Time-dependent internal density functional theory formalism and Kohn-Sham scheme for self-bound systems

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The stationary internal density functional theory (DFT) formalism and Kohn-Sham scheme are generalized to the time-dependent case. It is proven that, in the time-dependent case, the internal properties of a self-bound system (such as an atomic nucleus or a helium droplet) are all defined by the internal one-body density and the initial state. A time-dependent internal Kohn-Sham scheme is set up as a practical way to compute the internal density. The main difference from the traditional DFT formalism and Kohn-Sham scheme is the inclusion of the center-of-mass correlations in the functional.

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I. INTRODUCTION

Traditional density functional theory (DFT) [1–3] 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 nondegenerate system of N fermions or bosons [1] 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 [7] and its time-dependent generalization [4,5] provide a straightforward method to compute self-consistently the density in a quantum framework, defining the noninteracting system (i.e., the local single-particle potential), that 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 [9]), and one can apply the Jacobi coordinates method. This permits decoupling of the center-of-mass (c.m.) properties from the internal ones and the correct treatment of 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 $\sum_{i=1}^{N} v_{\text{ext}}(\mathbf{r}_i)$, 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, in contrast to the whole molecule, the pure electronic system is not a self-bound system: The v_{ext} potential breaks translational invariance and is required to reach bound states in the stationary case.

For other self-bound systems, such as isolated atomic nuclei or helium 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 (and 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 $\sum_{i=1}^{N} v_{\text{ext}}(\mathbf{r}_i)$ can be justified in the corresponding self-bound Hamiltonians (where $\mathbf{r}_i$ denote 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 $v_{\text{ext}} \to 0$, but this would lead to false and incoherent results because of the following:

(i) In the stationary case, the HK theorem is valid only for external potentials that lead to bound many-body states [13], which is no longer the case at the limit $v_{\text{ext}} \to 0$ for translationally invariant particle-particle interactions [14].
(ii) The form of v_{ext} is not translationally invariant, but translational invariance is a key feature of self-bound systems [10,14,15].
(iii) Traditional DFT concepts as formulated so far are not applicable in terms of a well-defined internal density ρ_{int}, that is, the density relative to the system's c.m., which is of experimental interest [8,14,16] (being for example measured in nuclear scattering experiments).

Instead of the traditional DFT potential $\sum_{i=1}^{N} v_{\text{ext}}(\mathbf{r}_i)$, one might be tempted to introduce an arbitrary translationally invariant potential of the form $\sum_{i=1}^{N} v_{\text{int}}(\mathbf{r}_i - \mathbf{R})$, where $\mathbf{R} = \frac{1}{N}\sum_{j=1}^{N} \mathbf{r}_j$ is the total c.m. of the particles. This potential is an "internal" potential (i.e., is seen in the c.m. frame), and in Ref. [14] we emphasized that it is the only form that satisfies all the key formal properties. However, v_{int} should be zero in the purely isolated self-bound case. This is why in Ref. [14] we presented it as a mathematical "auxiliary" to reach our goal and showed that it can be dropped in the end without affecting the conclusions. Through it (and using the Jacobi coordinates), we proved, in a different way than those found in Refs. [16,17], the stationary "internal DFT" theorem: The internal many-body state can be written as a functional of ρ_{int}. Then we formulated rigorously the corresponding "internal" KS scheme (in the c.m. frame). The main goal of this work is to take a first step toward 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 that allows reproduction of the exact internal density, which was not clear up to now.

It is to be noted that the stationary internal DFT formalism and KS scheme give a more fundamental justification than the Hartree-Fock (HF) framework to the stationary nuclear mean-field-like calculations. Indeed, the HF framework does not contain quantum correlations, nor does it treat correctly the redundant coordinate problem, which introduces a spurious coupling between the internal properties and the c.m. motion [10,20]. A way to overcome this problem in the stationary case is to perform a projected HF (projection before variation on c.m. momentum), which permits the restoration of Galilean invariance, but at the price of abandoning the independent-particle description [10,15,18]. Within the internal DFT formalism, we proved that the c.m. correlations can be included in the energy functional, thus in the KS potential [14], so that there would be no need for a c.m. projection if the ultimate functional was known.

It is of interest to generalize the stationary internal DFT formalism and KS scheme to the time-dependent case. It would provide a first step toward a fundamental justification to the use of density functionals in nuclear time-dependent calculations with an effective mean field [21,22] and would prove that the c.m. correlations can be included in the functional. This last point is even more interesting in that the spurious c.m. motion problem remains in time-dependent HF [23,24], but then the projected HF method becomes unmanageable and is not used in practice [24].

In this paper, I propose to set up the time-dependent internal DFT formalism and KS scheme. The paper is organized as follows: First the Jacobi coordinates method is applied to the time-dependent full many-body Hamiltonian to decouple the internal properties from the c.m. ones, and some useful "internal" observables, including the internal density, are defined (Sec. II); then it is shown that the internal many-body wave function (and thus the "internal" mean values of all the observables) can be written as a functional of the internal density (Sec. III); finally, the associated time-dependent internal KS scheme is developed as a practical scheme to compute the internal density (Sec. IV).

II. TIME-DEPENDENT N-BODY FORMULATION

A. General formulation

In the time-dependent domain, the introduction of an *explicitly* time-dependent internal potential of the form

$$\sum_{i=1}^{N} v_{\text{int}}(\mathbf{r}_i - \mathbf{R}; t) \tag{1}$$

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 no longer isolated (since they interact with the piece of matter into which they are inserted, which localizes the c.m.). In the time domain, the c.m. motion remains uncomparable to the experimental one (as will be discussed in more detail later), so that it would not make sense to introduce a time-dependent potential that would act on the c.m. motion. It is the internal properties that are of true experimental interest (since experimentalists always deduce those properties [25]). This justifies the introduction of an explicitly time-dependent potential of the form (1), which would act on the internal properties only, and models the internal effect (only) of time-dependent potentials used in experiments. Such a potential no longer appears simply as a mathematical auxiliary (as for the stationary internal DFT) 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_{int}, which contains an explicit time dependence:

$$H = \sum_{i=1}^{N} \frac{\mathbf{p}_i^2}{2m} + \sum_{\substack{i,j=1 \\ i>j}}^{N} u(\mathbf{r}_i - \mathbf{r}_j) + \sum_{i=1}^{N} v_{\text{int}}(\mathbf{r}_i - \mathbf{R}; t). \tag{2}$$

For the sake of simplicity we assume a two-body interaction u and N identical fermions or bosons. The generalization to three- to N-body 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

$$\begin{aligned} \xi_1 &= \mathbf{r}_2 - \mathbf{r}_1, \quad \xi_2 = \mathbf{r}_3 - \frac{\mathbf{r}_2 + \mathbf{r}_1}{2}, \ldots, \\ \xi_{N-1} &= \frac{N}{N-1}(\mathbf{r}_N - \mathbf{R}), \\ \mathbf{R} &= \frac{1}{N}\sum_{j=1}^{N} \mathbf{r}_j. \end{aligned} \tag{3}$$

The ξ_α are relative to the c.m. of the other $1, \ldots, \alpha - 1$ particles and are independent of $\mathbf{R}$. They are to be distinguished from the N "laboratory coordinates" $\mathbf{r}_i$ and the N "c.m. frame coordinates" $(\mathbf{r}_i - \mathbf{R})$ relative to the total c.m. $\mathbf{R}$. Because the $\{\mathbf{r}_i - \mathbf{r}_{j\neq i}\}$ and the $\{\mathbf{r}_i - \mathbf{R}\}$ can be rewritten as

functions of the ξ_α, the interaction u and the internal potential $v_{\rm int}$ can be rewritten as functions of the ξ_α. (Appendix A gives the expression of the $\{\mathbf{r}_i - \mathbf{R}\}$ as a function of the $\{\xi_\alpha\}$ coordinates.) We denote by U and V the interaction potential and the internal potential in the Jacobi coordinates representation:

$$\begin{aligned} \sum_{\substack{i,j=1\\ i>j}}^{N} u(\mathbf{r}_i - \mathbf{r}_j) \quad &\rightarrow \quad U(\xi_1, \ldots, \xi_{N-1}), \\ \sum_{i=1}^{N} v_{\rm int}(\mathbf{r}_i - \mathbf{R}; t) \quad &\rightarrow \quad V(\xi_1, \ldots, \xi_{N-1}; t). \end{aligned} \tag{4}$$

Of course we have $U[u]$ and $V[v_{\rm int}]$. The $V[v_{\rm int}]$ 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 $v_{\rm int}$ can lead to the same V, which we will develop later.

