simple form in this representation (see Appendix A). Moreover, various \( v_{\text{int}} \) can lead to the same \( V \), which we will develop later.

After having defined the conjugate momenta of \( R \) and \( \xi_\alpha \), we can separate \( H = H_{\text{CM}} + H_{\text{int}} \), where \( (M = Nm) \) is the total mass

\[
H_{\text{CM}} = -\frac{\hbar^2 \Delta_R}{2M}
\]

is a one-body operator acting in \( R \) space only, and \( (\tau_\alpha \) is the conjugate momentum of \( \xi_\alpha \) and \( \mu_\alpha = m \frac{\tau_\alpha}{\alpha + 1} \) the corresponding reduced mass

\[
H_{\text{int}} = \sum_{\alpha=1}^{N-1} \frac{\tau_\alpha^2}{2\mu_\alpha} + U[u](\xi_1, \ldots, \xi_{N-1})
\]

\[
+ V[v_{\text{int}}](\xi_1, \ldots, \xi_{N-1}; t) \]

is a \( (N-1) \) body operator in the \( \{\xi_\alpha\} \) space. It contains the interaction and the internal potential.

In the time-dependent case, we can choose freely the initial state \( \psi(r_1, \ldots, r_N; t_0) \). We start from an initial state which can be written

\[
\psi(r_1, \ldots, r_N; t_0) = \Gamma(R; t_0) \psi_{\text{int}}(\xi_1, \ldots, \xi_{N-1}; t_0)
\]  

in the Jacobi coordinates representation. This form does not mix the c.m. motion with the internal one (mixing them would not make sense because the c.m. motion does anyway not correspond to the experimental one) and corresponds to the form of the stationary state \( |10, 14\rangle \).

As \( H_{\text{CM}} \) and \( H_{\text{int}} \) act in two separate subspaces, the \( R \) and \( \{\xi_\alpha\} \) spaces (which implies \( [H_{\text{CM}}, H_{\text{int}}] = 0 \)), it is easy to show that the state \( |\psi(t)\rangle \) can be built at all time \( t \geq t_0 \) as a direct product of the form

\[
|\psi(r_1, \ldots, r_N; t)\rangle = \Gamma(R; t) |\psi_{\text{int}}(\xi_1, \ldots, \xi_{N-1}; t)\rangle,
\]

with

\[
H_{\text{CM}}|\Gamma(t)\rangle = i\hbar \partial_t |\Gamma(t)\rangle \quad (9)
\]

\[
H_{\text{int}}|\psi_{\text{int}}(t)\rangle = i\hbar \partial_t |\psi_{\text{int}}(t)\rangle.
\]  

Hence, the \( N \)-body wave function \( \psi \) can be separated into a one-body wave function \( \Gamma \) that depends on the position \( R \) of the c.m. only, and an "internal" \( (N-1) \) body wave function \( \psi_{\text{int}} \) that depends on the remaining \( (N-1) \) Jacobi coordinates \( \xi_\alpha \). Of course, \( \psi_{\text{int}} \) could also be written as a function of the \( N \) laboratory coordinates \( r_i \), but one of them would be redundant. \( \Gamma \) is solution of the free Schrödinger equation and describes the motion of the isolated system as a whole in any chosen inertial frame of reference (as the laboratory). If one starts from a normalizable initial state \( |\Gamma(t_0)\rangle \), \( |\Gamma(t)\rangle \) is condemned to spread more and more. In the stationary limit, the only solutions of Eq. (9) are plane waves, which are infinitely spread (thus not normalizable). This does not correspond to experimental situations, where the system is not isolated anymore: interactions with other systems finitely spread (thus not normalizable). This does not be normalizable. The formal decoupling between the c.m. motion and the internal properties obtained when using the Jacobi coordinates method permits to let the c.m. motion to the choice of experimental conditions, the internal properties being comparable to the experimental ones.

B. Some useful definitions.

We define some quantities and relations that will be useful for the next considerations. In \( |14, 28, 29\rangle \) is defined the internal one-body density

\[
\rho_{\text{int}}(r, t)/N \quad (11)
\]

\[
= \int dr_1 \ldots dr_N \delta(R) |\psi_{\text{int}}(r_1, \ldots, r_N; t)|^2 \delta(r - (r_j - R))
\]

\[
= \left(\frac{N}{N-1}\right)^3 \int d\xi_1 \cdots d\xi_{N-2} |\psi_{\text{int}}(\xi_1, \ldots, \xi_{N-2}, \xi_j, t)|^2.
\]

It is normalized to \( N \). The laboratory density \( \rho(r, t) \) is obtained by convolution of \( \rho_{\text{int}} \) with the c.m. wave function (following \( 28, 29 \)): \( \rho(r, t) = \int dR |\Gamma(R; t)|^2 \rho_{\text{int}}(r - R, t) \).

We also introduced in \( 14 \) the local part of the two-body internal density matrix

\[
\gamma_{\text{int}}(r, r'; t) \quad (12)
\]

\[
= \int dr_1 \cdots dr_N \delta(R) |\psi_{\text{int}}(r_1, \ldots, r_N; t)|^2
\]

\[
\times \delta(r - (r_j - R)) \delta(r' - (r_j - R)).
\]
the internal densities calculated with nates. As one of them is redundant, the fixed initial state δ
following similar steps than in [28, 29], we can show that R
or Bosons, i.e. a given particle-particle interaction potential the definition of the corresponding internal density (11)
Schrödinger equation (10). The main differences lie in we adapt the considerations of [4, 5] to the internal

\[ \rho_{\text{int}}(r, t) = \int d\xi_1 ... d\xi_{N-2} \{ \nabla_{\nu} \psi_{\text{int}}(\xi_1, ..., \xi_{N-2}, \nu; t) \hat{h} \delta_\nu \psi_{\text{int}}^*(\xi_1, ..., \xi_{N-2}, \nu; t) + c.c. \} \Bigg|_{\nu = \frac{\hbar}{i} r} \]

which will be a key equation for the next considerations.

