PHYSICAL REVIEW RESEARCH **6**, 013060 (2024)

Orbital-free quasidensity functional theory

Carlos L. Benavides-Riveros [*]
Pitaevskii BEC Center, CNR-INO and Dipartimento di Fisica, Universitá di Trento, I-38123 Trento, Italy
and Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Strasse 38, D-01187 Dresden, Germany

(Received 18 April 2023; accepted 22 December 2023; published 16 January 2024)

We develop an *orbital-free* functional framework to compute one-body quasiprobabilities for both fermionic and bosonic systems. Since the key variable is a quasidensity, this theory circumvents the problems of finding the Pauli potential or approximating the kinetic energy that are known to limit the applicability of standard orbital-free density functional theory. We present a set of strategies to (a) compute the one-body Wigner quasiprobability in an orbital-free manner from the knowledge of the universal functional and (b) obtain those functionals from the functionals of the one-body reduced density matrix (1-RDM). We find that the universal functional of optical lattices results from a translation, a contraction, and a rotation of the corresponding functional of the 1-RDM, revealing the strong connection between these two functional theories. Furthermore, we relate the key concepts of *Wigner negativity* and v *representability*.

DOI: 10.1103/PhysRevResearch.6.013060

I. INTRODUCTION

Detecting and understanding quantum features at the macroscopic level is one of the main theoretical and technological challenges of modern quantum sciences. Nowadays, state-of-the-art experiments can directly observe nonclassical behavior (as quantum superposition) in systems with a truly macroscopic number of particles [1–4]. A powerful theoretical and computational strategy to detect that *quantumness* is by directly measuring the system's corresponding Wigner function. Although normalized to unity, Wigner functions are quasiprobability distributions that can take negative values, a phenomenon that has no classical counterpart. As a result, in a wide range of research fields the detection of negativity of the Wigner functions has been linked to nonclassical features of quantum states and is broadly considered a distinctive signature of quantum entanglement [5–7], contextuality [8–10], quantum computation [11], quantum steering [12,13], or even quantum gravity [14].

Due to the exponentially large Hilbert spaces of quantum many-body systems, finding the corresponding Wigner function is, in general, a computationally prohibitive task. Yet when dealing with identical particles it is possible to circumvent the Hilbert space's exponential growth by relying upon universal functionals of certain reduced, more manageable, quantities. For instance, based on the important observation that electronic systems are fully determined by the ground-state density [15], density functional theory (DFT) is a prominent methodology in electronic structure calculations, with applications ranging from quantum chemistry and material science [16,17] to self-driving labs [18]. Unfortunately, although progress is being made to reconstruct many-body observables from the density [19,20], standard DFT is, in general, not suitable for describing many nonclassical features from which Wigner negativity is just a remarkable example.

A recent phase-space formulation of DFT employs, as the central variable, the one-particle Wigner quasidensity. Similar to the Hohenberg-Kohn theorems of DFT, this quasidensity is in a one-to-one correspondence with the respective ground state for interacting many-fermion/boson systems [21]. Its main feature is that the one-body Wigner function can be accessed directly, without precomputing the full wave function or finding the map to the ground-state density. This Wigner quasidensity functional theory (quasi-DFT) is a promising theoretical tool to model many-body problems while accounting for nonclassical features, strong interactions, and quantum correlations, with the same computational cost as standard DFT. As we show below, the theory has also the potential to bypass well-known problems of orbital-free DFT which, while achieving a computational linear scaling with the system size, requires expressing the kinetic energy in terms of the density [22,23]. Despite these clear advantages, there are neither orbital-free nor orbital-dependent functionals for quasi-DFT.

The main goal of this work is to unveil a set of equations that allows us to compute the fermionic/bosonic one-particle quasidensity in a fully orbital-free manner. We show that $\omega(\mathbf{r},\mathbf{p})$, the one-particle Wigner quasidensity, satisfies the following effective eigenequation:

$$h_{\text{eff}}(\mathbf{r},\mathbf{p}) \star \omega(\mathbf{r},\mathbf{p}) = \mu\, \omega(\mathbf{r},\mathbf{p}), \tag{1}$$

where $h_{\text{eff}}(\mathbf{r},\mathbf{p}) = \frac{1}{2}\mathbf{p}^2 + v_{\text{ext}}(\mathbf{r}) + v_{\text{eff}}(\mathbf{r},\mathbf{p})$. Here $v_{\text{ext}}(\mathbf{r})$ is the external potential, $v_{\text{eff}}(\mathbf{r},\mathbf{p})$ is the certain effective potential that we introduce below, and the symbol $\star$ is the so-called

---

[*] cl.benavidesriveros@unitn.it

star product of phase-space quantum mechanics. We show that introducing this $\star$ product in the context of functional theories has far-reaching consequences for functional theories applied to fermionic and bosonic systems. For instance, since it is possible to express this $\star$ product as a sum in powers of $\hbar$, it is possible to truncate at some order, and, as a result, an infinite sequence of differential equations will arise.

This work is organized as follows. First, we review orbital-free functional theories and discuss the Wigner formulation of DFT. Second, we derive an Euler-Lagrange equation for the one-body Wigner quasidensity. Next, we derive Eq. (1) and discuss its main theoretical implications. We then employ the Hubbard model to present a functional realization of quasi-DFT and perform a calculation with a large number of particles. We conclude with a summary and discuss some implications of our results.

II. FUNCTIONAL THEORIES OF THE MANY-BODY PROBLEM

The enormous success of DFT in electronic structure calculations is mainly due to the existence of a set of self-consistent one-particle equations that allow for the computation of the density $\rho(\mathbf{r})$ from one-particle Kohn-Sham orbitals $\{\phi_j(\mathbf{r})\}$ [24]:

$$\rho(\mathbf{r}) = \sum_{j=1}^{N} |\phi_j(\mathbf{r})|^2. \tag{2}$$

Although DFT is cheaper than wave-function methods, this Kohn-Sham formulation still has a well-known unfavorably computational scaling with the cube of the number of electrons [25]. Orbital-free DFT has a much more favorable, linear scaling with the system size [17,22], because to find the ground-state electron density, the total energy expression is directly minimized in the set of densities whose number of electrons is fixed. This yields the following Lagrangian:

$$\mathcal{L}[\rho(\mathbf{r})] = \mathcal{E}[\rho(\mathbf{r})] - \mu\left(\int \rho(\mathbf{r}) d\mathbf{r} - N\right), \tag{3}$$

with $\mathcal{E}[\rho(\mathbf{r})] = \mathcal{T}[\rho(\mathbf{r})] + \mathcal{E}_{\rm ext}[\rho(\mathbf{r})] + \mathcal{E}_{\rm H}[\rho(\mathbf{r})] + \mathcal{E}_{\rm xc}[\rho(\mathbf{r})]$ being the kinetic, external, Hartree, and exchange-correlation energy functionals. Applying a functional derivative with respect to the electron density yields the Euler-Lagrange equation of orbital-free DFT:

$$\frac{\delta \mathcal{T}[\rho(\mathbf{r})]}{\delta \rho(\mathbf{r})} + V_{\rm eff}(\mathbf{r}) = \mu, \tag{4}$$

where $V_{\rm eff}(\mathbf{r}) = V_{\rm ext}(\mathbf{r}) + V_{\rm H}(\mathbf{r}) + V_{\rm xc}(\mathbf{r})$ are the functional derivatives of the energy functionals.