After having defined the conjugate momenta of $\mathbf{R}$ and ξ_α, we can separate (2) into $H = H_{\rm CM} + H_{\rm int}$, where

$$H_{\rm CM} = -\frac{\hbar^2 \Delta_{\mathbf{R}}}{2M} \tag{5}$$

(with $M = Nm$ the total mass) is a one-body operator acting in $\mathbf{R}$ space only and

$$\begin{aligned} H_{\rm int} = &\sum_{\alpha=1}^{N-1} \frac{\tau_\alpha^2}{2\mu_\alpha} + U[u](\xi_1, \ldots, \xi_{N-1}) \\ &+ V[v_{\rm int}](\xi_1, \ldots, \xi_{N-1}; t) \end{aligned} \tag{6}$$

(with τ_α the conjugate momentum of ξ_α and $\mu_\alpha = m\frac{\alpha}{\alpha+1}$ the corresponding reduced mass). $H_{\rm int}$ 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 $\psi(\mathbf{r}_1, \ldots, \mathbf{r}_N; t_0)$. We start from an initial state, which can be written

$$\psi(\mathbf{r}_1, \ldots, \mathbf{r}_N; t_0) = \Gamma(\mathbf{R}; t_0)\ \psi_{\rm int}(\xi_1, \ldots, \xi_{N-1}; t_0) \tag{7}$$

in the Jacobi coordinates representation. This form does not mix the c.m. motion with the internal one (since mixing them would not make sense because the c.m. motion does not correspond to the experimental one) and corresponds to the form of the stationary state [10,14]. Because $H_{\rm CM}$ and $H_{\rm int}$ act in two separate subspaces, the $\mathbf{R}$ and $\{\xi_\alpha\}$ spaces (which implies $[H_{\rm CM}, H_{\rm int}] = 0$), it is easy to show that the state $|\psi(t)\rangle$ can be built at all times $t \geqslant t_0$ as a direct product of the form

$$\psi(\mathbf{r}_1, \ldots, \mathbf{r}_N; t) = \Gamma(\mathbf{R}; t)\ \psi_{\rm int}(\xi_1, \ldots, \xi_{N-1}; t), \tag{8}$$

with

$$H_{\rm CM}|\Gamma(t)\rangle = i\hbar\partial_t|\Gamma(t)\rangle, \tag{9}$$

$$H_{\rm int}|\psi_{\rm int}(t)\rangle = i\hbar\partial_t|\psi_{\rm int}(t)\rangle. \tag{10}$$

Hence, the N-body wave function ψ can be separated for all times into a one-body wave function Γ that depends on the position $\mathbf{R}$ of the c.m. only and an "internal" $(N-1)$ body wave function $\psi_{\rm int}$ that depends on the remaining $(N-1)$ Jacobi coordinates ξ_α. Of course, $\psi_{\rm int}$ could also be written as a function of the N laboratory coordinates $\mathbf{r}_i$, but one of them would be redundant. Γ 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 (such as the laboratory). If one starts from a normalizable initial state $|\Gamma(t_0)\rangle$, $|\Gamma(t)\rangle$ is destined to spread more and more. In the stationary limit, the only solutions of Eq. (9) are plane waves, which are infinitely spread (and thus not normalizable). This does not correspond to experimental situations, where the system is no longer isolated: Interactions with other systems of the experimental apparatus localize the c.m. But the formal decoupling between the c.m. motion and the internal properties obtained when using the Jacobi coordinates method allows one to deduce the internal properties, which can be compared to the experimental ones.

B. Some useful definitions

Is is helpful to define some quantities and relations that will be useful for the calculations to follow. In Refs. [14,26,27] the internal one-body density is defined as

$$\begin{aligned} \rho_{\rm int}(\mathbf{r}, t)/N = &\int d\mathbf{r}_1 \cdots d\mathbf{r}_N\ \delta(\mathbf{R})|\psi_{\rm int}(\mathbf{r}_1, \ldots, \mathbf{r}_N; t)|^2 \\ &\times \delta(\mathbf{r} - (\mathbf{r}_i - \mathbf{R})) \\ = &\left(\frac{N}{N-1}\right)^3 \int d\xi_1 \cdots d\xi_{N-2} \\ &\times \left|\psi_{\rm int}\left(\xi_1, \ldots, \xi_{N-2}, \frac{N\mathbf{r}}{N-1}; t\right)\right|^2. \end{aligned} \tag{11}$$

This density is normalized to N. The laboratory density $\rho(\mathbf{r}, t)$ is obtained by convolution of $\rho_{\rm int}$ with the c.m. wave function (following Refs. [26,27]): $\rho(\mathbf{r}, t) = \int d\mathbf{R}|\Gamma(\mathbf{R}, t)|^2 \rho_{\rm int}(\mathbf{r} - \mathbf{R}, t)$.

We also introduced in Ref. [14] the local part of the two-body internal density matrix

$$\begin{aligned} &\gamma_{\rm int}(\mathbf{r}, \mathbf{r}'; t) \\ &= \int d\mathbf{r}_1 \cdots d\mathbf{r}_N\ \delta(\mathbf{R})|\psi_{\rm int}(\mathbf{r}_1, \ldots, \mathbf{r}_N; t)|^2 \\ &\quad \times \delta(\mathbf{r} - (\mathbf{r}_i - \mathbf{R}))\delta(\mathbf{r}' - (\mathbf{r}_{j\neq i} - \mathbf{R})) \\ &= \frac{N(N-1)}{2}\left(\frac{N-1}{N-2}\right)^3 \left(\frac{N}{N-1}\right)^3 \int d\xi_1 \cdots d\xi_{N-3} \\ &\quad \times \left|\psi_{\rm int}\left(\xi_1, \ldots, \xi_{N-3}, \frac{\mathbf{r}' + (N-1)\mathbf{r}}{N-2}, \frac{N\mathbf{r}'}{N-1}; t\right)\right|^2. \end{aligned} \tag{12}$$

It has the required normalization to $N(N-1)/2$. Following steps similar to those in Refs. [26,27], one can show that the local part of the two-body laboratory density matrix $\gamma(\mathbf{r}, \mathbf{r}', t)$ is obtained by convolution of $\gamma_{\rm int}$ with the c.m. wave function: $\gamma(\mathbf{r}, \mathbf{r}'; t) = \int d\mathbf{R}|\Gamma(\mathbf{R}, t)|^2 \gamma_{\rm int}(\mathbf{r} - \mathbf{R}, \mathbf{r}' - \mathbf{R}; t)$.

The definitions of $\rho_{\rm int}(\mathbf{r}, t)$ and $\gamma_{\rm int}(\mathbf{r}, \mathbf{r}'; t)$ show clearly that they are defined in the c.m. frame, that is, that the $\mathbf{r}$, $\mathbf{r}'$ coordinates are measured in the c.m. frame [see the δ relations in Eqs. (11) and (12)]. Compared to the traditional definitions, a $\delta(\mathbf{R})$ term appears in the definition of the internal densities

calculated with $\psi_{\rm int}$ in $\{\mathbf{r}_i\}$ coordinates. As one of them is redundant, the $\delta(\mathbf{R})$ term represents the dependence of the redundant coordinate on the others [28].

Another quantity that will be very useful is the one-body internal probability current, defined in Appendix B as

$$\mathbf{j}_{\rm int}(\mathbf{r},t)/N = \frac{\hbar}{2mi}\left(\frac{N}{N-1}\right)^3 \times \int d\xi_1 \dots d\xi_{N-2}\psi^*_{\rm int}(\xi_1,\dots,\xi_{N-2},\nu;t) \times \nabla_\nu \psi_{\rm int}(\xi_1,\dots,\xi_{N-2},\nu;t)|_{\nu=\frac{N}{N-1}\mathbf{r}} + \text{c.c.} \quad (13)$$

(where c.c. denotes the complex conjugate), which satisfies the "internal" continuity equation

$$\partial_t \rho_{\rm int}(\mathbf{r},t) + \nabla\cdot\mathbf{j}_{\rm int}(\mathbf{r},t) = 0. \quad (14)$$

Using Eqs. (13), (6), and (10), we obtain the relation

$$\begin{aligned} & i\frac{\partial}{\partial t}\mathbf{j}_{\rm int}(\mathbf{r},t) \\ &= \frac{N}{2mi}\left(\frac{N}{N-1}\right)^3 \int d\xi_1\dots d\xi_{N-2}\Big\{\nabla_\nu\psi_{\rm int}(\xi_1,\dots,\xi_{N-2},\nu;t) i\hbar\partial_t\psi^*_{\rm int}(\xi_1,\dots,\xi_{N-2},\nu;t) \\ &\quad + \nabla_\nu\big(i\hbar\partial_t\psi_{\rm int}(\xi_1,\dots,\xi_{N-2},\nu;t)\big)\psi^*_{\rm int}(\xi_1,\dots,\xi_{N-2},\nu;t) + \text{c.c.}\Big\}\Big|_{\nu=\frac{N}{N-1}\mathbf{r}} \\ &= \frac{N}{2mi}\left(\frac{N}{N-1}\right)^3 \int d\xi_1\dots d\xi_{N-2}\Big\{\nabla_\nu\psi_{\rm int}(\xi_1,\dots,\nu;t)\frac{\hbar^2\Delta_\nu}{2\mu_{N-1}}\psi^*_{\rm int}(\xi_1,\dots,\nu;t) \\ &\quad - \psi^*_{\rm int}(\xi_1,\dots,\nu;t)\nabla_\nu\frac{\hbar^2\Delta_\nu}{2\mu_{N-1}}\psi_{\rm int}(\xi_1,\dots,\nu;t) \\ &\quad + \psi^*_{\rm int}(\xi_1,\dots,\nu;t)\nabla_\nu\big(U[u](\xi_1,\dots,\nu) + V[v_{\rm int}](\xi_1,\dots,\nu;t)\big)\psi_{\rm int}(\xi_1,\dots,\nu;t) + \text{c.c.}\Big\}\Big|_{\nu=\frac{N}{N-1}\mathbf{r}}, \end{aligned} \quad (15)$$

which will be a key equation for what follows.