III. TIME-DEPENDENT INTERNAL DFT THEOREM.

A. Preliminaries.

To prove the time-dependent Internal DFT theorem, we adapt the considerations of [4, 5] to the internal Schrödinger equation [10]. The main differences lie in the definition of the corresponding internal density [11] and probability current [13], and in the fact that the potential \( V[\psi_{\text{int}}](\xi_1, ..., \xi_{N-1}; t) \) cannot be written as the sum of one-body potentials in the Jacobi coordinates representation (which introduces some subtleties due to the c.m. correlations and will bring us to use the integral mean value theorem to reach our goal).

In what follows, we consider a given type of Fermions or Bosons, i.e. a given particle-particle interaction \( v \). Solving the “internal” Schrödinger equation [10] for a fixed initial state \( |\psi_{\text{int}}(t_0)\rangle \) and for various internal po-

tentials \( V[\psi_{\text{int}}](\xi_1, ..., \xi_{N-1}; t) \) defines two maps [4, 5] \( F : V[\psi_{\text{int}}]|(\xi_1, ..., \xi_{N-1}; t) \rightarrow |\psi_{\text{int}}(t)\rangle \)
\( G : V[\psi_{\text{int}}]|(\xi_1, ..., \xi_{N-1}; t) \rightarrow \rho_{\text{int}}(r, t) \). (16)

We first notice that two potentials \( v_{\text{int}} \) and \( v'_{\text{int}} \) which lead to two potentials \( V[\psi_{\text{int}}]|(\xi_1, ..., \xi_{N-1}; t) \) and \( V[v'_{\text{int}}]|(\xi_1, ..., \xi_{N-1}; t) \) that differ by a scalar function of time only \( c(t) \), will give two wave functions that differ by a phase \( e^{-i\alpha(t)/\hbar} \) only [4, 5]:

\[ V[v'_{\text{int}}]|(\xi_1, ..., \xi_{N-1}; t) - V[\psi_{\text{int}}]|(\xi_1, ..., \xi_{N-1}; t) = c(t) \]
\[ \Rightarrow |\psi'_{\text{int}}(t)\rangle = e^{-i\alpha(t)/\hbar} |\psi_{\text{int}}(t)\rangle, \]

with \( \alpha(t) = c(t) \). (17)

Then, \( |\psi_{\text{int}}(t)\rangle \) and \( |\psi'_{\text{int}}(t)\rangle \) will give the same density \( \rho_{\text{int}}(r, t) = \rho'_{\text{int}}(r, t) \). The consequence is that the map \( G \) is not fully invertible.

Let us discuss a bit about the condition (17). The form [4] for \( V[v_{\text{int}}] \) implies \( V[v'_{\text{int}}] - V[\psi_{\text{int}}] = V[v'_{\text{int}} - v_{\text{int}}] \).

We define \( \Delta v_{\text{int}}(r; t) = v'_{\text{int}}(r; t) - v_{\text{int}}(r; t) \). (18)
It is to be noted that the condition \( \Delta v_{\text{int}}(r; t) \neq c(t)/N \) is necessary but not sufficient to ensure the condition \( \Delta v_{\text{int}}(\xi_1; \ldots, \xi_{N-1}; t) \neq c(t) \). Indeed, it is possible to have \( \Delta v_{\text{int}}(r; t) \neq c(t)/N \) and nevertheless \( \Delta v_{\text{int}}(\xi_1; \ldots, \xi_{N-1}; t) = c(t) \), because compensations due to the c.m. correlations can happen.

Let us reason on the two particles case, where only one Jacobi coordinate is sufficient to describe the internal properties. We have (see Appendix A): \( V[\Delta v_{\text{int}}](\xi_1; \ldots, \xi_{N-1}; t) \neq c(t) \). We see that if \( \Delta v_{\text{int}}(r; t) \) is an odd function of \( r \) at all time (up to an additional time-dependent function), we have \( \Delta v_{\text{int}}(\xi_1; t) = c(t) \Rightarrow \rho_{\text{int}} = \rho'_{\text{int}} \). This is due to the c.m. correlations, that the non-trivial form of \( V \) reflects. If \( \Delta v_{\text{int}} \) tends to move the first particle in one direction, the second particle will tend to move in the opposite direction because of the c.m. correlations. But if this potential counter-acts perfectly the motion of the second particle (as does an odd potential in the c.m. frame), then the particles remain stuck and the density unchanged.

The same can occur for an arbitrary number of particles. For instance, as \( \sum_{i=1}^{N} (r_i - R) = 0 \), it is obvious with (14) and (15) that every \( \Delta v_{\text{int}}(r; t) = \mathbf{b}(r)t + \mathbf{c}(t)/N \) will yield \( V[\Delta v_{\text{int}}] = c(t) \) even if this form for \( \Delta v_{\text{int}} \) leads to internal potentials which are not null at infinity. Again, this is because if a potential counter acts perfectly the motion due to the c.m. correlations, the particles remain stuck and the density unchanged. In what follows, we consider only internal potentials \( v_{\text{int}} \) and \( v'_{\text{int}} \) that lead to \( V[\Delta v_{\text{int}}] \neq c(t) \).

We come back to Eq. (17) and denote \( |\psi_{\text{int}}(t)\rangle = e^{-i\alpha(t)/h}|\psi_{\text{int}}^0(t)\rangle \) where we define \( \psi_{\text{int}}^0 \) as the wave function obtained for the choice \( c(t) = 0 \), i.e. associated to a \( V[v_{\text{int}}](\xi_1; \ldots, \xi_{N-1}; t) \) where no additive time-dependent function can be split. If we prove that the map \( G \) is invertible up to an additive time-dependent function \( c(t) \), then \( \psi_{\text{int}}^0 \) is fixed by \( \rho_{\text{int}} \) through the relation \( |\psi_{\text{int}}^0(t)\rangle = F G^{-1} \rho_{\text{int}}(r, t) \), which implies that \( \psi_{\text{int}}^0(t) \) can be written as a functional of the internal density \( \rho_{\text{int}} \) defined in (11). Consequently, any expectation value of an operator \( \hat{O} \) which does not contain a time derivative can be written as a functional of the internal density (as the phase cancels out): \( \langle \psi_{\text{int}}(t) | \hat{O} | \psi_{\text{int}}(t) \rangle = \langle \psi_{\text{int}}^0(\rho_{\text{int}}(t)) | \hat{O} | \psi_{\text{int}}^0(\rho_{\text{int}}(t)) \rangle \).