However, this derivation makes it clear that the computational advantage of orbital-free DFT is counterbalanced by the fact that the quantum mechanical kinetic energy functional is unknown, reducing both its accuracy and its applicability. Efforts are being made to solve this problem either by writing it as a classical, approximate function of the electron density, or, more recently, by training deep neural networks [22].

A parallel intellectual effort is the one-particle reduced density matrix functional theory (1-RDMFT) whose purpose is to seek a universal functional of the one-body reduced density matrix (1-RDM) [26–28]:

$$\gamma(\mathbf{r}, \mathbf{r}') = \sum_j n_j \varphi_j(\mathbf{r}) \varphi_j^*(\mathbf{r}'), \tag{5}$$

for fermionic [29–31], bosonic [32–35], or relativistic [36] interacting particles. Similar to DFT, 1-RDMFT is based on a one-to-one correspondence between the ground state and its corresponding 1-RDM. Yet, although this theory is in a better position than DFT to tackle strong correlations due to the presence of fractional occupations n_j [37,38], its broad use has been hampered by the absence of Kohn-Sham-like equations for the natural orbitals $\varphi_j(\mathbf{r})$ (i.e., the eigenvectors of the 1-RDM) [28,39], and therefore, 1-RDMFT is computationally much more demanding than DFT [40].

While DFT and 1-RDMFT are somehow connected theories [e.g., the diagonal of the 1-RDM $\gamma(\mathbf{r}, \mathbf{r})$ gives the density $\rho(\mathbf{r})$], they are in practice quite different. One of the main differences lies in the fact that the functionals of 1-RDM are usually written as a *de facto* reconstruction of the two-body reduced density matrix (2-RDM) in terms of the 1-RDM, i.e., the occupation numbers n_j and the natural orbitals $\varphi_j(\mathbf{r})$ [41,42]. For instance, for two-electron systems in the singlet case the exact 2-RDM can be written in terms of the 1-RDM [43]:

$$\Gamma(\mathbf{r}, \mathbf{s}; \mathbf{r}', \mathbf{s}') = \frac{1}{2} \sum_{ij} \sqrt{n_i n_j} \varphi_i(\mathbf{r}) \varphi_i(\mathbf{s}') \varphi_j^*(\mathbf{r}') \varphi_j^*(\mathbf{s}'). \tag{6}$$

This has two consequences: first, the theory takes in practice the form of a natural orbital functional theory where $\Gamma[\{n_j\}, \{\varphi_j\}]$ is the functional to be evaluated [44], and second, the attempt to build an orbital-free formulation of 1-RDMFT is nonexistent, despite the fact that it could boost its broad applicability and its computational efficiency. This work can be seen as an attempt to fill this gap and initiate such an orbital-free framework for 1-RDMFT.

III. WIGNER QUASI-DFT

Before presenting our novel conceptual framework we recap the phase-space formulation of quantum mechanics. Next, we present two complementary equations to perform orbital-free calculations of the one-body Wigner quasidensity.

A. Phase-space formulation of quantum mechanics

In the phase-space formulation of quantum mechanics, observables are represented by symbols, i.e., functions of position $\mathbf{r}$ and momentum $\mathbf{p}$ coordinates. Out of many choices, Wigner functions host a quite natural representation of quantum mechanics [45], mainly because in the classical limit it turns out to be the phase-space distribution of statistical mechanics. While there are other phase-space distributions (e.g., Berezin's Q or P functions), the Wigner functions are the only ones that are real and give the correct marginal probabilities (i.e., they give the densities of DFT by partial integration). Furthermore, as indicated in the Introduction, the negativity of the Wigner function is broadly considered a distinctive signature of non-classical quantum features.

In this phase-space formulation, quantum operators correspond uniquely to phase-space classical functions via the Weyl correspondence. Operator products correspond to $\star$ products. This noncommutative star (twisted or Moyal) product is commonly defined by the phase-space pseudodifferential operator [46]:

$$\star \equiv \exp[i\hbar(\overleftarrow{\partial}_r\overrightarrow{\partial}_p - \overleftarrow{\partial}_p\overrightarrow{\partial}_r)/2], \tag{7}$$

where the arrows denote that a given derivative acts only on a function standing on the left or right. This product is defined by $\mathcal{Q}(f \star g) = \mathcal{Q}(f)\mathcal{Q}(g)$, where $\mathcal{Q}(f)$ is the quantized operator version (by the Weyl rule) of the phase-space function f [47]. The eigenvalue problem for the Hamiltonian H reads $H \star f_n = E_n f_n = f_n \star H$. Another important property is that the integral of the $\star$ multiplication reduces to a plain integral, namely, $\int d\Omega\,(f \star g) = \int d\Omega\,(g \star f) = \int d\Omega f g$, with $d\Omega = d^3r d^3p$.

B. One-body quasidensity

By definition, the one-body Wigner quasidensities are given in terms of the 1-RDM $\gamma(\mathbf{r}, \sigma; \mathbf{r}', \sigma')$ by the following relation:

$$\omega^{\sigma\sigma'}(\mathbf{r}, \mathbf{p}) = \frac{1}{\pi^3}\int \gamma(\mathbf{r} - \mathbf{z}, \sigma; \mathbf{r} + \mathbf{z}, \sigma')\, e^{2i\mathbf{p}\cdot\mathbf{z}}\, d^3z, \tag{8}$$

where $\sigma \in \{\uparrow, \downarrow\}$ are the spin variables. Notice that the marginal $\sum_\sigma \int \omega^{\sigma\sigma}(\mathbf{r}, \mathbf{p})\, d^3p$ gives exactly the density $\rho(\mathbf{r})$ (the central object of DFT) [48].