III. TIME-DEPENDENT INTERNAL DFT THEOREM

A. Preliminaries

To prove the time-dependent internal DFT theorem, we adapt the considerations of Refs. [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 $V[v_{\rm int}](\xi_1,\dots,\xi_{N-1};t)$ cannot be written as the sum of one-body potentials in the Jacobi coordinates representation (which introduces some subtleties owing 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 fermion or boson (i.e., a given particle-particle interaction u). Solving the "internal" Schrödinger equation (10) for a fixed initial state $|\psi_{\rm int}(t_0)\rangle$ and for various internal potentials $V[v_{\rm int}](\xi_1,\dots,\xi_{N-1};t)$ defines two maps [4,5]

$$\begin{aligned} F &: V[v_{\rm int}](\xi_1,\dots,\xi_{N-1};t) \to |\psi_{\rm int}(t)\rangle, \\ G &: V[v_{\rm int}](\xi_1,\dots,\xi_{N-1};t) \to \rho_{\rm int}(\mathbf{r},t). \end{aligned} \quad (16)$$

We first notice that two potentials $v_{\rm int}$ and $v'_{\rm int}$, which lead to two potentials $V[v_{\rm int}](\xi_1,\dots,\xi_{N-1};t)$ and $V[v'_{\rm int}](\xi_1,\dots,\xi_{N-1};t)$ 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]:

$$\begin{aligned} & V[v'_{\rm int}](\xi_1,\dots,\xi_{N-1};t) - V[v_{\rm int}](\xi_1,\dots,\xi_{N-1};t) = c(t) \\ & \quad \Rightarrow |\psi'_{\rm int}(t)\rangle = e^{-i\alpha(t)/\hbar}|\psi_{\rm int}(t)\rangle, \\ & \text{with } \dot\alpha(t) = c(t). \end{aligned} \quad (17)$$

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

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

$$\Delta v_{\rm int}(\mathbf{r};t) = v'_{\rm int}(\mathbf{r};t) - v_{\rm int}(\mathbf{r};t). \quad (18)$$

It is to be noted that the condition $\Delta v_{\rm int}(\mathbf{r};t) \neq c(t)/N$ is necessary but not sufficient to ensure the condition (17), which can be rewritten as $V[\Delta v_{\rm int}](\xi_1,\dots,\xi_{N-1};t) \neq c(t)$. Indeed, it is possible to have $\Delta v_{\rm int}(\mathbf{r};t) \neq c(t)/N$ and nevertheless $V[\Delta v_{\rm int}](\xi_1,\dots,\xi_{N-1};t) = c(t)$, because of possible compensation from the c.m. correlations.

Let us focus on the two-particle case, where only one Jacobi coordinate is sufficient to describe the internal properties. We have (see Appendix A) $V[\Delta v_{\rm int}](\xi_1;t) = \Delta v_{\rm int}(-\frac{1}{2}\xi_1;t) + \Delta v_{\rm int}(\frac{1}{2}\xi_1;t) = \sum_{i=1}^2 v_{\rm int}(\mathbf{r}_i - \mathbf{R};t)$. We see that if $\Delta v_{\rm int}(\mathbf{r};t)$ is an odd function of $\mathbf{r}$ at all t (up to an additional time-dependent function), we have $V[\Delta v_{\rm int}](\xi_1;t) = c(t) \Rightarrow \rho_{\rm int} =$

$\rho'_{\rm int}$. This is due to the c.m. correlations, which the nontrivial form of V reflects. If $\Delta v_{\rm int}$ tends to move the first particle in one direction (in the c.m. frame), the second particle will tend to move in the opposite direction because of the c.m. correlations. But if this potential counteracts 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 remains unchanged.

The same can occur for an arbitrary number of particles. For instance, as $\sum_{i=1}^{N}(\mathbf{r}_i - \mathbf{R}) = 0$, it is obvious from Eqs. (4) and (18) that every $\Delta v_{\rm int}(\mathbf{r};t) = \mathbf{b}(t)\cdot\mathbf{r} + c(t)/N$ will yield $V[\Delta v_{\rm int}] = c(t)$ (even if this form for $\Delta v_{\rm int}$ leads to internal potentials that are not null at infinity). Again, this is because if a potential counteracts perfectly the motion due to the c.m. correlations, the particles remain stuck and the density remains unchanged. In what follows, we consider only internal potentials $v_{\rm int}$ and $v'_{\rm int}$ that lead to $V[\Delta v_{\rm int}] \neq c(t)$.

Let us return to Eq. (17) and denote $|\psi_{\rm int}(t)) = e^{-i\alpha(t)/\hbar}|\psi^0_{\rm int}(t))$, where we define $\psi^0_{\rm int}$ as the wave function obtained for the choice $c(t) = 0$, that is, associated with a $V[v_{\rm int}](\xi_1,\ldots,\xi_{N-1};t)$ 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 $\psi^0_{\rm int}$ is fixed by $\rho_{\rm int}$ through the relation $|\psi^0_{\rm int}(t)) = FG^{-1}\rho_{\rm int}(\mathbf{r},t)$, which implies that $|\psi^0_{\rm int}(t))$ can be written as a functional of the internal density $\rho_{\rm int}$ defined in (11). Consequently, any expectation value of an operator $\hat{O}$ that does not contain a time derivative can be written as a functional of the internal density (since the phase cancels out): $(\psi_{\rm int}(t)|\hat{O}|\psi_{\rm int}(t)) = (\psi^0_{\rm int}[\rho_{\rm int}](t)|\hat{O}|\psi^0_{\rm int}[\rho_{\rm int}](t))$.

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

B. The proof

We start from a *fixed initial state* $|\psi_{\rm int}(t_0))$ and propagate it with two potentials $v_{\rm int}$ and $v'_{\rm int}$ that give $V[\Delta v_{\rm int}](\xi_1,\ldots,\xi_{N-1};t) \neq c(t)$. We deduce from Eq. (15)

$$i\frac{\partial}{\partial t}\left(\mathbf{j}_{\rm int}(\mathbf{r},t) - \mathbf{j}'_{\rm int}(\mathbf{r},t)\right)\Big|_{t=t_0} = \frac{N}{mi}\left(\frac{N}{N-1}\right)^3\int d\xi_1\ldots d\xi_{N-2}|\psi_{\rm int}(\xi_1,\ldots,\nu;t_0)|^2\nabla_\nu V[\Delta v_{\rm int}](\xi_1,\ldots,\nu;t_0)\Big|_{\nu=\frac{N}{N-1}\mathbf{r}}. \quad (19)$$

Using the "internal" continuity relation (14) we obtain

$$\frac{\partial^2}{\partial t^2}\left(\rho_{\rm int}(\mathbf{r},t) - \rho'_{\rm int}(\mathbf{r},t)\right)\Big|_{t=t_0} = \frac{N}{m}\left(\frac{N}{N-1}\right)^3\nabla_{\mathbf{r}}\cdot\int d\xi_1\ldots d\xi_{N-2}\left|\psi_{\rm int}\left(\xi_1,\ldots,\frac{N}{N-1}\mathbf{r};t_0\right)\right|^2\nabla_\nu V[\Delta v_{\rm int}](\xi_1,\ldots,\nu;t_0)\Big|_{\nu=\frac{N}{N-1}\mathbf{r}}. \quad (20)$$

We now make the only hypothesis that is used in this derivation. Following Refs. [4,5] we restrict the set of potentials $v_{\rm int}$ to those that can be expanded into Taylor series with respect to time at the initial time t_0 (which is a reasonable hypothesis for physical potentials). As we supposed that $V[\Delta v_{\rm int}](\xi_1,\ldots,\xi_{N-1};t) \neq c(t)$, we have

$$V[\Delta v_{\rm int}](\xi_1,\ldots,\xi_{N-1};t) \neq c(t) \quad \Rightarrow \quad \exists k : w_k(\xi_1,\ldots,\xi_{N-1};t_0) \neq \text{constant} \quad (21)$$

(with k a positive integer), where

$$w_k(\xi_1,\ldots,\xi_{N-1};t_0) = \frac{\partial^k}{\partial t^k}V[\Delta v_{\rm int}](\xi_1,\ldots,\xi_{N-1};t)\Big|_{t=t_0}. \quad (22)$$

It is to be noted that the condition $\frac{\partial^k}{\partial t^k}\Delta v_{\rm int}(\mathbf{r};t)|_{t=t_0} \neq$ constant $\Rightarrow \nabla_r\frac{\partial^k}{\partial t^k}\Delta v_{\rm int}(\mathbf{r};t)|_{t=t_0} \neq \overrightarrow{0}$ is necessary to ensure the condition (21) [see Eqs. (4) and (22)], but it is not sufficient.