We thus have to show that a propagation of (10) with two potentials \( v_{\text{int}} \) and \( v'_{\text{int}} \) that yield \( V[\Delta v_{\text{int}}](\xi_1; \ldots, \xi_{N-1}; t) \neq c(t) \) will produce two different internal densities \( \rho_{\text{int}} \) and \( \rho'_{\text{int}} \).

### B. The proof.

We start from a fixed initial state \( |\psi_{\text{int}}(t_0)\rangle \) and propagate it with two with two potentials \( v_{\text{int}} \) and \( v'_{\text{int}} \) that give \( V[\Delta v_{\text{int}}](\xi_1; \ldots, \xi_{N-1}; t) \neq c(t) \). We deduce from Eq. (18)
In what follows, we consider \( k \) as the smallest positive integer such that (24) is verified. Then, if we apply \( k \) time derivatives to the Eq. (20), we straightforwardly obtain

\[
\frac{\partial^{k+2}}{\partial t^{k+2}}\left( \rho_{\text{int}}(r,t) - \rho'_{\text{int}}(r,t) \right) \bigg|_{t_0} = \frac{N}{m(N-1)} \int d\xi_1 ... d\xi_{N-2} |\psi_{\text{int}}(\xi_1, ..., \nu; t_0)|^2 \Delta w_k(\xi_1, ..., \nu; t_0) \bigg|_{\nu=\frac{r}{N}} (23)
\]

As, for every physical potential, \( \nabla \xi_{N-1} w_k(\xi_1, ..., \xi_{N-1}; t_0) \) is a real and continuous function in the whole position space, and as \( |\psi_{\text{int}}(\xi_1, ..., \xi_{N-1}; t_0)|^2 \) is a real and positive function in the whole position space, we can apply the integral mean value theorem generalized to many variables functions (demonstrated in Appendix C) to the previous expression. We obtain

\[
\exists (\beta_1, ..., \beta_{N-2}) : m \frac{\partial^{k+2}}{\partial t^{k+2}}\left( \rho_{\text{int}}(r,t) - \rho'_{\text{int}}(r,t) \right) \bigg|_{t_0} = \nabla_r \left[ \nabla \xi_{N-1} w_k(\beta_1, ..., \beta_{N-2}; \frac{N}{N-1} r; t_0) N \left( \frac{N}{N-1} \right)^3 \int d\xi_1 ... d\xi_{N-2} |\psi_{\text{int}}(\xi_1, ..., \frac{N}{N-1} r; t_0)|^2 \right]
\]

\[= \nabla_r \left[ \nabla \xi_{N-1} w_k(\beta_1, ..., \beta_{N-2}; \frac{N}{N-1} r; t_0) \rho_{\text{int}}(r,t) \right]. \quad (24)
\]

To prove the one-to-one correspondence \( V[\xi_{\text{int}}(\xi_1, ..., \xi_{N-1}; t) \mapsto \rho_{\text{int}}(r,t) \) it remains to show that (24) cannot vanish for \( \psi_{\text{int}} \) and \( \psi'_{\text{int}} \) that lead to the relation (21). Then the internal densities \( \rho_{\text{int}}(r,t) \) and \( \rho'_{\text{int}}(r,t) \) would become different infinitesimally later than \( t_0 \). We use the reductio ad absurdum method, in the spirit of Refs. [4, 5]. We suppose that (24) vanishes, which implies:

\[
0 = \frac{N-1}{N} \int dr \Delta w_k(\beta_1, ..., \beta_{N-2}; \frac{N}{N-1} r; t_0) \nabla_r \left[ \nabla \xi_{N-1} w_k(\beta_1, ..., \beta_{N-2}; \frac{N}{N-1} r; t_0) \rho_{\text{int}}(r,t) \right]
\]

\[= - \int dr \left[ \nabla \xi_{N-1} w_k(\beta_1, ..., \beta_{N-2}; \frac{N}{N-1} r; t_0) \right]^2 \rho_{\text{int}}(r,t). \quad (25)
\]

As \( w_k \) is a many-body function, the Eq. (21) does not imply that \( \nabla \xi_{N-1} w_k(\beta_1, ..., \beta_{N-2}, \xi_{N-1}; t_0) \neq \bar{0} \) in the general case. However, we check if this relation holds for the particular form (31) we choose for \( V \). Inserting the results of Appendix A in (4) and (22), we obtain, if \( N > 2 \) (the case \( N = 2 \) will be discussed later on)

\[
\frac{\partial^k}{\partial t^k} \Delta \rho_{\text{int}} \left( \frac{N-1}{N} \xi_{N-1}; t \right) \bigg|_{t_0} = \sum_{i=1}^{N-2} \frac{\partial^k}{\partial t^k} \Delta \rho_{\text{int}} \left( \frac{i}{N} \xi_{N-1} - 1 \right) \bigg|_{t_0} + \frac{\partial^k}{\partial t^k} \Delta \rho_{\text{int}} \left( \sum_{i=1}^{N-2} \frac{1}{\xi_{N-1}} \xi_{N-1} - 1 \right) \bigg|_{t_0},
\]

where we defined

\[
\gamma_{N-2} = \frac{N-2}{N-1} \beta_{N-2} \quad \text{and} \quad \forall i \in [1, N-3] : \gamma_i = \frac{i}{i+1} \beta_i - \sum_{\alpha=i+1}^{N-2} \frac{1}{\alpha+1} \beta_{\alpha}. \quad (27)
\]