A generalization of the Hohenberg-Kohn [15] and Gilbert [26] theorems to Hamiltonians of the form $H = h + V$, with a *fixed* two-particle interaction V, proves the existence of a universal Wigner functional $\mathcal{F}_V[\omega]$ of the one-body quasidensity ω [21]. Indeed, for any choice of the one-particle phase-space Hamiltonian $h(\mathbf{r}, \mathbf{p}) = \frac{1}{2}\mathbf{p}^2 + v_{\text{ext}}(\mathbf{r}, \mathbf{p})$, the energy functional

$$\mathcal{E}[\omega] \equiv \int h(\mathbf{r}, \mathbf{p})\, \omega(\mathbf{r}, \mathbf{p})\, d\Omega + \mathcal{F}_V[\omega] \geqslant E_{\text{gs}}, \tag{9}$$

with $d\Omega = d^3r d^3p$ being the phase-space infinitesimal volume, is bounded from below by the exact ground-state energy. The equality in Eq. (9) holds when $\mathcal{E}[\omega]$ is evaluated using the ground-state one-body quasidensity ω_{gs}.

Two similarities arise with both DFT and 1-RDMFT. As in standard DFT, the functional $\mathcal{F}_V[\omega]$ is completely independent of any external (phase-space) potential $v(\mathbf{r}, \mathbf{p})$. As in 1-RDMFT, it is also completely independent of the kinetic energy and depends only on the fixed two-particle interaction V.

The (universal) functional $\mathcal{F}_V[\omega]$ obeys a constrained-search formulation, by considering only many-body wavefunctions that integrate to the same ω:

$$\mathcal{F}_V[\omega] = \min_{\Psi \to \omega} \langle \Psi | V | \Psi \rangle. \tag{10}$$

While the functional is unknown, it is known that, due to the exact knowledge of the kinetic energy functional, it has some better scaling properties than the functionals in DFT. For instance, by defining $\omega_\lambda = \omega(\lambda\mathbf{r}, \lambda^{-1}\mathbf{p})$, one can show that $\mathcal{F}_V[\omega_\lambda] = \lambda\mathcal{F}_V[\omega]$ [21].

Furthermore, in stark contrast to DFT, *any* one-particle observable $o(\mathbf{r}, \mathbf{p})$ can be computed directly from the ground-state one-particle Wigner function by using a rather simple linear functional: $\langle \hat{o} \rangle = \int o(\mathbf{r}, \mathbf{p})\omega(\mathbf{r}, \mathbf{p}) d\Omega$. While two-particle observables remain challenging, we present below a way to derive the interacting part of the ground-state energy for discrete systems from the knowledge of the functional of 1-RDMFT.

Finally, in a quite natural way, one-body quasidensities inherit the representability conditions of the 1-RDM. In fact, due to unitary invariance, those can be expressed as conditions on the eigenvalues of γ [49]. Therefore, it is convenient to use the spectral representation of ω (i.e., $\omega = \sum_i n_i f_i$) to find its representability conditions. In general, $n_i \geqslant 0$. In addition, in the case of fermions,

$$\omega \star \omega \leqslant \omega, \tag{11}$$

which is just a consequence of the Pauli exclusion principle [50].

C. Two equations for the one-body quasidensity

We now exhibit our first result: the exact equation for the phase-space one-body quasidensity. Let $\mathcal{E}[\omega]$ be the energy functional of the Wigner function (9), subject to the constraint $\int d\Omega\, \omega(\mathbf{r}, \mathbf{p}) = N$. The N-particle phase-space density which minimizes such a functional is found by applying a functional derivative of the Lagrangian $\mathcal{E}[\omega] - \mu N$ with respect to ω, yielding the Euler-Lagrange equation of Wigner quasi-DFT:

$$h(\mathbf{r}, \mathbf{p}) + \frac{\delta\mathcal{F}_V[\omega]}{\delta\omega(\mathbf{r}, \mathbf{p})} = \mu. \tag{12}$$

There is an important consequence of this result. As is well known, one of the central problems in orbital-free DFT is approximating the kinetic energy functional in terms of the density [51,52] or, alternatively, the Pauli potential [53,54]. It is, indeed, particularly crucial that the Pauli principle be captured precisely in the kinetic energy. As we can see in Eq. (12), this important problem is completely absent in the phase-space formalism. First, the kinetic energy and the external potential are exact, rather simple phase-space functions, and no approximation is needed. Second, the representability condition of the Wigner function (11) guarantees that the Pauli principle is fulfilled. As a consequence, our orbital-free quasi-DFT needs only to approximate the universal functional $\mathcal{F}_V[\omega]$.

We present the second result of our work. Inspired by the work of Levy, Perdew, and Sahni [55], we exhibit an exact eigenequation for the one-particle quasidensity that does not require natural orbitals. Indeed, operation on the right of the Eq. (3), evaluated at the exact ground-state one-body quasidensity, with the same function, yields a $\star$-eigenvalue equation,

$$h_{\text{eff}}(\mathbf{r}, \mathbf{p}) \star \omega_{\text{gs}}(\mathbf{r}, \mathbf{p}) = \mu\, \omega_{\text{gs}}(\mathbf{r}, \mathbf{p}), \tag{13}$$

where $h_{\text{eff}}(\mathbf{r}, \mathbf{p}) = h(\mathbf{r}, \mathbf{p}) + \delta\mathcal{F}_V[\omega]/\delta\omega(\mathbf{r}, \mathbf{p})|_{\omega=\omega_{\text{gs}}}$. In the same way, operation from the left gives

$$\omega_{\text{gs}}(\mathbf{r}, \mathbf{p}) \star h_{\text{eff}}(\mathbf{r}, \mathbf{p}) = \mu\, \omega_{\text{gs}}(\mathbf{r}, \mathbf{p}), \tag{14}$$

These equations can be compared with the one from orbital-free DFT for $\sqrt{\rho(\mathbf{r})}$ [56]:

$$\left[-\tfrac{1}{2}\nabla^2 + v_{\text{ext}}(\mathbf{r}) + v_{\text{eff}}(\mathbf{r})\right]\sqrt{\rho(\mathbf{r})} = \mu\sqrt{\rho(\mathbf{r})}, \tag{15}$$

where $v_{\rm ext}(\mathbf{r})$ is the external potential and $v_{\rm eff}(\mathbf{r})$ is a sum of the Kohn-Sham $v_{\rm KS}(\mathbf{r})$ and the Pauli $v_\theta(\mathbf{r})$ potentials. While the advantage of Eq. (15) over the more common Kohn-Sham approach is evident, a general expression for $v_\theta(\mathbf{r})$ in terms of the density is unknown [54]. Strikingly, in the phase-space formulation, only the derivative of the functional is needed and no Pauli potential is required.