In what follows, we consider k as the *smallest* positive integer such that (21) is verified. Then, if we apply k time derivatives to Eq. (20), we straightforwardly obtain

$$\frac{\partial^{k+2}}{\partial t^{k+2}}\left(\rho_{\rm int}(\mathbf{r},t) - \rho'_{\rm int}(\mathbf{r},t)\right)\Big|_{t=t_0} = \frac{N}{m}\left(\frac{N}{N-1}\right)^3\nabla_{\mathbf{r}}\cdot\int d\xi_1\ldots d\xi_{N-2}|\psi_{\rm int}(\xi_1,\ldots,\nu;t_0)|^2\nabla_\nu w_k(\xi_1,\ldots,\nu;t_0)\Big|_{\nu=\frac{N}{N-1}\mathbf{r}}. \quad (23)$$

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

$$\begin{aligned}
&\exists(\beta_1, \ldots, \beta_{N-2}) : m \frac{\partial^{k+2}}{\partial t^{k+2}} \left(\rho_{\text{int}}(\mathbf{r}, t) - \rho'_{\text{int}}(\mathbf{r}, t)\right)\Big|_{t=t_0} \\
&= \nabla_{\mathbf{r}} \cdot \left[\nabla_{\frac{N\mathbf{r}}{N-1}} w_k \left(\beta_1, \ldots, \beta_{N-2}, \frac{N}{N-1}\mathbf{r}; t_0\right) N \left(\frac{N}{N-1}\right)^3 \int d\xi_1 \ldots d\xi_{N-2} \left|\psi_{\text{int}} \left(\xi_1, \ldots, \frac{N}{N-1}\mathbf{r}; t_0\right)\right|^2 \right] \\
&= \nabla_{\mathbf{r}} \cdot \left[\nabla_{\frac{N\mathbf{r}}{N-1}} w_k \left(\beta_1, \ldots, \beta_{N-2}, \frac{N}{N-1}\mathbf{r}; t_0\right) \rho_{\text{int}}(\mathbf{r}, t_0) \right]. \qquad (24)
\end{aligned}$$

To prove the one-to-one correspondence $V[v_{\text{int}}](\xi_1, \ldots, \xi_{N-1}; t) \leftrightarrow \rho_{\text{int}}(\mathbf{r}, t)$ it remains to be shown that (24) cannot vanish for v_{int} and v'_{int} that satisfy the relation (21). Then the internal densities $\rho_{\text{int}}(\mathbf{r}, t)$ and $\rho'_{\text{int}}(\mathbf{r}, t)$ 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

$$\begin{aligned}
0 &= \frac{N-1}{N} \int d\mathbf{r} w_k \left(\beta_1, \ldots, \beta_{N-2}, \frac{N}{N-1}\mathbf{r}; t_0\right) \nabla_{\mathbf{r}} \cdot \left[\nabla_{\frac{N\mathbf{r}}{N-1}} w_k \left(\beta_1, \ldots, \beta_{N-2}, \frac{N}{N-1}\mathbf{r}; t_0\right) \rho_{\text{int}}(\mathbf{r}, t_0) \right] \\
&= -\int d\mathbf{r} \left[\nabla_{\frac{N\mathbf{r}}{N-1}} w_k \left(\beta_1, \ldots, \beta_{N-2}, \frac{N}{N-1}\mathbf{r}; t_0\right) \right]^2 \rho_{\text{int}}(\mathbf{r}, t_0). \qquad (25)
\end{aligned}$$

As w_k is a many-body function, Eq. (21) does not imply that $\forall(\beta_1, \ldots, \beta_{N-2}) : \nabla_{\xi_{N-1}} w_k(\beta_1, \ldots, \beta_{N-2}, \xi_{N-1}; t_0) \neq \overrightarrow{0}$ in the general case. However, we check whether this relation holds for the particular form (4) we choose for V.

Inserting the results of Appendix A in Eqs. (4) and (22), we obtain, if $N > 2$,

$$\begin{aligned}
&w_k(\beta_1, \ldots, \beta_{N-2}, \xi_{N-1}; t_0) \\
&= \frac{\partial^k}{\partial t^k} \Delta v_{\text{int}} \left(\frac{N-1}{N}\xi_{N-1}; t\right)\Big|_{t=t_0} + \sum_{i=1}^{N-2} \frac{\partial^k}{\partial t^k} \Delta v_{\text{int}} \left(\gamma_i - \frac{1}{N}\xi_{N-1}; t\right)\Big|_{t=t_0} + \frac{\partial^k}{\partial t^k} \Delta v_{\text{int}} \left(-\sum_{i=1}^{N-2} \gamma_i - \frac{1}{N}\xi_{N-1}; t\right)\Big|_{t=t_0}, \qquad (26)
\end{aligned}$$

where we defined

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

The form of the third term of the right-hand side of Eq. (26) comes from the fact that $\sum_{i=1}^{N}(\mathbf{r}_i - \mathbf{R}) = 0$, which implies, using 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 $(\gamma_1, \ldots, \gamma_{N-2})$ is perfectly defined by the set $(\beta_1, \ldots, \beta_{N-2})$ and vice versa. We now can calculate

$$\nabla_{\xi_{N-1}} w_k(\beta_1, \ldots, \beta_{N-2}, \xi_{N-1}; t_0) = \frac{N-1}{N}\mathbf{D}\left(\frac{N-1}{N}\xi_{N-1}\right) - \frac{1}{N}\sum_{i=1}^{N-2}\mathbf{D}\left(\gamma_i - \frac{1}{N}\xi_{N-1}\right) - \frac{1}{N}\mathbf{D}\left(-\sum_{i=1}^{N-2}\gamma_i - \frac{1}{N}\xi_{N-1}\right), \qquad (28)$$

where we introduced

$$\mathbf{D}(\mathbf{r}) = \nabla_{\mathbf{r}} \frac{\partial^k}{\partial t^k} \Delta v_{\text{int}}(\mathbf{r}; t)\Big|_{t=t_0} \tag{29}$$

for simplicity. We now check whether $\exists(\beta_1, \ldots, \beta_{N-2})$: $\nabla_{\xi_{N-1}} w_k(\beta_1, \ldots, \beta_{N-2}, \xi_{N-1}; t_0) = \overrightarrow{0}$, which is equivalent, according to Eqs. (27) and (28), to checking whether

$$\exists(\gamma_1, \ldots, \gamma_{N-2}) : \quad (N-1)\mathbf{D}\big((N-1)\mathbf{r}\big) = \sum_{i=1}^{N-2} \mathbf{D}(\gamma_i - \mathbf{r}) + \mathbf{D}\left(-\sum_{i=1}^{N-2} \gamma_i - \mathbf{r}\right). \tag{30}$$

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

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

Thus, we can conclude that in our case

$$\forall(\beta_1, \ldots, \beta_{N-2}) : \nabla_{\xi_{N-1}} w_k(\beta_1, \ldots, \beta_{N-2}, \xi_{N-1}; t_0) \neq \overrightarrow{0}\,.$$

We immediately deduce the incompatibility of this relation, which is a consequence of (21) and of the particular form (4) of V, with Eq. (25). Thus, the hypothesis we made is absurd: Eq. (24) cannot vanish if $V[\Delta v_{\text{int}}] \neq c(t)$, so that the internal densities $\rho_{\text{int}}(\mathbf{r}, t)$ and $\rho'_{\text{int}}(\mathbf{r}, t)$ become different infinitesimally later than t_0. As a consequence, the map G, defined in Eq. (16), is invertible (up to an additive time-dependent function) and $|\psi^0_{\text{int}}(t))$ can be written as a functional of the internal density [using the notation in Eq. (17)]. Thus, any expectation value of an operator $\hat{O}$ that does not contain a time derivative can be written as a functional of ρ_{int} as the phase cancels out. This proves the time-dependent internal DFT theorem (which is a variant of the Runge-Gross theorem [4,5] for self-bound systems and internal densities).

Keep in mind that all the previous reasoning holds only for a fixed initial state $\psi_{\text{int}}(t_0)$ (and a given type of particle), so that ψ^0_{int} is not only a functional of ρ_{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_{ext} of the 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 $\mathbf{r}_i$, the nuclear c.m. coordinate as $\mathbf{R}^{\text{nucl}}$, and the N electronic coordinates related to the c.m. of the nuclei as $\mathbf{r}'_i = \mathbf{r}_i - \mathbf{R}^{\text{nucl}}$. 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 $\mathbf{R}^{\text{nucl}}$, 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 $\Gamma(\mathbf{R}^{\text{nucl}})$ wave function. The redundant coordinate problem (and 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 using the clamped nuclei approximation, the interaction of the electrons with the nuclear background is described by a potential of the form $\sum_{i=1}^{N} v_{\text{ext}}(\mathbf{r}_i - \mathbf{R}^{\text{nucl}})$, which becomes $\sum_i v_{\text{ext}}(\mathbf{r}'_i)$ when moving to the c.m. frame. We then recover the form of the traditional DFT potential. The potential v_{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

$$\sum_{i=1}^{N} v_{\text{ext}}(\mathbf{r}_i - \mathbf{R}^{\text{nucl}}; t) \tag{31}$$

can then contain an explicit time dependence in addition to the part that describes the interaction of the electrons with the nuclear background. We recover the traditional time-dependent DFT potential [4,5,29,30] when moving in the c.m. frame.