The form of the third term of the right hand side of Eq. (26) comes from the fact that \( \sum_{i=1}^{N} (r_i - \bar{R}) = 0 \), which implies, using the Appendix A that \( - \sum_{\alpha=1}^{N-2} \frac{1}{\alpha+1} \beta_{\alpha} = - \sum_{i=1}^{N-2} \gamma_i \). We see from Eq. (27) that the set \( (\gamma_1, ..., \gamma_{N-2}) \) is perfectly defined by the set \( (\beta_1, ..., \beta_{N-2}) \) and vice versa. We now can calculate

\[
\nabla \xi_{N-1} w_k(\beta_1, ..., \beta_{N-2}, \xi_{N-1}; t_0) = \frac{N-1}{N} \frac{1}{\xi_{N-1}} \sum_{i=1}^{N-2} \Delta \rho_{\text{int}} \left( \frac{i}{N} \xi_{N-1} - 1 \right) - \frac{1}{N} \sum_{i=1}^{N-2} \gamma_i \xi_{N-1} - 1 \Delta \rho_{\text{int}} (- \sum_{i=1}^{N-2} \gamma_i \xi_{N-1} - 1), \quad (28)
\]

where we introduced

\[
\mathbf{D}(\mathbf{r}) = \nabla_r \frac{\partial^k}{\partial t^k} \Delta \rho_{\text{int}} (\mathbf{r}; t) \bigg|_{t_0}. \quad (29)
\]

for simplicity. We now check if \( \exists (\beta_1, ..., \beta_{N-2}) : \nabla \xi_{N-1} w_k(\beta_1, ..., \beta_{N-2}, \xi_{N-1}; t_0) = \bar{0} \), which is equivalent, according to (27) and (28), to check if

\[
\exists (\gamma_1, ..., \gamma_{N-2}) : (N-1) \mathbf{D}((N-1) \mathbf{r}) = \sum_{i=1}^{N-2} \Delta \mathbf{D}(\gamma_i - \mathbf{r}) + \Delta \mathbf{D}(- \sum_{i=1}^{N-2} \gamma_i - \mathbf{r}). \quad (30)
\]

Some mathematical considerations show that this equation cannot be fulfilled for all \( \mathbf{r} \) when \( N > 2 \), whatever the set of \( (\gamma_1, ..., \gamma_{N-2}) \), instead if \( \mathbf{D}(\mathbf{r}) = \text{const}. \) But
if $D(r) = \text{const.}$, then $\Delta v_{\text{int}}(r; t)$ should be equal to $b(t) + c(t)/N$, according to \(29\), which is forbidden by the condition \(21\), cf. discussion of the \(\text{III A}\).

It remains to discuss the case $N = 2$. It is easy to show that then, we have $\nabla_{\xi_{N-1}} w_k(\xi_{N-1}; t_0) = -\frac{1}{2}D(-\frac{1}{2}\xi_{N-1}) + \frac{1}{2}D(\frac{1}{2}\xi_{N-1})$, which is null if $D(r)$ is any par function of $r$. But if $D(r)$ is par, then $\frac{\partial}{\partial r^2}\Delta v_{\text{int}}(r; t)$ should be an odd function of $r$ (up to an additional time-dependent function), according to \(29\), which is also forbidden by the condition \(21\), cf. discussion of the \(\text{III A}\).

Thus, we can conclude that, in our case
\[
\forall(\beta_1, ..., \beta_{N-2}) : \nabla_{\xi_{N-1}} w_k(\beta_1, ..., \beta_{N-2}, \xi_{N-1}; t_0) \neq 0.
\]

We immediately deduce the incompatibility of this relation, which is a consequence of \(21\) and of the particular form \(1\) of $V$, with \(23\). Thus, the hypothesis we made is absurd: Eq. \(23\) cannot vanish if $V[\Delta v_{\text{int}}] \neq c(t)$, so that the internal densities $\rho_{\text{int}}(r, t)$ and $\rho_{\text{nucl}}(r, t)$ become different infinitesimally later than $t_0$. As a consequence, the map $G$, defined in \(18\), is invertible (up to an additive time-dependent function) and $|\psi_{\text{nucl}}(t)|$ can be written as a functional of the internal density (we use the notation \(17\)). Thus, any expectation value of an operator $\hat{O}$ which does not contain a time derivative can be written as a functional of $\rho_{\text{int}}$ as the phase cancels out. This achieves to prove the time-dependent Internal DFT theorem (which is a variant of the Runge-Gross theorem \(4, 5\) for self-bound systems and internal densities).

Mind that all the previous reasonings hold only for a fixed initial state $\psi_{\text{int}}(t_0)$ (and a given type of particle), so that $\psi_{\text{int}}^0$ is not only a functional of $\rho_{\text{int}}$, but also depends on $\psi_{\text{int}}(t_0)$. This will be discussed further.

C. Link with traditional (time-dependent) DFT.

We stress here the link and differences between the traditional DFT and internal DFT potentials. We recall that the form of the potential $v_{\text{ext}}$ of traditional DFT can be fundamentally justified starting from the laboratory Hamiltonian of an isolated molecule where the nuclei are treated explicitly. As a molecule is a self-bound system, one can apply the Jacobi coordinates method. We denote the $N$ electronic coordinates related to the laboratory frame as $r_i$, the nuclear c.m. coordinate as $R^{\text{nucl}}$ and the $N$ electronic coordinates related to the c.m. of the nuclei as $r_i^1 = r_i - R^{\text{nucl}}$. A key point concerning the molecules is that, as the nuclei are much heavier than the electrons, the c.m. of the whole molecule coincides with $R^{\text{nucl}}$, and it is an excellent approximation to apply the Jacobi coordinates to the nuclear coordinates only. As a result, the c.m. motion will be described by a $\Gamma(R^{\text{nucl}})$ wave function. The redundant coordinate problem (thus the c.m. correlations) will concern the nuclei only, and will be “external” to the electronic problem: the $N$ electrons are still described by $N$ coordinates. Then, if one decouples the electronic motion from the nuclear one doing the clamped nuclei approximation, the interaction of the electrons with the nuclear background is described by a potential of the form $\sum_{i=1}^N v_{\text{ext}}(r_i - R^{\text{nucl}})$, which becomes $\sum_{i=1}^N v_{\text{ext}}(r_i)$ when moving to the c.m. frame. We then recover the form of the traditional DFT potential.

The potential $v_{\text{ext}}$, which is internal for the (self-bound) molecular problem, becomes external for the pure electronic problem. Those considerations also hold in the time domain, the difference being that the potential
\[
\sum_{i=1}^N v_{\text{ext}}(r_i - R^{\text{nucl}}; t)
\]
can then contain an explicit time dependence in addition to the part which describes the interaction of the electrons with the nuclear background. We recover the traditional time-dependent DFT potential \(4, 5, 26, 27\) when moving in the c.m. frame.