Introducing the product $\star$ in the context of functional theories has additional advantages: it is possible to truncate the expansion at some power of $\hbar$, and, as a result, a sequence of different differential equations will arise. Indeed, the formula (13) allows for a Wigner-Moyal expansion of the equation for the quasidensity [57]:

$$\sum_n \frac{i^n\hbar^n}{2^n n!} h_{\rm eff}(\overleftarrow{\partial}_r\overrightarrow{\partial}_p - \overleftarrow{\partial}_p\overrightarrow{\partial}_r)^n\omega = \mu\,\omega, \tag{16}$$

which can be separated into two equations, namely,

$$\sum_n \frac{(-)^n\hbar^{2n}}{2^{2n}(2n+1)!} h_{\rm eff}(\overleftarrow{\partial}_r\overrightarrow{\partial}_p - \overleftarrow{\partial}_p\overrightarrow{\partial}_r)^{2n+1}\omega = 0 \tag{17}$$

and

$$\sum_n \frac{(-)^n\hbar^{2n}}{2^{2n}(2n)!} h_{\rm eff}(\overleftarrow{\partial}_r\overrightarrow{\partial}_p - \overleftarrow{\partial}_p\overrightarrow{\partial}_r)^{2n}\omega = \mu\,\omega. \tag{18}$$

These equations comprise a quite rich structure. For instance, up to order $O(\hbar^3)$, we have $\{h_{\rm eff}, \omega\} = 0$, where $\{,\}$ denotes the Poisson bracket of classical mechanics, and

$$h_{\rm eff}\left[1 - \frac{\hbar^2}{8}(\overleftarrow{\partial}_r\overrightarrow{\partial}_p - \overleftarrow{\partial}_p\overrightarrow{\partial}_r)^2\right]\omega = \mu\,\omega. \tag{19}$$

To be solved, these equations require, of course, some approximate form of the exact functional $\mathcal{F}_V[\omega]$ and its functional derivative. Unfortunately, explicit functionals of ω are absent in the literature (save the exchange part of the Hartree-Fock functional that we present in Appendix B). In the next section, we propose and work out a strategy to find such functionals.

IV. FUNCTIONAL REALIZATION

As already mentioned, although 1-RDMFT functionals could be Wigner transformed, almost all of them are written in terms of natural orbitals [42,58–67], so they are not suited for our purposes. Let us, therefore, illustrate the potential of orbital-free quasi-DFT by discussing the generalized Fermi and Bose-Hubbard models, whose standard version has been broadly used to unveil aspects of functional theories [32,68–72]. For two sites, the interacting Hamiltonian, containing all particle-conserving quartic terms, can be written with three parameters in the following way:

$$\begin{aligned} V(u_1,u_2,u_3) = {} & u_1\sum_{j=l,r}\hat{n}_j(\hat{n}_j-1) \\ & + u_2\hat{n}_l\hat{n}_r + u_3\big[(b_l^\dagger)^2 b_r^2 + (b_r^\dagger)^2 b_l^2\big], \end{aligned} \tag{20}$$

where $b_j^\dagger$, b_j, and $\hat{n}_j$ are the corresponding creation, annihilation and particle-number operators on the left and right sites $j \in \{l, r\}$. Normalizing to 1 and assuming real-valued matrix elements, the 1-RDM can be represented in the lattice-site basis $|l\rangle$, $|r\rangle$ as

$$\gamma = \tfrac{1}{2} + \vec{\gamma}\cdot\vec{\sigma}, \tag{21}$$

where $\vec{\gamma} = (\gamma_{lr}, 0, \gamma_{ll} - \frac{1}{2})$, $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ is the vector of Pauli matrices, and $\gamma_{ij} = \langle i|\gamma|j\rangle$.

To write the corresponding (discrete) Wigner transformation we follow Refs. [73–76], where the Wigner function is represented on a grid of twice the dimension of the underlying Hilbert space $\{j, n\}$. For the momentum basis, we choose the one in which the hopping term of the Hubbard Hamiltonian is diagonal: $|n\rangle = [|l\rangle + (-1)^n|r\rangle]/\sqrt{2}$ for $n \in \{0, 1\}$. This grid can be seen as a two-dimensional vector space over a finite field, in which the Wigner function is defined as

$\omega_{l,1}$	$\omega_{r,1}$
$\omega_{l,0}$	$\omega_{r,0}$

As explained in Appendix A, for a qubit, a phase-space point operator is [76]

$$\begin{aligned} \Omega(n,\phi_m) = {} & \tfrac{1}{2}[1 + (-1)^m(|0\rangle\langle 0| - |1\rangle\langle 1|) \\ & + (-1)^n(|0\rangle\langle 1| + |1\rangle\langle 0|) \\ & + i(-1)^{n+m}(|0\rangle\langle 1| - |1\rangle\langle 0|)]. \end{aligned} \tag{22}$$

Therefore, by computing $\omega(n,\phi_m) = \frac{1}{4}{\rm Tr}[\gamma\Omega(n,\phi_m)]$, one finds in vectorized form the following equation:

$$|\omega\rangle\!\rangle = \tfrac{1}{2}\mathcal{D}|\gamma\rangle\!\rangle, \tag{23}$$

where

$$|\omega\rangle\!\rangle = \begin{pmatrix}\omega_{l,0}\\ \omega_{l,1}\end{pmatrix} \quad\text{and}\quad \mathcal{D} = \begin{pmatrix}1 & 1\\ 1 & -1\end{pmatrix}, \tag{24}$$

with $\omega_{r,1} = \frac{1}{2} - \omega_{l,0}$ and $\omega_{r,0} = \frac{1}{2} - \omega_{l,1}$. Notice that $\mathcal{D}$ is an orthogonal matrix. Inverting Eq. (23) one gets $|\gamma(\omega)\rangle\!\rangle = \mathcal{D}|\omega\rangle\!\rangle$. Thus, the one-body Wigner quasidensity can now be computed as

$$\omega_{j,n} = \tfrac{1}{2}[\gamma_{jj} + (-1)^n\gamma_{lr}]. \tag{25}$$

As it should be, the marginal densities are recovered by the partial sums: $\sum_n \omega_{j,n} = \gamma_{jj}$ and $\sum_j \omega_{j,n} = \tilde{\gamma}_{nn}$, where $\tilde{\gamma}_{nn}$ is the momentum density. Since $\omega_{l,0}$ and $\omega_{l,1}$ determine $\omega_{r,0}$ and $\omega_{r,1}$, we take $\omega_{l,0}$ and $\omega_{l,1}$ as our two degrees of freedom.