This line of reasoning gives us the link between the traditional DFT potential expressed with the laboratory coordinates, Eq. (31), 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 not comparable to the experimental one). The difference is that as, in the molecular case, some particles are much heavier than others, it is a very good approximation to associate the c.m. of the whole molecule with $\mathbf{R}^{\text{nucl}}$, which allows us to neglect the c.m. correlations for the electronic system and justifies 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 is the case for nuclei or He droplets, the total c.m. ($\mathbf{R}$) 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 KOHN-SHAM SCHEME

A practical scheme to calculate the internal density $\rho_{\rm int}$ is now presented. It consists of the generalization of the stationary internal KS scheme of Ref. [14] to the time-dependent case. First, we note that for any normalizable initial state $|\psi_{\rm int}(t_0))$, which is the only allowed one, the "internal" Schrödinger equation (10) stems from a variational principle on the "internal" quantum action [4,31,32]

$$A_{\rm int} = \int_{t_0}^{t_1} dt (\psi_{\rm int}(t)|i\hbar\partial_t - H_{\rm int}|\psi_{\rm int}(t)). \tag{32}$$

As the function $c(t)$ possibly contained in the potential $V_{\rm int}$ is perfectly canceled by the time derivative of the corresponding phase $e^{-i\alpha(t)/\hbar}$ of $\psi_{\rm int}$ [see Eq. (17)], we have $A_{\rm int} = \int_{t_0}^{t_1} dt(\psi^0_{\rm int}[\rho_{\rm int}](t)|i\hbar\partial_t - H_{\rm int}|\psi^0_{\rm int}[\rho_{\rm int}](t))$ if $V_{\rm int}$ is chosen so that no additive time-dependent function can be split. Thus, the internal quantum action can be considered as a functional of $\rho_{\rm int}$. Its $\int_{t_0}^{t_1} dt(\psi^0_{\rm int}(t)|i\hbar\partial_t - \sum_{\alpha=1}^{N-1}\frac{\tau_\alpha^2}{2\mu_\alpha} - U[u]|\psi^0_{\rm int}(t))$ part is a universal functional of $\rho_{\rm int}$ in the sense that, for a given type of particle (a given interaction u), the same dependence on $\rho_{\rm int}$ holds for every $V[v_{\rm int}]$ and thus $v_{\rm int}$ [see Eq. (4)].

Using Eq. (6), we develop the "internal" quantum action as

$$\begin{aligned} A_{\rm int}[\rho_{\rm int}] = &\int_{t_0}^{t_1} dt \left(\psi^0_{\rm int}(t)\left|i\hbar\partial_t - \sum_{\alpha=1}^{N-1}\frac{\tau_\alpha^2}{2\mu_\alpha}\right|\psi^0_{\rm int}(t)\right) \\ &- \int_{t_0}^{t_1} dt \left(\psi^0_{\rm int}(t)\,|U[u](\xi_1,\ldots,\xi_{N-1})|\,\psi^0_{\rm int}(t)\right) \\ &- \int_{t_0}^{t_1} dt\big(\psi^0_{\rm int}(t)\big|V[v_{\rm int}](\xi_1,\ldots,\xi_{N-1};t)\big|\psi^0_{\rm int}(t)\big). \end{aligned} \tag{33}$$

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

$$\begin{aligned} &\big(\psi^0_{\rm int}(t)|F(\xi_1,\ldots,\xi_{N-1};t)|\psi^0_{\rm int}(t)\big) \\ &\quad= \int d\xi_1\cdots d\xi_{N-1} F(\xi_1,\ldots,\xi_{N-1};t) \\ &\qquad\times\left|\psi^0_{\rm int}(\xi_1,\ldots,\xi_{N-1};t)\right|^2 \\ &\quad= \int d\mathbf{R} d\xi_1\cdots d\xi_{N-1}\delta(\mathbf{R})F(\xi_1,\ldots,\xi_{N-1};t) \\ &\qquad\times\left|\psi^0_{\rm int}(\xi_1,\ldots,\xi_{N-1};t)\right|^2 \\ &\quad= \int d\mathbf{r}_1\cdots d\mathbf{r}_N\delta(\mathbf{R})f(\mathbf{r}_1,\ldots,\mathbf{r}_N;t) \\ &\qquad\times\left|\psi^0_{int}(\mathbf{r}_1,\ldots,\mathbf{r}_N;t)\right|^2. \end{aligned} \tag{34}$$

We see that the "internal mean values" calculated with $\psi_{\rm int}$ expressed as a function of the $(N-1)$ coordinates ξ_α, can also be calculated with $\psi_{\rm int}$ expressed as a function of the N coordinates $\mathbf{r}_i$. Since one of them is redundant, a $\delta(\mathbf{R})$ that represents the dependence of the redundant coordinate on the others appears [28].

Equation (34) leads to

$$\begin{aligned} &\big(\psi^0_{\rm int}(t)\big|V[v_{\rm int}](\xi_1,\ldots,\xi_{N-1};t)\big|\psi^0_{\rm int}(t)\big) \\ &\quad= \int d\mathbf{r}_1\cdots d\mathbf{r}_N\ \delta(\mathbf{R})\sum_{i=1}^{N} v_{\rm int}(\mathbf{r}_i-\mathbf{R};t) \\ &\qquad\times\left|\psi^0_{\rm int}(\mathbf{r}_1,\ldots,\mathbf{r}_N;t)\right|^2 \\ &\quad= \sum_{i=1}^{N}\int d\mathbf{r}\ v_{\rm int}(\mathbf{r};t)\int d\mathbf{r}_1\cdots d\mathbf{r}_N\ \delta(\mathbf{R}) \\ &\qquad\times\left|\psi^0_{\rm int}(\mathbf{r}_1,\ldots,\mathbf{r}_N;t)\right|^2\delta(\mathbf{r}-(\mathbf{r}_i-\mathbf{R})) \\ &\quad= \sum_{i=1}^{N}\int d\mathbf{r}\ v_{\rm int}(\mathbf{r};t)\frac{\rho_{\rm int}(\mathbf{r},t)}{N} \\ &\quad= \int d\mathbf{r}\ v_{\rm int}(\mathbf{r};t)\rho_{\rm int}(\mathbf{r},t), \end{aligned} \tag{35}$$

where we used Eq. (11) to obtain the penultimate equality. We see that the potential $\sum_{i=1}^{N} v_{\rm int}(\mathbf{r}_i-\mathbf{R};t)$, which 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 (where one must keep in mind that $\rho_{\rm int}$ is defined in the c.m. frame, i.e., that $\mathbf{r}$ is measured in the c.m. frame; cf. Sec. II B).

Applying Eq. (34) to the second term of the action integral (33) gives $(\psi^0_{\rm int}(t)|U[u](\xi_1,\ldots,\xi_{N-1})|\psi^0_{\rm int}(t)) = \frac{1}{2}\int d\mathbf{r}d\mathbf{r}'\gamma_{\rm int}(\mathbf{r},\mathbf{r}';t)u(\mathbf{r}-\mathbf{r}')$, where $\gamma_{\rm int}$ is defined in Eq. (12).

The action integral (33) can thus be rewritten as

$$\begin{aligned} A_{\rm int}[\rho_{\rm int}] = &\int_{t_0}^{t_1} dt \left(\psi^0_{\rm int}(t)\left|i\hbar\partial_t - \sum_{\alpha=1}^{N-1}\frac{\tau_\alpha^2}{2\mu_\alpha}\right|\psi^0_{\rm int}(t)\right) \\ &- \frac{1}{2}\int_{t_0}^{t_1} dt\int d\mathbf{r}d\mathbf{r}'\gamma_{\rm int}(\mathbf{r},\mathbf{r}';t)u(\mathbf{r}-\mathbf{r}') \\ &- \int_{t_0}^{t_1} dt\int d\mathbf{r}\ v_{\rm int}(\mathbf{r};t)\rho_{\rm int}(\mathbf{r},t). \end{aligned} \tag{36}$$

Up to now we have not made any hypotheses. To recover the associated internal time-dependent KS scheme, we assume, so as to obtain the traditional time-dependent KS scheme [4,5], that there exists, *in the c.m. frame*, an N-body noninteracting system (i.e., a local single-particle potential v_S)

$$\left(-\frac{\hbar^2\Delta}{2m} + v_S(\mathbf{r},t)\right)\varphi^i_{\rm int}(\mathbf{r},t) = i\hbar\partial_t\varphi^i_{\rm int}(\mathbf{r},t) \tag{37}$$

that reproduces *exactly* the density $\rho_{\rm int}$ of the interacting system

$$\rho_{\rm int}(\mathbf{r},t) = \sum_{i=1}^{N}\left|\varphi^i_{\rm int}(\mathbf{r},t)\right|^2 \tag{38}$$

(where we keep in mind that $\rho_{\rm int}$ is defined in the c.m. frame). 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 Eq. (37) we implicitly supposed that the particles are fermions (but a KS scheme to describe boson condensates can be set similarly equaling all the $\varphi^i_{\rm int}$). Uniqueness of the potential $v_S(\mathbf{r},t)$ for a given density $\rho_{\rm int}(\mathbf{r},t)$ [and initial $|\varphi^i_{\rm int}(t_0)\rangle$] that yield the correct initial density $\rho_{\rm int}(\mathbf{r},t_0)$] 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 *noninteracting v-representability* problem, remains, as in traditional (time-dependent) DFT [1,5].