Those reasonings explicit the link between the traditional DFT potential expressed with the laboratory coordinates, Eq. \(23\), and the Internal DFT potential expressed with the laboratory coordinates, Eq. \(1\). They both act only on the internal properties, and not on the c.m. motion (because it is anyway not comparable to the experimental one). The difference is that as, in the molecular case, some particles are much heavier than the other, it is a very good approximation to assimilate the c.m. of the whole molecule with $R^{\text{nucl}}$, which permits to neglect the c.m. correlations for the electronic system, and to justify the clamped nuclei approximation. This simplifies greatly the electronic problem and the traditional DFT can be used to study it. When the particles constituting the self-bound system have nearly the same masses, as it is the case for the nuclei or the He droplets, the total c.m. ($R$) should be calculated with all the particles, so that the c.m. correlations will concern all the particles, and no clamped approximation can be justified. Then, we should use the formalism proposed here.

IV. TIME-DEPENDENT INTERNAL KÖHN-SHAM SCHEME.

We now provide a practical scheme to calculate the internal density $\rho_{\text{int}}$, which consists in the generalization of the stationary Internal KS scheme of \(14\) to the time-dependent case. First, we note that for any normalizable initial state $|\psi_{\text{int}}(t_0)|$, which are the only allowed, the “internal” Schrödinger equation \(10\) stems from a variational principle on the “internal” quantum action \(4, 31, 52\)
\[
A_{\text{int}} = \int_{t_0}^{t_1} dt (\psi_{\text{int}}(t)|i\hbar\hat{H}_t - H_{\text{int}}|\psi_{\text{int}}(t)).
\]
As the function $c(t)$ possibly contained in the potential $V_{\text{int}}$ is perfectly canceled by the time derivative of the
corresponding phase $e^{-ia(t)/\hbar}$ of $\psi_{\text{int}}$, see (17), we have

$$A_{\text{int}} = \int_{t_0}^{t_1} dt \langle \psi_{\text{int}}(t) | \frac{i}{\hbar} \partial_t - H | \psi_{\text{int}}(t) \rangle$$

where we used (11) to obtain the penultimate equality.

We see that the potential $\sum_{i=1}^N v_{\text{int}}(\mathbf{r}_i - \mathbf{R}; t)$ that is $N$ body with respect to the laboratory coordinates (and $(N-1)$ body when expressed with Jacobi coordinates), becomes one body (and local) when expressed with the c.m. frame coordinates (mind that $\rho_{\text{int}}$ is defined in the c.m. frame, cf. (11B).

Applying (33) to the second term of the action integral (32) gives ($\psi_{\text{int}}(t)|U[u](\xi_1, ..., \xi_{N-1})|\psi_{\text{int}}(t)\rangle = \frac{\hbar}{2} \int d\mathbf{r} d\mathbf{r}' \gamma_{\text{int}}(\mathbf{r}, \mathbf{r}'; t) u(\mathbf{r} - \mathbf{r}')$, where $\gamma_{\text{int}}$ is defined in (12).

The action integral (32) can thus be rewritten

$$A_{\text{int}}[\rho_{\text{int}}] = \int_{t_0}^{t_1} dt \langle \psi_{\text{int}}(t) | i\hbar \partial_t - \hbar \gamma_{\text{int}}(\mathbf{r}, \mathbf{r}'; t) u(\mathbf{r} - \mathbf{r}') \rangle$$

To rewrite its last two terms in a more convenient way, we establish a useful relation. For any function $f(\mathbf{r}_1, ..., \mathbf{r}_n; t)$ of the laboratory coordinates, expressible with the Jacobi coordinates [we denote $\psi_{\text{int}}(t)|F(\xi_1, ..., \xi_{N-1}; t)\rangle$, we have

$$F(\xi_1, ..., \xi_{N-1}; t)\rangle = \int d\mathbf{r}_1 \cdots d\mathbf{r}_N F(\mathbf{r}_1, ..., \mathbf{r}_N; t)|\psi_{\text{int}}(\mathbf{r}_1, ..., \mathbf{r}_N; t)\rangle$$

We see that the "internal mean values" calculated with $\psi_{\text{int}}$ expressed as a function of the $(N-1)$ coordinates $\xi_i$, can also be calculated with $\psi_{\text{int}}$ expressed as a function of the $N$ coordinates $\mathbf{r}_i$. As one of them is redundant, a $\delta(\mathbf{R})$ which represents the dependence of the redundant coordinate on the others appears (20).

The relation (21) leads to

$$F(\xi_1, ..., \xi_{N-1}; t)\rangle = \int \mathbf{R} = \int \mathbf{R} \cdot \mathbf{r}_N \delta(\mathbf{R}) F(\mathbf{r}_1, ..., \mathbf{r}_N; t)|\psi_{\text{int}}(\mathbf{r}_1, ..., \mathbf{r}_N; t)\rangle$$

Even if only $(N-1)$ coordinates are sufficient to describe the internal properties, they still describe a system of $N$ particles. Thus, we have to introduce $N$ orbitals in the KS scheme (as we did) if we want them to be interpreted (to first order only) as single-particle orbitals and obtain a scheme comparable (but not equivalent) to mean-field like calculations with effective interactions.

In (37) we implicitly supposed that the particles are Fermions (a KS scheme to describe Boson condensates can be set similarly equaling all the $\varphi_{\text{int}}^i$). Uniqueness of the potential $v_S(\mathbf{r}, t)$ for a given density $\rho_{\text{int}}(t)$ (and initial $|\varphi_{\text{int}}(t_0)\rangle$ which yield the correct initial density $\rho_{\text{int}}(t_0)$) is ensured by a direct application of the traditional time-dependent DFT formalism (4, 5). Of course, the question of the validity of the KS hypothesis, known as the non-interacting $\text{v}$-representability problem, remains, as in traditional (time-dependent) DFT (1, 2).