Using the semipositivity of γ, it is straightforward to check that the representability condition reads as follows:

$$\left(\omega_{l,0} - \tfrac{1}{4}\right)^2 + \left(\omega_{l,1} - \tfrac{1}{4}\right)^2 \leqslant \tfrac{1}{8}. \tag{26}$$

In Fig. 1 are presented two different realizations of the Hamiltonian (20) for both 1-RDMFT and quasi-DFT. It can be seen that the functional of quasi-DFT results from the respective 1-RDMFT functional after a translation, a contraction, and a rotation of 45°. This result is general for lattice systems, as indicated in Appendix A. Finally, notice that, due to the general form of the formula (25), the same result will hold for the Fermi-Hubbard dimer. After applying Eq. (12) [or a discrete version of Eq. (13)], one can find $\omega_{\rm gs}$ for specific

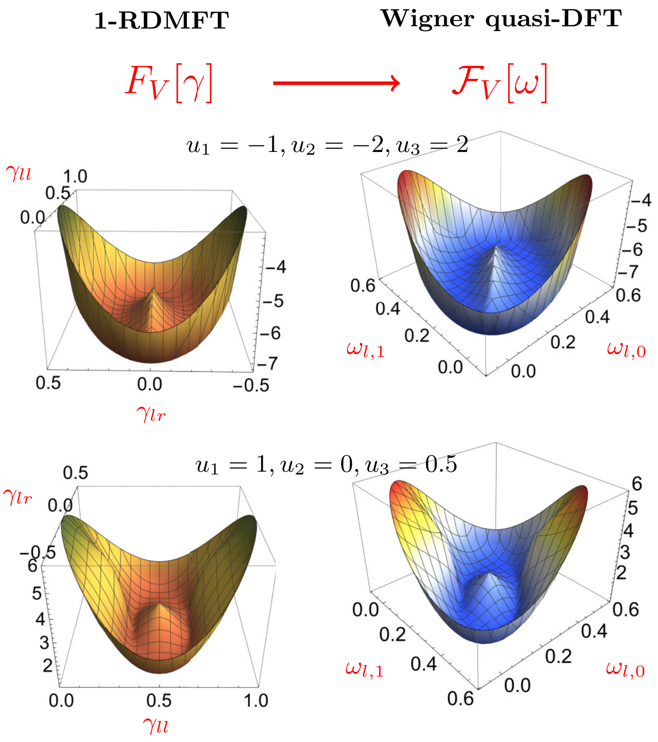

FIG. 1. Universal functionals of 1-RDMFT $F_V[\gamma]$ and Wigner quasi-DFT $\mathcal{F}_V[\gamma]$ for two realizations of the generalized Hubbard dimer (20) for three particles.

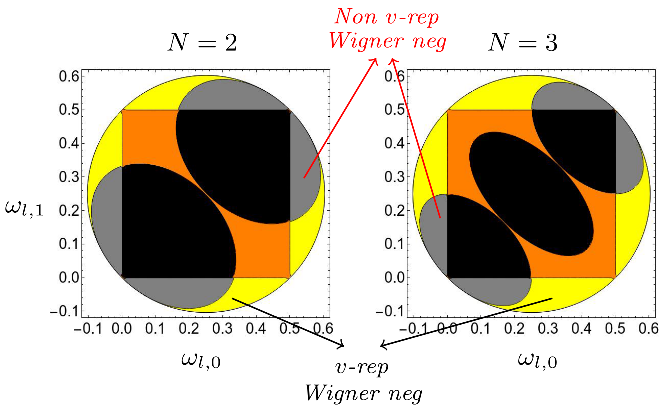

FIG. 2. Representation of the domain of Wigner one-body quasidensities for the Bose-Hubbard dimer (20) with $u_1 = 1$ and $u_2 = u_3 = 0$, for two and three bosons. Wigner positive $\omega > 0$ are represented in black (non-v-representable) and orange (v-representable). Wigner negative ω are represented in yellow (v-representable) and gray (non-v-representable).

values of t (the strength of the hopping term) and $v_l - v_r$ (the external potential).

Finally, we present the last result of this section. The connection between 1-RDM and quasidensities presented above allows us to relate two important concepts in Wigner and functional theories: *Wigner negativity* and *v representability*. The question is: *which fermionic or bosonic ground states give place to Wigner one-body quasidensities with negative values?* We answer explicitly this question for two and three particles for the standard Hubbard dimer in Fig. 2: There are four disconnected ground-state regions of Wigner negativity (the yellow areas). Relating these two important concepts seems to be new in the literature.

V. FUNCTIONAL FOR CONDENSATES

In this section, we present results for the Wigner quasidensity for an arbitrary number of particles close to the condensate phase. We use the spherical symmetry of the quasidensity (26) to parametrize it with a radius ρ and an angle φ, namely, $\omega_{l,0}(\rho,\varphi) = \frac{1}{4}[1+\sqrt{2}\rho\cos(\varphi)]$ and $\omega_{l,1}(\rho,\varphi) = \frac{1}{4}[1+\sqrt{2}\rho\sin(\varphi)]$. While an analytical expression for the universal functional is beyond the scope of what is currently possible, it is still possible to provide an expression for the condensate state. Following Refs. [32,34], let us perform the following rotation of the original site basis: $|\rho\rangle = \cos(\varphi/2)|l\rangle + \sin(\varphi/2)|r\rangle$ and $|\varphi\rangle = -\sin(\varphi/2)|l\rangle + \cos(\varphi/2)|r\rangle$. In this new basis, states of the form

$$|N-n,n\rangle_\rho \equiv \frac{1}{\sqrt{(N-n)!n!}}(a^\dagger_\rho)^{N-n}(a^\dagger_\varphi)^n|0\rangle.$$

map to one-body quasidensities with the same φ and only ρ changes. The idea is to perform the transformation of the entire Hamiltonian to the states $|\rho\rangle$ and $|\varphi\rangle$. This will allow us to perform the calculation of the functional in the vicinity of the condensate phase analytically. For simplicity let us consider the Hamiltonian $V(u_1,u_2,0)$ of Eq. (20), which we write from now on as

$$V(u_1,u_2) = u_1\sum_{j=l,r}\hat{n}_j(\hat{n}_j-1) + u_2\,\hat{n}_l\hat{n}_r. \tag{27}$$

Let us separate the Hamiltonian into two terms: $\sum_{j=l,r}\hat{n}_j(\hat{n}_j-1) \equiv \hat{h}_1(\rho,\varphi)$ and $\hat{n}_l\hat{n}_r \equiv \hat{h}_2(\rho,\varphi)$.