To use similar kinds of notation to that in traditional DFT, we add and subtract to the internal action integral (36) the internal Hartree term $A_H[\rho_{\rm int}] = \frac{1}{2}\int_{t_0}^{t_1} dt \int d\mathbf{r}d\mathbf{r}'\rho_{\rm int}(\mathbf{r},t)\rho_{\rm int}(\mathbf{r}',t)u(\mathbf{r}-\mathbf{r}')$, the noninteracting kinetic energy term $\int_{t_0}^{t_1} dt \sum_{i=1}^{N}(\varphi^i_{\rm int}(t)|\frac{\mathbf{p}^2}{2m}|\varphi^i_{\rm int}(t))$, and the $\int_{t_0}^{t_1} dt \sum_{i=1}^{N}(\varphi^i_{\rm int}(t)|i\hbar\partial_t|\varphi^i_{\rm int}(t))$ term. This allows us to rewrite the "internal" action integral (36) as

$$A_{\rm int} = \int_{t_0}^{t_1} dt \sum_{i=1}^{N}\left(\varphi^i_{\rm int}(t)\left|i\hbar\partial_t - \frac{\mathbf{p}^2}{2m}\right|\varphi^i_{\rm int}(t)\right) - A_H[\rho_{\rm int}] - A_{\rm XC}[\rho_{\rm int}] - \int_{t_0}^{t_1} dt \int d\mathbf{r}\, v_{\rm int}(\mathbf{r};t)\rho_{\rm int}(\mathbf{r},t), \tag{39}$$

where the internal exchange-correlation part is defined as

$$\begin{aligned} A_{\rm XC}[\rho_{\rm int}] = {} & \frac{1}{2}\int_{t_0}^{t_1} dt \int d\mathbf{r}\, d\mathbf{r}'\big(\gamma_{\rm int}(\mathbf{r},\mathbf{r}';t) - \rho_{\rm int}(\mathbf{r},t)\rho_{\rm int}(\mathbf{r}',t)\big)u(\mathbf{r}-\mathbf{r}') \\ & + \int_{t_0}^{t_1} dt \left(\left(\psi^0_{\rm int}(t)\left|\sum_{\alpha=1}^{N-1}\frac{\tau^2_\alpha}{2\mu_\alpha}\right|\psi^0_{\rm int}(t)\right) - \sum_{i=1}^{N}\left(\varphi^i_{\rm int}(t)\left|\frac{\mathbf{p}^2}{2m}\right|\varphi^i_{\rm int}(t)\right)\right) \\ & - \int_{t_0}^{t_1} dt \left(\big(\psi^0_{\rm int}(t)|i\hbar\partial_t|\psi^0_{\rm int}(t)\big) - \sum_{i=1}^{N}\big(\varphi^i_{\rm int}(t)|i\hbar\partial_t|\varphi^i_{\rm int}(t)\big)\right). \end{aligned} \tag{40}$$

We see that it contains the exchange-correlation that comes from the interaction u [the first line of Eq. (40)] but also the correlations contained in the interacting kinetic energy [the second line of Eq. (40)] and in the interacting "$i\hbar\partial_t$" term [the third line of Eq. (40)]. A key point is that, because the KS assumption implies $\varphi^i_{\rm int}[\rho_{\rm int}]$ [1,4,5], $A_{\rm XC}[\rho_{\rm int}](t)$ can be written as a functional of $\rho_{\rm int}$ [for given $|\psi^0_{\rm int}(t_0)\rangle$ and $\{|\varphi^i_{\rm int}(t_0)\rangle\}$] that yields the same initial density $\rho_{\rm int}(\mathbf{r},t_0)$].

It remains to vary the "internal" quantum action (39) to obtain the equations of motion (which define $\rho_{\rm int}$). Vignale [32] showed recently that the correct formulation of the variational principle is not to stationarize the quantum action (i.e., $\delta A_{\rm int}[\rho_{\rm int}] = 0$) as done so far [4,5,31], but

$$\delta A_{\rm int}[\rho_{\rm int}] = i(\psi_{\rm int}[\rho_{\rm int}](t_1)|\delta\psi_{\rm int}[\rho_{\rm int}](t_1)) - i\big(\psi^S_{\rm int}[\rho_{\rm int}](t_1)\big|\delta\psi^S_{\rm int}[\rho_{\rm int}](t_1)\big) \tag{41}$$

(where $\psi^S_{\rm int}$ is the Slater determinant constructed from the $\varphi^i_{\rm int}$). The two formulations lead to identical final results for theorems derived from symmetries of the action functional because compensations occur [32], but Vignales's formulation allows us to solve the causality paradox of the previous formulation.

Varying Eq. (41) with respect to the $\varphi^{i*}_{\rm int}(\mathbf{r},t)$, with $t \in [t_0,t_1]$, leads straightforwardly to the internal time-dependent KS equations for the $\varphi^i_{\rm int}$:

$$\left(-\frac{\hbar^2}{2m}\Delta + U_H[\rho_{\rm int}] + U_{\rm XC}[\rho_{\rm int}] + v_{\rm int}\right)\varphi^i_{\rm int} = i\hbar\partial_t\varphi^i_{\rm int}, \tag{42}$$

with the potentials

$$\begin{aligned} U_H[\rho_{\rm int}](\mathbf{r},t) &= \frac{\delta A_H[\rho_{\rm int}]}{\delta\rho_{\rm int}(\mathbf{r},t)}, \\ U_{\rm XC}[\rho_{\rm int}](\mathbf{r},t) &= \frac{\delta A_{\rm XC}[\rho_{\rm int}]}{\delta\rho_{\rm int}(\mathbf{r},t)} \\ &\quad - i\left(\psi_{\rm int}[\rho_{\rm int}](t_1)\left|\frac{\delta\psi_{\rm int}[\rho_{\rm int}](t_1)}{\delta\rho_{\rm int}(\mathbf{r},t)}\right.\right) \\ &\quad + i\left(\psi^S_{\rm int}[\rho_{\rm int}](t_1)\left|\frac{\delta\psi^S_{\rm int}[\rho_{\rm int}](t_1)}{\delta\rho_{\rm int}(\mathbf{r},t)}\right.\right), \end{aligned} \tag{43}$$

which are local as expected [$v_S = U_H[\rho_{\rm int}] + U_{\rm XC}[\rho_{\rm int}] + v_{\rm int}$ with the notation of Eq. (37)]. Note that the variational formulation of Vignale [32] leads to the addition of the last two terms in the definition of $U_{\rm XC}[\rho_{\rm int}](\mathbf{r},t)$ [see Eq. (43)], compared to the traditional result obtained by stationarization of the action. It is those terms that allow us to solve the causality paradox [32].

Equations (42) have the same form as the traditional time-dependent KS equations formulated in the laboratory frame for non-translationally invariant Hamiltonians [4,5,7] and allow us to define $\rho_{\rm int}$ through Eq. (38). Here, we have justified their form *in the c.m. frame* for self-bound systems described with translationally invariant Hamiltonians.

But there is a major difference with the traditional DFT formalism. Following similar steps as in Eq. (34), one can show that the interacting kinetic energy term and the interacting "$i\hbar\partial_t$" term can be rewritten [28] as

$$\left(\psi^0_{\rm int}(t)\left|\sum_{\alpha=1}^{N-1}\frac{\tau_\alpha^2}{2\mu_\alpha}\right|\psi^0_{\rm int}(t)\right)=\int d\mathbf{r}_1\cdots d\mathbf{r}_N\delta(\mathbf{R})\psi^{0*}_{\rm int}(\mathbf{r}_1,\ldots,\mathbf{r}_N;t)\sum_{i=1}^{N}\frac{\mathbf{p}_i^2}{2m}\psi^0_{\rm int}(\mathbf{r}_1,\ldots,\mathbf{r}_N;t),$$
$$\left(\psi^0_{\rm int}(t)|i\hbar\partial_t|\psi^0_{\rm int}(t)\right)=\int d\mathbf{r}_1\cdots d\mathbf{r}_N\delta(\mathbf{R})\psi^{0*}_{\rm int}(\mathbf{r}_1,\ldots,\mathbf{r}_N;t)i\hbar\partial_t\psi^0_{\rm int}(\mathbf{r}_1,\ldots,\mathbf{r}_N;t). \quad (44)$$

This makes it clear that the difference between the interacting kinetic energy term and the noninteracting kinetic energy term $\sum_{i=1}^{N}\int d\mathbf{r}\,\varphi^{i*}_{\rm int}(\mathbf{r})\frac{\mathbf{p}^2}{2m}\varphi^i_{\rm int}(\mathbf{r})$ [found in the exchange-correlation functional (40)], and also the difference between the interacting "$i\hbar\partial_t$" term and the noninteracting "$i\hbar\partial_t$" term $\sum_{i=1}^{N}(\varphi^i_{\rm int}(t)|i\hbar\partial_t|\varphi^i_{\rm int}(t))$ [found in (40)], 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 Eq. (44), which does not appear in traditional time-dependent DFT [4,5]. The inclusion of the c.m. correlations in the exchange-correlation functional (40) and potential (43) is the main difference from the traditional KS scheme, and it is a key issue for self-bound systems such as atomic nuclei.