To use similar kinds of notations than the traditional DFT ones, we add and subtract to the internal action integral (35) the internal Hartree term
\[ A_{H}[\rho_{\text{int}}] = \frac{1}{2} \int_{t_0}^{t_1} dt \int d\mathbf{r} d\mathbf{r}' \rho_{\text{int}}(r, t) \rho_{\text{int}}(r', t) u(\mathbf{r} - \mathbf{r'}), \]

the non-interacting kinetic energy term

\[ \int_{t_0}^{t_1} dt \sum_{i=1}^{N} (\varphi_{\text{int}}^i(t) | \frac{\mathbf{p}^2}{2m} | \varphi_{\text{int}}^i(t)) \] and the

\[ \int_{t_0}^{t_1} dt \sum_{i=1}^{N} (\varphi_{\text{int}}^i(t) i\hbar \partial_t | \varphi_{\text{int}}^i(t)) \] term. This permits to rewrite the “internal” action integral \(^\text{30}\) as

\[ A_{\text{int}} = \int_{t_0}^{t_1} dt \sum_{i=1}^{N} (\varphi_{\text{int}}^i(t) | i\hbar \partial_t - \frac{\mathbf{p}^2}{2m} | \varphi_{\text{int}}^i(t)) - A_{H}[\rho_{\text{int}}] \]

where the internal exchange-correlation part is defined as

\[ A_{XC}[\rho_{\text{int}}] = \frac{1}{2} \int_{t_0}^{t_1} dt \int d\mathbf{r} d\mathbf{r}' \left( \gamma_{\text{int}}(\mathbf{r}, \mathbf{r}'; t) - \rho_{\text{int}}(r, t) \rho_{\text{int}}(r', t) \right) u(\mathbf{r} - \mathbf{r}') \]

\[ + \int_{t_0}^{t_1} dt \left( \psi^0_{\text{int}}(t) \sum_{\alpha=1}^{N-1} \frac{\alpha^2}{2\mu_\alpha} | \psi^0_{\text{int}}(t) \right) - \sum_{i=1}^{N} (\varphi_{\text{int}}^i(t) | \frac{\mathbf{p}^2}{2m} | \varphi_{\text{int}}^i(t)) \]

\[ - \int_{t_0}^{t_1} dt \left( \psi^0_{\text{int}}(t) i\hbar \partial_t | \psi^0_{\text{int}}(t) \right) - \sum_{i=1}^{N} (\varphi_{\text{int}}^i(t) | i\hbar \partial_t | \varphi_{\text{int}}^i(t)) \). \(^\text{(40)}\)

We see that it contains the exchange-correlation which comes from the interaction \(u\) (first line of \(^\text{40}\)), but also the correlations contained in the interacting kinetic energy (second line of \(^\text{40}\)) and in the interacting “i\(\hbar\partial_t\)” term (third line of \(^\text{40}\)). A key point is that, as the KS assumption implies \(\varphi_{\text{int}}^i[\rho_{\text{int}}] \) \(^\text{1, 2, 3}\), \(A_{XC}[\rho_{\text{int}}](t)\) can be written as a functional of \(\rho_{\text{int}}\) (for given \(\psi^0_{\text{int}}(t_0)\)) and \(\{ | \varphi_{\text{int}}^i(t_0) \}\) which yield the same initial density \(\rho_{\text{int}}(\mathbf{r}, t_0)\).

It remains to vary the “internal” quantum action \(^\text{30}\) to obtain the equations of motion (which define \(\rho_{\text{int}}\)). Vignale, see Ref. \(^\text{32}\), showed recently that the correct formulation of the variational principle is not to stationarize the quantum action, i.e. \(\delta A_{\text{int}}[\rho_{\text{int}}] = 0\) as done so far \(^\text{1, 2, 31}\), but

\[ \delta A_{\text{int}}[\rho_{\text{int}}] = \delta A_{H}[\rho_{\text{int}}] + \frac{1}{2} \left( \psi_{\text{int}}[\rho_{\text{int}}](t_1) | \delta \psi_{\text{int}}[\rho_{\text{int}}](t_1) \right) \]

\[ \left( - \frac{\hbar^2}{2m} + U_{H}[\rho_{\text{int}}] + U_{XC}[\rho_{\text{int}}] + v_{\text{int}} \right) \varphi_{\text{int}}^i = i\hbar \partial_t \varphi_{\text{int}}^i \]

\(^\text{(42)}\)

with the potentials

\[ \psi^S_{\text{int}} = \frac{\partial}{\partial \rho_{\text{int}}(\mathbf{r}, t)} \psi_{\text{int}}[\rho_{\text{int}}](t_1) \]

\(^\text{(41)}\)

(\(\psi^S_{\text{int}}\) is the Slater determinant constructed from the \(\varphi_{\text{int}}^i\)). The two formulations lead to identical final results for theorems derived form symmetries of the action functional because compensations occur \(^\text{32}\), but Vignale’s formulation permits to solve the causality paradox of the previous formulation.

Varying \(^\text{41}\) with respect to the \(\varphi_{\text{int}}^i(\mathbf{r}, t)\), with \(t \in [t_0, t_1]\), leads straightforwardly to the Internal time-dependent KS equations for the \(\varphi_{\text{int}}^i\)

\[ \left( - \frac{\hbar^2}{2m} + U_{H}[\rho_{\text{int}}] + U_{XC}[\rho_{\text{int}}] + v_{\text{int}} \right) \varphi_{\text{int}}^i = i\hbar \partial_t \varphi_{\text{int}}^i \]

\(^\text{(42)}\)

which are local as expected (\(v_S = U_{H}[\rho_{\text{int}}] + U_{XC}[\rho_{\text{int}}] + v_{\text{int}}\) with the notations of Eq. \(^\text{37}\)). Note that the variational formulation of Vignale \(^\text{32}\) leads to the addition of the last two terms in the definition of \(U_{XC}[\rho_{\text{int}}](\mathbf{r}, t)\), see Eq. \(^\text{49}\), compared to the traditional result obtained by stationarization of the action. It are those terms which permit to solve the causality paradox \(^\text{32}\).

Equations \(^\text{42}\) have the same form as the traditional time-dependent KS equations formulated for non-translationally invariant Hamiltonians \(^\text{1, 2, 3}\) and permit to define \(\rho_{\text{int}}\) through \(^\text{48}\). Here, we have justified their form in the c.m. frame for self-bound systems de-
scribed with translationally invariant Hamiltonians.