The first term $\hat{h}_1(\rho,\varphi)$ was computed in Ref. [32,34], giving

$$\begin{aligned}\hat{h}_1(\rho,\varphi)\\ &= \left[1-\tfrac{1}{2}\sin^2(\varphi)\right]\left(\hat{n}^2_\varphi+\hat{n}^2_\rho\right) + \tfrac{1}{2}\sin^2(\varphi)(N+4\hat{n}_\rho\hat{n}_\varphi)\\ &\quad + \tfrac{1}{2}\sin^2(\varphi)[(\hat{a}^\dagger_\rho)^2(\hat{a}_\varphi)^2 + (\hat{a}^\dagger_\varphi)^2(\hat{a}_\rho)^2] + \hat{\kappa}_1(\rho,\varphi),\end{aligned} \tag{28}$$

where $\hat{\kappa}_1(\rho,\varphi)$ is an operator that contains only one-particle excitations, which plays no role in our derivation. For the second term we obtain

$$\begin{aligned}\hat{h}_2(\rho,\varphi)\\ &= \tfrac{1}{4}\sin^2(\varphi)\left(\hat{n}^2_\varphi+\hat{n}^2_\rho\right) + [1-\sin^2(\varphi)]\hat{n}_\rho\hat{n}_\varphi + \tfrac{1}{4}N\sin^2(\varphi)\\ &\quad - \tfrac{1}{4}\sin^2(\varphi)[(\hat{a}^\dagger_\rho)^2(\hat{a}_\varphi)^2 + (\hat{a}^\dagger_\varphi)^2(\hat{a}_\rho)^2] + \hat{\kappa}_2(\rho,\varphi).\end{aligned} \tag{29}$$

Here the operator $k_2(\rho,\varphi)$ contains only one-particle excitations, and will also play no role in our derivation. Next, we write the state $|\Psi_{\widetilde{\rm BEC}}\rangle = \alpha_0|N,0\rangle_\rho \pm \alpha_1|N-2,2\rangle_\rho$ and compute the expectation value $\langle\Psi_{\widetilde{\rm BEC}}|V(u_1,u_2)|\Psi_{\widetilde{\rm BEC}}\rangle$. It will be convenient to define $\delta = 2\alpha_1^2 = 1-\rho$. The exact relation with the Wigner function amounts to $\omega_{l,0}(\varphi,\delta) = \frac{1}{2}[\cos^2(\varphi/2)(N-\delta) + \sin^2(\varphi/2)\delta + \cos(\varphi/2)\sin(\varphi/2)(N-\delta)$. We then obtain an expression for the universal functional (with an explicit dependence with the number of particles N):

$$\mathcal{F}_N[\omega(\varphi,\delta)] = E^{(0)}_{u_1,u_2}(\varphi) - E^{(1/2)}_{u_1,u_2}(\varphi)\delta^{1/2} \\ + E^{(1)}_{u_1,u_2}(\varphi)\delta + O(\delta^{3/2}),$$

where $E^{(0)}_{u_1,u_2}(\varphi) = [N(N-1) - \frac{1}{2}N(N-1)\sin^2(\varphi)]u_1 + \frac{1}{4}N(N+1)\sin^2(\varphi)u_2$, $E^{(1/2)}_{u_1,u_2}(\varphi) = \sqrt{N(N-1)}(u_1 - \frac{1}{2}u_2)\sin^2(\varphi)$, and $E^{(1)}_{u_1,u_2}(\varphi) = (N-2)[(u_2 - 2u_1) + \frac{1}{2}\sin^2(\varphi)(6u_1 - 3u_2)]$. This functional describes an arbitrary number of atoms in the phase space. Notice that $u_2 = 2u_1$ exactly cancels the term $\delta^{1/2}$ that gives the Bose-Einstein condensate (BEC) force.

VI. CONCLUSION

Unveiling the role of quantum effects at the classical level is a crucial problem for many areas of quantum science ranging from developing quantum technologies to the characterization of quantum entanglement. The Wigner quasiprobability is usually employed as a probe of such quantumness. In this work we have presented and developed a set of strategies to (a) compute $\omega(\mathbf{r},\mathbf{p})$, the (fermionic or bosonic) one-body Wigner quasidensity in an orbital-free manner from the knowledge of the universal functional, and (b) to obtain those functionals from functionals of the 1-RDM. By providing an Euler-Lagrange equation and a Wigner-Moyal eigenequation, we showed that $\omega(\mathbf{r},\mathbf{p})$ can be computed as a stationary point without referring to orbital equations, circumventing some known problems of orbital-free DFT (e.g., approximating the kinetic energy functional or finding the Pauli potential). We would like to emphasize that one of the most important aspects of our results is that the $\star$ product gives a rich structure for extracting the corresponding one-particle Wigner function. In that sense, quasi-DFT is a functional theory that can connect directly with DFT and with semiclassical expansions of the many-body problem. There are several potential research directions from our results: First, one could develop machine-learning quasi-DFT functionals, which is now current practice for standard DFT [77–80]. Second, since Wigner negativities carry important quantum information it will be interesting to see what information they can unveil for electronic correlations [81,82] or fermionic entanglement [83]. Finally, it could be quite promising to tackle—within this orbital-free framework—quantum excitations in the same spirit of state-average calculations [84] or the recently formulated **w**-1-RDMFT [85].

ACKNOWLEDGMENTS

I gratefully thank L. Colmenarez, J. Liebert, E. Kraisler, and J. Maki for insightful discussions and for providing important comments on the manuscript. I also thank Ana Maria Rey and the warm atmosphere of her group at JILA where this paper took its final shape. This work is funded by the European Union's Horizon Europe Research and Innovation program under Marie Skłodowska-Curie Grant No. 101065295—RDMFTforbosons. Views and opinions expressed are however those of the author only and do not necessarily reflect those of the European Union or the European Research Executive Agency.

APPENDIX A: DISCRETE WIGNER FORMALISM FOR THE HUBBARD MODEL

Here we apply the discrete Wigner formalism to the Hubbard model of L sites. This is defined in the L-dimensional Hilbert space $\mathcal{H}^L$ whose position basis is $\mathcal{S} = \{|1\rangle,\dots,|L\rangle\}$. Another orthonormal basis for the same Hilbert space is $\{|\phi_0\rangle,\dots,|\phi_{L-1}\rangle\}$, defined by the Fourier transform:

$$|\phi_m\rangle = \frac{1}{\sqrt{L}}\sum_{n=1}^{L} e^{in\phi_m}|n\rangle, \tag{A1}$$

with $\phi_m = \frac{2\pi}{L}m$. The set of pairs $\{n,\phi_m\}_{n,m}$ constitutes an $L\times L$ grid. This is the phase space Γ^L associated with the Hilbert space $\mathcal{H}^L$ [74].