Keep in mind that all the previous considerations only hold for fixed initial states $|\psi_{\rm int}(t_0))$ and $\{|\varphi^i_{\rm int}(t_0))\}$ (and also for a fixed type of particle), which should of course give the same initial density $\rho_{\rm int}(\mathbf{r},t_0)$. As a consequence, $\psi^0_{\rm int}$ is not only a functional of $\rho_{\rm int}$ but also depends on the initial state $|\psi_{\rm int}(t_0))$, and $U_{\rm XC}$, Eq. (43), also depends on the initial orbitals $\{|\varphi^i_{\rm int}(t_0))\}$. An important difference from the ground-state internal DFT formalism presented in Ref. [14] is that $|\psi_{\rm int}(t_0))$ and the $\{|\varphi^i_{\rm int}(t_0))\}$ cannot necessarily be written as functionals of $\rho_{\rm int}(\mathbf{r},t_0)$. However, as emphasized in Refs. [4,5], if one starts from initial states $|\psi_{\rm int}(t_0))$ and $\{|\varphi^i_{\rm int}(t_0))\}$ that are nondegenerate ground states (i.e., that can be written as functionals of $\rho_{\rm int}(\mathbf{r},t_0)$ [14]), $\psi_{\rm int}$ and $U_{\rm XC}$ become functionals of $\rho_{\rm int}(\mathbf{r},t)$ alone. Then, in the limit of stationary ground states, the theory reduces to the stationary internal DFT formalism.

We recall that, as in traditional DFT, the previously discussed functionals are defined only for internal densities $\rho_{\rm int}$ that correspond to some internal potential $v_{\rm int}$, 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 made, as is done in obtaining 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 to calculate the internal density, 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 toward justifying the use of density functionals for time-dependent nuclear mean-field-like calculations with effective interactions [21,22], proving that there exists an ultimate functional that allows us to reproduce the exact internal density (up to the noninteracting v-representability question). If this functional was known, there would be no need for a c.m. correction.

In practical terms, 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, $v_{\rm int}$ is zero but the dependency on the initial state allows us to start from a state that corresponds to two nuclei with different velocities, or "boosts" (chosen such that 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 [21] to Coulomb excitation [33]. One of the nuclei can also simply consist in a particle such as a proton, to describe the excitation of a nucleus by diffusion.

A case where a nonzero $v_{\rm int}$ would be interesting is the case of laser irradiation (in which $v_{\rm int}$ would then contain a laser potential switched on at $t > t_0$). This is not of major interest in the nuclear case because, experimentally, we do not yet have lasers that are suited to the study of laser irradiation of a nucleus. However, 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 that describes the c.m. correlations; in addition to its practical interest, this question would also give interesting arguments concerning the noninteracting 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 [34] (which is not directly observable). This is more complicated because rotation does not decouple from internal motion, but it should shed some light on the symmetry-breaking question.

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APPENDIX A: EXPRESSION OF THE $\{\mathbf{r}_i - \mathbf{R}\}$ AS FUNCTIONS OF THE $\{\xi_\alpha\}$ COORDINATES

Using the relations (3), one could show that the $\{\mathbf{r}_i - \mathbf{R}\}$ can all be written as a function of the $\{\xi_\alpha\}$ coordinates. We obtain, as a result,

$$\begin{aligned}
&\mathbf{r}_N - \mathbf{R} = \frac{N-1}{N}\xi_{N-1},\\
&\mathbf{r}_{N-1} - \mathbf{R} = \frac{N-2}{N-1}\xi_{N-2} - \frac{1}{N}\xi_{N-1},\\
&\mathbf{r}_{N-2} - \mathbf{R} = \frac{N-3}{N-2}\xi_{N-3} - \frac{1}{N-1}\xi_{N-2} - \frac{1}{N}\xi_{N-1},\\
&\quad\vdots\\
&\forall i \in [2, N-2]:\\
&\qquad \mathbf{r}_i - \mathbf{R} = \frac{i-1}{i}\xi_{i-1} - \sum_{\alpha=i}^{N-2}\frac{1}{\alpha+1}\xi_\alpha - \frac{1}{N}\xi_{N-1},\\
&\quad\vdots\\
&\mathbf{r}_1 - \mathbf{R} = -\sum_{\alpha=1}^{N-2}\frac{1}{\alpha+1}\xi_\alpha - \frac{1}{N}\xi_{N-1}.
\end{aligned}$$

The ξ_{N-1} is the only Jacobi coordinate to appear in the expressions (as a function of the Jacobi coordinates) of *all* the $\{\mathbf{r}_i - \mathbf{R}\}$. Thus, all the $v_{\rm int}(\mathbf{r}_i - \mathbf{R}; t)$ terms that enter into the calculation of the total internal potential, Eq. (4), will contain ξ_{N-1}.

APPENDIX B: EXPRESSION OF THE INTERNAL PROBABILITY CURRENT $\mathbf{j}_{\rm int}(\mathbf{r}, t)$

The one-body total laboratory density and probability current are defined as

$$\begin{aligned}
\rho(\mathbf{r}, t) &= N\int d\mathbf{r}_1 \ldots d\mathbf{r}_{N-1}|\psi(\mathbf{r}_1, \ldots, \mathbf{r}_{N-1}, \mathbf{r}; t)|^2,\\
\mathbf{j}(\mathbf{r}, t) &= \frac{\hbar}{2mi}N\int d\mathbf{r}_1 \ldots d\mathbf{r}_{N-1}\psi^*(\mathbf{r}_1, \ldots, \mathbf{r}_{N-1}, \mathbf{r}; t)\\
&\quad\times\nabla_r\psi(\mathbf{r}_1, \ldots, \mathbf{r}_{N-1}, \mathbf{r}; t) + \text{c.c.}
\end{aligned} \tag{B1}$$

(where c.c. denotes the complex conjugate). They satisfy the "laboratory" continuity equation

$$\partial_t\rho(\mathbf{r}, t) + \nabla_r\cdot\mathbf{j}(\mathbf{r}, t) = 0. \tag{B2}$$

We perform some manipulations on the laboratory probability current $\mathbf{j}$ using the Jacobi coordinates:

$$\begin{aligned}
\mathbf{j}(\mathbf{r}, t) &= \frac{\hbar}{2mi}N\int d\mathbf{r}_1 \ldots d\mathbf{r}_{N-1}d\mathbf{r}_N\delta(\mathbf{r} - \mathbf{r}_N)\psi^*(\mathbf{r}_1, \ldots, \mathbf{r}_{N-1}, \mathbf{r}_N; t)\nabla_{r_N}\psi(\mathbf{r}_1, \ldots, \mathbf{r}_{N-1}, \mathbf{r}_N; t) + \text{c.c.}\\
&= \frac{\hbar}{2mi}N\int d\mathbf{R}d\xi_1 \ldots d\xi_{N-1}\left(\frac{N}{N-1}\right)^3\delta\left(\xi_{N-1} - \frac{N}{N-1}(\mathbf{r} - \mathbf{R})\right)\\
&\quad\times\Gamma^*(\mathbf{R}, t)\psi^*_{\rm int}(\xi_1, \ldots, \xi_{N-1}; t)\left(\nabla_{\xi_{N-1}} + \frac{\nabla_R}{N}\right)\Gamma(\mathbf{R}, t)\psi_{\rm int}(\xi_1, \ldots, \xi_{N-1}; t) + \text{c.c.}\\
&= \int d\mathbf{R}|\Gamma(\mathbf{R}, t)|^2\times\frac{\hbar}{2mi}N\left(\frac{N}{N-1}\right)^3\int d\xi_1 \ldots d\xi_{N-2}\psi^*_{\rm int}(\xi_1, \ldots, \xi_{N-2}, \nu; t)\nabla_\nu\psi_{\rm int}(\xi_1, \ldots, \xi_{N-2}, \nu; t)\Big|_{\nu=\frac{N}{N-1}(\mathbf{r}-\mathbf{R})}\\
&\quad + \frac{\hbar}{2mi}\int d\mathbf{R}\Gamma^*(\mathbf{R}, t)\frac{\nabla_R}{N}\Gamma(\mathbf{R}, t)\times N\left(\frac{N}{N-1}\right)^3\int d\xi_1 \ldots d\xi_{N-2}\left|\psi_{\rm int}(\xi_1, \ldots, \xi_{N-2}, \frac{N}{N-1}(\mathbf{r} - \mathbf{R}); t)\right|^2 + \text{c.c.}\\
&= \int d\mathbf{R}|\Gamma(\mathbf{R}, t)|^2\mathbf{j}_{\rm int}(\mathbf{r} - \mathbf{R}, t) + \int d\mathbf{R}\rho_{\rm int}(\mathbf{r} - \mathbf{R}, t)\mathbf{j}_\Gamma(\mathbf{R}, t),
\end{aligned} \tag{B3}$$