But there is a major difference with the traditional DFT formalism. Following similar steps as in Eq. (54),

\[
(\psi_{\text{int}}^0(t)) \frac{N-1}{\alpha=1} \frac{\delta^2}{2\hbar\alpha} \psi_{\text{int}}^0(t) = \int d\mathbf{r}_1 \cdots d\mathbf{r}_N \delta(\mathbf{R}) \psi_{\text{int}}^0(\mathbf{r}_1, \ldots, \mathbf{r}_N; t) \sum_{i=1}^N \frac{P_i^2}{2m} \psi_{\text{int}}^0(\mathbf{r}_1, \ldots, \mathbf{r}_N; t)
\]

which makes it clear that the differences with the non-interacting kinetic energy term \(\sum_{i=1}^N \int d\mathbf{r}_i \psi_{\text{int}}^0(\mathbf{r}) \frac{P_i^2}{2m} \psi_{\text{int}}^0(\mathbf{r})\) and the non-interacting "\(i\hbar \partial_t\) term" \(\sum_{i=1}^N (\psi_{\text{int}}^0(\mathbf{r}_i(t)) | i\hbar \partial_t | \psi_{\text{int}}^0(\mathbf{r}_i(t)))\) (found in the exchange-correlation functional \(ERF\)) come, on the one hand, from the correlations neglected in the traditional independent-particle framework, but also from the c.m. correlations described by the \(\delta(\mathbf{R})\) term in (44), which does not appear in traditional time-dependent DFT (4). The inclusion of the c.m. correlations in the exchange-correlation functional \(ERF\) and potential \(V_{\text{xc}}\) is the main difference with the traditional KS scheme, and is a key issue for self bound-systems as atomic nuclei.

Mind that all the previous considerations only hold for fixed initial states \(\psi_{\text{int}}(t_0)\) and \(\{|\phi_{\text{int}}(t_0)\}\) which should of course give the same initial density \(\rho_{\text{int}}(\mathbf{r}, t_0)\) (and also for a fixed type of particle). As a consequence, \(\psi_{\text{int}}^0\) is not only a functional of \(\rho_{\text{int}}\), but also depends on the initial state \(\psi_{\text{int}}(t_0)\), and \(U_{\text{xc}}, \) Eq. (43), also depends on the initial orbitals \(|\phi_{\text{int}}(t_0)\rangle\). An important difference to the ground state Internal DFT formalism / KS scheme presented in (14) is that \(\psi_{\text{int}}(t_0)\) and the \(|\phi_{\text{int}}(t_0)\rangle\) cannot necessarily be written as functionals of \(\rho_{\text{int}}(\mathbf{r}, t_0)\). However, as underlined in (4, 5), if one starts from initial states \(\psi_{\text{int}}(t_0)\) and \(|\phi_{\text{int}}(t_0)\rangle\) that are non-degenerate ground states, i.e. that can be written as functionals of \(\rho_{\text{int}}(\mathbf{r}, t_0)\), \(\psi_{\text{int}}\) and \(U_{\text{xc}}\) become functionals of \(\rho_{\text{int}}(\mathbf{r}, t)\) alone. Then, in the limit of stationary ground states, the theory reduces to the stationary Internal DFT / KS.

We recall that, as in traditional DFT, the previously discussed functionals are defined only for internal densities \(\rho_{\text{int}}\) which correspond to some internal potential \(v_{\text{int}}\), called \(v\)-representable internal densities (4, 5). Up to now, we do not know exactly how large the set of \(v\)-representable densities is. This has to be kept in mind when variations with arbitrary densities are done, as to obtain the time-dependent KS equations.

\[V. \ CONCLUSION.\]

In summary, we have shown that, for a fixed initial state, the internal wave function, which describes the internal properties of a time-dependent self-bound system, can be written (up to a trivial phase) as a functional of the internal density. This implies that the "internal" expectation values of any observable (which does not contain a time derivative), that are of experimental interest, can be regarded as functionals of the internal density. Then, we set up, in the c.m. frame, a practical scheme which permits to calculate the internal density and whose form is similar to the traditional time-dependent KS equations, the difference being that the exchange-correlation functional contains the c.m. correlations.

This work is a first step towards the justification to the use of density functionals for time-dependent nuclear mean-field like calculations with effective interactions (23, 24), proving that there exists an ultimate functional which permits to reproduce the exact internal density (up to the non-interacting \(v\)-representability question). If this functional was known, there would be no need for a c.m. correction.

Practically speaking, the time-dependent Internal KS scheme can describe, for instance in the nuclear case, the collision of two nuclei in the frame attached to the total c.m. of the nuclei. Then, \(v_{\text{int}}\) is zero but the dependency to the initial state allows to start from a state which corresponds to two nuclei with different velocities, or "boosts" (chosen such as the total kinetic momentum is zero because we are in the c.m. frame). According to the choice of the boosts, we can describe a wide variety of physical phenomena, from nuclear fusion (23) to Coulomb excitation (23). One of the nuclei can also simply consist in a particle as a proton, to describe the excitation of a nucleus by diffusion.

A case where a non-zero \(v_{\text{int}}\) would be interesting could be the case of the laser irradiation \((v_{\text{int}}\) would then contain a laser potential switched on at \(t > t_0\)). This is not of major interest in the nuclear case because, experimentally speaking, we do not yet have lasers that are suited to the study of the laser irradiation of a nucleus. How-
ever, this could be interesting in view of a generalization of this work to the whole molecule (following from the generalization to different types of particles, which is underway).

Many questions remain open. In particular, the question of the form of the potential which describes the c.m. correlations; in addition to its practical interest, this question would also give interesting arguments concerning the non-interacting v-representability question. Generalization to different types of particles (Fermions or Bosons) appears desirable. Finally, the same reasoning should be applied to rotational invariance to formulate the theory in terms of the so-called "intrinsic" one-body density \( \rho \) (which is not directly observable). This is more complicated because rotation does not decouple from internal motion, but it should be interesting concerning the symmetry breaking question.