The operators

$$\hat{n} = \sum_n n|n\rangle\langle n| \tag{A2}$$

and $\hat{\phi} = \sum_m \phi_m|\phi_m\rangle\langle\phi_m|$ can be used to construct the following unitary operators:

$$\hat{V} = \exp\left(i\frac{2\pi}{L}\hat{n}\right) \quad \text{and} \quad \hat{U} = \exp(i\hat{\phi}), \tag{A3}$$

which satisfy the Weyl relation [76]

$$\hat{D}(k,l) \equiv \exp\left(-i\frac{\pi kl}{L}\right)\hat{U}^k\hat{V}^l = \exp\left(i\frac{\pi kl}{L}\right)\hat{U}^l\hat{V}^k, \tag{A4}$$

for $k,l\in\mathbb{Z}$. With this operator, the authors of Ref. [76] define the phase-space point operator as

$$\hat{\Omega}_\kappa(n,\phi_m) = \frac{1}{L}\sum_{k,l}\kappa(k,l)\hat{D}(k,l)e^{-i\left(k\phi_m+\frac{2\pi}{L}ln\right)}, \tag{A5}$$

with a kernel $\kappa(k,l)$, whose properties are determined by the properties of $\hat{\Omega}_\kappa$. For instance, by the operator's hermiticity condition, $\hat{\Omega}_\kappa(n,\phi_m) = \hat{\Omega}^\dagger_\kappa(n,\phi_m)$ that holds for all $(n,\phi_m)\in\Gamma^L$, which is needed to map phase-space functions to Hermitian operators, results in the condition $\kappa^*(k,l) = (-1)^{L+k+l}\kappa(L-k,L-l)$. For odd $L = 2N+1$, for instance, a kernel can be chosen to be [76] $\kappa(k,l) = \cos(\pi kl/L)$.

The map between $f(n,\phi_m)$, a real function in Γ^L, and $\hat{f}$, an operator in $\mathcal{H}^L$, is realized by means of the following relation:

$$\hat{f} = \frac{1}{L}\sum_{m,n} f(n,\phi_m)\hat{\Omega}_\kappa(n,\phi_m). \tag{A6}$$

Equation (A6) can now be used to find the Wigner quasidistribution. Since the average value of the observable represented by the operator $\hat{f}$ in a state defined by the density operator $\hat{\gamma}$ reads

$$\mathrm{Tr}[\hat{\gamma}\hat{f}] = \frac{1}{L}\sum_{m,n} f(n,\phi_m)\mathrm{Tr}[\hat{\gamma}\hat{\Omega}_\kappa(n,\phi_m)], \tag{A7}$$

a natural definition for the Wigner quasiprobability (for the kernel κ) arises: $\omega(n,\phi_m) = \mathrm{Tr}[\hat{\gamma}\hat{\Omega}_\kappa(n,\phi_m)]$. From this definition, one can write

$$\omega(n,\phi_m)=\sum_{m',n'}\mathcal{D}(n,\phi_m;n',m')\langle n'|\hat{\gamma}|m'\rangle, \tag{A8}$$

where

$$\begin{aligned}\mathcal{D}(n,\phi_m;n',m')\\ &=\frac{1}{L^2}\sum_{k,l,s}\kappa(k,l)e^{i\left(\frac{\pi kl}{L}+k\phi_s+n'\phi_{s+l}-m'\phi_s-k\phi_m-\frac{2\pi}{L}ln\right)}.\end{aligned}$$

If one vectorizes both ω and γ, to wit,

$$|\omega\rangle\rangle=\begin{pmatrix}\omega(1,\phi_0)\\ \omega(1,\phi_1)\\ \omega(1,\phi_2)\\ \vdots\end{pmatrix}\quad\text{and}\quad|\gamma\rangle\rangle=\begin{pmatrix}\langle 1|\gamma|1\rangle\\ \langle 1|\gamma|2\rangle\\ \langle 1|\gamma|3\rangle\\ \vdots\end{pmatrix}, \tag{A9}$$

one can formally write Eq. (A8) as $|\omega\rangle\rangle=\hat{\mathcal{D}}|\gamma\rangle\rangle$.

APPENDIX B: HARTREE-FOCK IN PHASE SPACE

In this last section, we investigate the form of the orbital-free Hartree-Fock equations in phase space for a system of N electrons. As the respective wave function is a single Slater determinant, the one-body reduced-density matrix is a projector:

$$\gamma(\mathbf{r},\mathbf{r}')=\sum_{n=1}^{N}\varphi_n(\mathbf{r})\varphi_n^*(\mathbf{r}'),$$

with $\int\varphi_n(\mathbf{r})\varphi_m^*(\mathbf{r})d^3\mathbf{r}=\delta_{nm}$. The first result we prove is that the corresponding Wigner function satisfies $\omega\star\omega=\omega$.

Proof. Let us first define the Wigner phase-space orbitals $\chi_n(\mathbf{r},\mathbf{p})=\int\varphi_n(\mathbf{r}-\mathbf{z})\varphi_n^*(\mathbf{r}+\mathbf{z})e^{2i\mathbf{p}\cdot\mathbf{z}}d^3\mathbf{z}$. They satisfy the following equation:

$$\begin{aligned}&\chi_n(\mathbf{r},\mathbf{p})\star\chi_m(\mathbf{r},\mathbf{p})\\
&=\int\chi_n(\mathbf{r}',\mathbf{p}')\,\chi_m(\mathbf{r}'',\mathbf{p}'')e^{2i(\mathbf{r}\cdot\mathbf{p}'-\mathbf{r}'\cdot\mathbf{p}+\mathbf{r}'\cdot\mathbf{p}''-\mathbf{r}''\cdot\mathbf{p}'+\mathbf{r}''\cdot\mathbf{p}-\mathbf{r}\cdot\mathbf{p}'')}\,d\Omega'\,d\Omega''\\
&=\int\varphi_n(\mathbf{r}'-\mathbf{z}')\,\varphi_n^*(\mathbf{r}'+\mathbf{z}')\,\varphi_m(\mathbf{r}''-\mathbf{z}'')\,\varphi_m^*(\mathbf{r}''+\mathbf{z}'')\,e^{2i(\mathbf{r}''-\mathbf{r}')\cdot\mathbf{p}}e^{2i(\mathbf{z}'+\mathbf{r}-\mathbf{r}'')\cdot\mathbf{p}'}e^{2i(\mathbf{z}''+\mathbf{r}'-\mathbf{r})\cdot\mathbf{p}''}d\Omega'\,d\Omega''\,d^3\mathbf{z}'\,d^3\mathbf{z}''\\
&=\int\varphi_n(\mathbf{r}'-\mathbf{z}')\,\varphi_n^*(\mathbf{r}'+\mathbf{z}')\,\varphi_m(\mathbf{r}''-\mathbf{z}'')\,\varphi_m^*(\mathbf{r}''+\mathbf{z}'')e^{2i(\mathbf{r}''-\mathbf{r}')\cdot\mathbf{p}}\,\delta(\mathbf{z}'+\mathbf{r}-\mathbf{r}'')\,\delta(\mathbf{z}''+\mathbf{r}'-\mathbf{r})\,d^3\mathbf{r}'\,d^3\mathbf{r}''\,d^3\mathbf{z}'\,d^3\mathbf{z}''\\
&=\int\varphi_n(\mathbf{r}'+\mathbf{r}-\mathbf{r}'')\,\varphi_n^*(\mathbf{r}'-\mathbf{r}+\mathbf{r}'')\,\varphi_m(\mathbf{r}''+\mathbf{r}'-\mathbf{r})\,\varphi_m^*(\mathbf{r}''-\mathbf{r}'+\mathbf{r})e^{2i(\mathbf{r}''-\mathbf{r}')\cdot\mathbf{p}}\,d^3\mathbf{r}'d^3\mathbf{r}''.\end{aligned}$$