where the second equality is obtained using $\nabla_{r_N} = \nabla_{\xi_{N-1}} + \nabla_R/N$ (by the definition of the Jacobi coordinates) and where we introduced the internal one-body density (11), the c.m. probability current $\mathbf{j}_\Gamma(\mathbf{R}, t) = \frac{\hbar}{2Mi}\Gamma^*(\mathbf{R}, t)\nabla_R\Gamma(\mathbf{R}, t) + \text{c.c.}$ (where $M = Nm$ 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 the internal probability current, both convolved, respectively, by the internal one-body density and the c.m. one-body density. One can show that $\mathbf{j}_\Gamma$ and $\mathbf{j}_{\rm int}$ satisfy both independent continuity relations. It is trivial, using Eqs. (5) and (9), for $\mathbf{j}_\Gamma$. For $\mathbf{j}_{\rm int}$, we calculate [with the help of Eqs. (6) and (10)]

$$\begin{aligned}
&\left(\frac{N}{N-1}\right)^3\partial_t\int d\xi_1 \ldots d\xi_{N-1}\delta\left(\xi_{N-1} - \frac{N}{N-1}\mathbf{r}\right)|\psi_{\rm int}(\xi_1, \ldots, \xi_{N-1}; t)|^2\\
&= -\frac{1}{i\hbar}\left(\frac{N}{N-1}\right)^3\int d\xi_1 \ldots d\xi_{N-1}\delta\left(\xi_{N-1} - \frac{N}{N-1}\mathbf{r}\right)\psi^*_{\rm int}(\xi_1, \ldots, \xi_{N-1}; t)\frac{\hbar^2}{2\mu_{N-1}}\Delta_{\xi_{N-1}}\psi_{\rm int}(\xi_1, \ldots, \xi_{N-1}; t) + \text{c.c.}
\end{aligned}$$

$$= -\frac{\hbar}{2\mu_{N-1}i}\left(\frac{N}{N-1}\right)^3 \int d\xi_1 \ldots d\xi_{N-2}\psi^*_{\rm int}(\xi_1,\ldots,\xi_{N-2},\nu;t)\Delta_\nu\psi_{\rm int}(\xi_1,\ldots,\xi_{N-2},\nu;t)\Big|_{\nu=\frac{N}{N-1}\mathbf{r}} + \text{c.c.}$$

$$= -\frac{\hbar}{2\mu_{N-1}i}\left(\frac{N}{N-1}\right)^3 \nabla_\nu \cdot \int d\xi_1 \ldots d\xi_{N-2}\psi^*_{\rm int}(\xi_1,\ldots,\xi_{N-2},\nu;t)\nabla_\nu\psi_{\rm int}(\xi_1,\ldots,\xi_{N-2},\nu;t)\Big|_{\nu=\frac{N}{N-1}\mathbf{r}} + \text{c.c.}$$

$$= -\frac{\hbar}{2mi}\left(\frac{N}{N-1}\right)^3 \nabla_{\mathbf{r}} \cdot \int d\xi_1 \ldots d\xi_{N-2}\psi^*_{\rm int}(\xi_1,\ldots,\xi_{N-2},\nu;t)\nabla_\nu\psi_{\rm int}(\xi_1,\ldots,\xi_{N-2},\nu;t)\Big|_{\nu=\frac{N}{N-1}\mathbf{r}} + \text{c.c.}$$

(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 Eqs. (11) and (13), the "internal" continuity equation

$$\partial_t \rho_{\rm int}(\mathbf{r},t) + \nabla_{\mathbf{r}} \cdot \mathbf{j}_{\rm int}(\mathbf{r},t) = 0.$$

This reinforces the interpretation of $\mathbf{j}_{\rm int}$ as the internal probability current.

APPENDIX C: INTEGRAL MEAN VALUE THEOREM FOR FUNCTIONS OF MANY VARIABLES

We give the generalization of the mean value theorem [35] to functions of an arbitrary number of variables. One starts from two real functions of A variables,

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

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

$$m = \inf\{g(x_1,\ldots,x_A);(x_1,\ldots,x_A)\in D\},$$
$$M = \sup\{g(x_1,\ldots,x_A);(x_1,\ldots,x_A)\in D\}.$$

Because $f \geqslant 0$, $mf \leqslant fg \leqslant Mf$, which we integrate to get

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

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

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

[1] R. M. Dreizler and E. K. U. Gross, *Density Functional Theory* (Springer-Verlag, Berlin, 1990).
[2] W. Kohn, Rev. Mod. Phys. **71**, 1253 (1999).
[3] C. Fiolhais, F. Nogueira, and M. Marques, eds., *A Primer in Density Functional Theory*, Lecture Notes in Physics 620 (Springer-Verlag, Berlin/Heidelberg, 2003).
[4] E. Runge and E. K. U. Gross, Phys. Rev. Lett. **52**, 997 (1984).
[5] E. K. U. Gross, C. A. Ullrich, and U. J. Grossmann, in *Density Functional Theory*, edited by E. K. U. Gross and R. M. Dreizler (Plenum Press, New York, 1994), p. 149.
[6] P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964).
[7] W. Kohn and L. J. Sham, Phys. Rev. **140**, 1133 (1965).
[8] T. Kreibich and E. K. U. Gross, Phys. Rev. Lett. **86**, 2984 (2001).
[9] Translational invariance of the wave function, which implies that the mean values of the observables do not depend on the position of the c.m., is a weaker condition than the more fundamental Galilean invariance of the wave function [10].
[10] K. W. Schmid, Eur. Phys. J. A **12**, 29 (2001).
[11] A multicomponent DFT formalism, for an explicit quantum treatment of both the electrons and the nuclei of a molecule, has been proposed in Refs. [8, 12].
[12] T. Kreibich, R. van Leeuwen, and E. K. U. Gross, Phys. Rev. A **78**, 022501 (2008).
[13] E. H. Lieb, Int. J. Quantum Chem. **24**, 243 (1983).
[14] J. Messud, M. Bender, and E. Suraud, Phys. Rev. C **80**, 054314 (2009).
[15] R. E. Peierls and D. J. Thouless, Nucl. Phys. **38**, 154 (1962).
[16] J. Engel, Phys. Rev. C **75**, 014306 (2007).
[17] N. Barnea, Phys. Rev. C **76**, 067302 (2007).
[18] M. Bender, P.-H. Heenen, and P.-G. Reinhard, Rev. Mod. Phys. **75**, 121 (2003).

[19] M. Barranco, R. Guardiola, S. Hernandez, R. Mayol, J. Navarro, and M. Pi, J. Low Temp. Phys. **142**, 1 (2006).
[20] P. Ring and P. Schuck, *The Nuclear Many-Body Problem* (Springer-Verlag, New York/Heidelberg/Berlin, 1980).
[21] C. Simenel, B. Avez, and D. Lacroix, Lecture given at the "Joliot Curie" school, arXiv:0806.2714.
[22] J. W. Negele, Rev. Mod. Phys. **54**, 913 (1982).
[23] J. M. Irvine, J. Phys. G **6**, L175 (1980).
[24] A. S. Umar and V. E. Oberacker, J. Phys. G **36**, 025101 (2009).
[25] Experimentalists use the c.m. observables (position, momentum, or kinetic energy of the c.m.) to transform all the other observables into the c.m. frame, using the Galilean invariance.
[26] B. G. Giraud, Phys. Rev. C **77**, 014311 (2008).
[27] K. Yabana, Prog. Theor. Phys. **76**, 414 (1986).
[28] More generally, we should introduce a $\delta(\mathbf{R}-\mathbf{a})$, where $\mathbf{a}$ is an arbitrary translation vector, instead of the $\delta(\mathbf{R})$. This is linked to the translational invariance. This paper uses $\mathbf{a}=\mathbf{0}$, which leads to perfectly equivalent results and permits simpler notation.
[29] E. K. U. Gross and W. Kohn, Adv. Quantum Chem. **21**, 255 (1990).
[30] M. A. L. Marques and E. K. U. Gross, Annu. Rev. Phys. Chem. **55**, 427 (2004).
[31] A. K. Kerman and S. E. Koonin, Ann. Phys. (NY) **100**, 332 (1976).
[32] G. Vignale, Phys. Rev. A **77**, 062511 (2008).
[33] K. Alder, A. Bohr, T. Huus, B. Mottelson, and A. Winther, Rev. Mod. Phys. **28**, 432 (1956).
[34] B. G. Giraud, B. K. Jennings, and B. R. Barrett, Phys. Rev. A **78**, 032507 (2008).
[35] J. Bass, *Cours de Mathématiques* (Masson et Cie., Paris, 1964).