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**APPENDIX A: EXPRESSION OF THE \( \{r_i - R\} \) AS FUNCTIONS OF THE \( \{\xi_\alpha\} \) COORDINATES.**

Using the relations (B3), one could show that the \( \{r_i - R\} \) can all be written in function of the \( \{\xi_\alpha\} \) coordinates. We obtain, as a result

\[
\begin{align*}
    r_N - R &= N - 1, \\
    r_{N-1} - R &= N - 2, \\
    r_{N-2} - R &= N - 3.
\end{align*}
\]

\[
\begin{align*}
    r_{N-1} &= \frac{N}{N} \xi_{N-1}, \\
    r_{N-2} &= \frac{N}{N} \xi_{N-2}, \\
    r_{N-3} &= \frac{N}{N} \xi_{N-3}.
\end{align*}
\]

We do some manipulations on the laboratory probability current \( j \) using the Jacobi coordinates (c.c. denotes the complex conjugate; the second equality is obtained using \( \nabla_{\mathbf{r}_N} = \nabla_{\xi_{N-1}} + \nabla_{\mathbf{R}} / N \), by definition of the Jacobi coordinates).
= \int d\mathbf{R} |\Gamma(\mathbf{R}, t)|^2 j_{\text{int}}(\mathbf{r} - \mathbf{R}, t) + \int d\mathbf{R} \rho_{\text{int}}(\mathbf{r} - \mathbf{R}, t) j_{\text{f}}(\mathbf{R}, t), \tag{B3}

where we introduced the internal one-body density (11), the c.m. probability current \( j_{\text{f}}(\mathbf{R}, t) = \frac{\hbar}{i} \Gamma^*(\mathbf{R}, t) \nabla \Gamma(\mathbf{R}, t) + \text{c.c.} \) (\( M = Nm \) is the total mass), and the internal probability current (13). The meaning of Eq. (B3) is clear: the laboratory probability current is the sum of the c.m. probability current and of the internal probability current, both convoluted respectively with the internal one-body density and the c.m. one-body density. One can show that \( j_{\text{f}} \) and \( j_{\text{int}} \) satisfy both independent continuity relations. It is trivial, using (5) and (9), for \( j_{\text{f}} \). For \( j_{\text{int}} \), we calculate (with the help of (6) and (10))

\[
\left( \frac{N}{N-1} \right)^3 \frac{1}{\hbar} \left( \frac{N}{N-1} \right)^3 \int d\xi_1 \ldots d\xi_{N-1} \delta(\xi_{N-1} - \frac{N}{N-1} \mathbf{r}) |\psi_{\text{int}}(\xi_1, \ldots, \xi_{N-1}; t)|^2 \\
= - \frac{\hbar}{2\mu_{N-1}} \left( \frac{N}{N-1} \right)^3 \int d\xi_1 \ldots d\xi_{N-2} \psi_{\text{int}}^*(\xi_1, \ldots, \xi_{N-2}; t) \frac{\hbar^2}{2\mu_{N-1}} \Delta(\xi_{N-1}) \psi_{\text{int}}(\xi_1, \ldots, \xi_{N-1}; t) + \text{c.c.} \\
= - \frac{\hbar}{2\mu_{N-1}} \left( \frac{N}{N-1} \right)^3 \nabla_{\nu} \int d\xi_1 \ldots d\xi_{N-2} \psi_{\text{int}}^*(\xi_1, \ldots, \xi_{N-2}; \nu; t) \nabla_{\nu} \psi_{\text{int}}(\xi_1, \ldots, \xi_{N-2}; \nu; t) \bigg|_{\nu = \frac{N}{N-1} \mathbf{r}} + \text{c.c.} \\
= - \frac{\hbar}{2\mu_{N-1}} \left( \frac{N}{N-1} \right)^3 \nabla_{\nu} \int d\xi_1 \ldots d\xi_{N-2} \psi_{\text{int}}^*(\xi_1, \ldots, \xi_{N-2}; \nu; t) \nabla_{\nu} \psi_{\text{int}}(\xi_1, \ldots, \xi_{N-2}; \nu; t) \bigg|_{\nu = \frac{N}{N-1} \mathbf{r}} + \text{c.c.}
\]

(To obtain the last equality we used the fact that, by definition, \( \mu_{N-1} = \frac{N-1}{N} m \). From this relation we deduce, using (11) and (13), the “internal” continuity equation

\[
\partial_t \rho_{\text{int}}(\mathbf{r}, t) + \nabla_{\mathbf{r}} j_{\text{int}}(\mathbf{r}, t) = 0.
\]

This reinforces the interpretation of \( j_{\text{int}} \) as the internal probability current.

\section*{APPENDIX C: INTEGRAL MEAN VALUE THEOREM FOR FUNCTIONS OF MANY VARIABLES.}

We give the generalization of the mean value theorem to functions of an arbitrary number of variables. One starts from two real functions of \( A \) variables

\[
\forall (x_1, \ldots, x_A) \in \Re : \quad f : [x_1, \ldots, x_A] \mapsto \Re \\
g : [x_1, \ldots, x_A] \mapsto \Re.
\]

We suppose that they are integrable in a domain \( D \), that \( f \geq 0 \) in \( D \), and that \( g \) is continuous in \( D \). We define

\[
m = \inf \{ g(x_1, \ldots, x_A) : (x_1, \ldots, x_A) \in D \} \\
M = \sup \{ g(x_1, \ldots, x_A) : (x_1, \ldots, x_A) \in D \}.
\]

As \( f \geq 0 \), \( mf \leq fg \leq Mf \), which we integrate

\[
m \int_D dx_1 \ldots dx_A f(x_1, \ldots, x_A) \leq \int_D dx_1 \ldots dx_A f(x_1, \ldots, x_A) g(x_1, \ldots, x_A) \leq M \int_D dx_1 \ldots dx_A f(x_1, \ldots, x_A) \\
\Rightarrow \exists C \in [m, M] : \int_D dx_1 \ldots dx_A f(x_1, \ldots, x_A) g(x_1, \ldots, x_A) = C \int_D dx_1 \ldots dx_A f(x_1, \ldots, x_A).
\]

As we supposed that \( g \) is continuous in \( D \), we deduce that \( \exists (x'_1, \ldots, x'_A) \in D : g(x'_1, \ldots, x'_A) = C \), which implies

\[
\exists (x'_1, \ldots, x'_A) \in D : \int_D dx_1 \ldots dx_A f(x_1, \ldots, x_A) g(x_1, \ldots, x_A) = g(x'_1, \ldots, x'_A) \int_D dx_1 \ldots dx_A f(x_1, \ldots, x_A).
\]

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