Letting $\mathbf{u}=\mathbf{r}''-\mathbf{r}'$ and $\mathbf{v}=\mathbf{r}'+\mathbf{r}''-\mathbf{r}$, we have

$$\chi_n(\mathbf{r},\mathbf{p})\star\chi_m(\mathbf{r},\mathbf{p})=\int\varphi_n(\mathbf{r}-\mathbf{u})\,\varphi_m^*(\mathbf{r}+\mathbf{u})\,e^{2i\mathbf{u}\cdot\mathbf{p}}d^3\mathbf{u}\int\varphi_m(\mathbf{v})\,\varphi_n^*(\mathbf{v})\,d^3\mathbf{v}=\delta_{nm}\chi_n(\mathbf{r},\mathbf{p}).$$

As a consequence,

$$\omega(\mathbf{r},\mathbf{p})\star\omega(\mathbf{r},\mathbf{p})=\sum_{nm}\chi_n(\mathbf{r},\mathbf{p})\star\chi_m(\mathbf{r},\mathbf{p})=\sum_{nm}\chi_n(\mathbf{r},\mathbf{p})\delta_{nm}=\omega(\mathbf{r},\mathbf{p}). \tag{B1}$$

■

This result indicates that we have to solve the Hartree-Fock functional subject to the condition $\omega\star\omega=\omega$ and the normalization $\int\omega(\mathbf{r},\mathbf{p})d\Omega=N$. Using the Lagrange multipliers $\alpha(\mathbf{r},\mathbf{p})$ and β, the variational problem reads as follows:

$$\delta\left\{\mathcal{E}_{\rm HF}[\omega]-\int\alpha(\mathbf{r},\mathbf{p})[\omega(\mathbf{r},\mathbf{p})\star\omega(\mathbf{r},\mathbf{p})-\omega(\mathbf{r},\mathbf{p})]\,d\Omega-\beta\left[\int\omega(\mathbf{r},\mathbf{p})\,d\Omega-N\right]\right\}=0. \tag{B2}$$

Before performing the variation note that

$$\begin{aligned}\frac{\delta}{\delta\omega(\mathbf{r},\mathbf{p})}\int\alpha(\mathbf{r},\mathbf{p})\,\omega(\mathbf{r},\mathbf{p})\star\omega(\mathbf{r},\mathbf{p})\,d\Omega&=\frac{\delta}{\delta\omega(\mathbf{r},\mathbf{p})}\int\alpha(\mathbf{r},\mathbf{p})\,\omega(\mathbf{r}',\mathbf{p}')\,\omega(\mathbf{r}'',\mathbf{p}'')e^{2i(\mathbf{r}\cdot\mathbf{p}'-\mathbf{r}'\cdot\mathbf{p}+\mathbf{r}'\cdot\mathbf{p}''-\mathbf{r}''\cdot\mathbf{p}'+\mathbf{r}''\cdot\mathbf{p}-\mathbf{r}\cdot\mathbf{p}'')}\,d\Omega\,d\Omega'\,d\Omega''\\
&=\int\alpha(\mathbf{r}',\mathbf{p}')\,\omega(\mathbf{r}'',\mathbf{p}'')e^{2i(\mathbf{r}'\cdot\mathbf{p}-\mathbf{r}\cdot\mathbf{p}'+\mathbf{r}\cdot\mathbf{p}''-\mathbf{r}''\cdot\mathbf{p}+\mathbf{r}''\cdot\mathbf{p}'-\mathbf{r}'\cdot\mathbf{p}'')}\,d\Omega'\,d\Omega''\\
&\quad+\int\alpha(\mathbf{r}'',\mathbf{p}'')\,\omega(\mathbf{r}',\mathbf{p}')e^{2i(\mathbf{r}''\cdot\mathbf{p}'-\mathbf{r}'\cdot\mathbf{p}''+\mathbf{r}'\cdot\mathbf{p}-\mathbf{r}\cdot\mathbf{p}'+\mathbf{r}\cdot\mathbf{p}''-\mathbf{r}''\cdot\mathbf{p})}\,d\Omega'\,d\Omega''\\
&=\omega(\mathbf{r},\mathbf{p})\star\alpha(\mathbf{r},\mathbf{p})+\alpha(\mathbf{r},\mathbf{p})\star\omega(\mathbf{r},\mathbf{p}).\end{aligned} \tag{B3}$$

Using this result in Eq. (B2), we obtain

$$L(\mathbf{r},\mathbf{p})\equiv f_{\rm HF}(\mathbf{r},\mathbf{p})-\omega(\mathbf{r},\mathbf{p})\star\alpha(\mathbf{r},\mathbf{p})-\alpha(\mathbf{r},\mathbf{p})\star\omega(\mathbf{r},\mathbf{p})+\alpha(\mathbf{r},\mathbf{p})-\beta=0, \tag{B4}$$

where $f_{\rm HF}(\mathbf{r},\mathbf{p})=\delta\mathcal{E}_{\rm HF}[\omega]/\delta\omega(\mathbf{r},\mathbf{p})$. Multiplying (with the $\star$ product) this equation on the left by $\omega(\mathbf{r},\mathbf{p})$ [i.e, $\omega(\mathbf{r},\mathbf{p})\star L(\mathbf{r},\mathbf{p})$] and on the right [i.e., $L(\mathbf{r},\mathbf{p})\star\omega(\mathbf{r},\mathbf{p})$], and then subtracting both equations, we obtain that $\omega\,\star$ anticommutes with $f_{\rm HF}(\mathbf{r},\mathbf{p})$:

$$[f_{\rm HF}(\mathbf{r},\mathbf{p}),\omega(\mathbf{r},\mathbf{p})]_\star\equiv f_{\rm HF}(\mathbf{r},\mathbf{p})\star\omega(\mathbf{r},\mathbf{p})-\omega(\mathbf{r},\mathbf{p})\star f_{\rm HF}(\mathbf{r},\mathbf{p})=0. \tag{B5}$$

This is the equation of $\omega(\mathbf{r},\mathbf{p})$ within Hartree-Fock theory. Recall that it admits an expansion in $\hbar$. For this reason, this equation allows a semiclassical expansion that does not exist in the double-coordinate representation.

To finish the calculation we give now the explicit form of $f_{\mathrm{HF}}(\mathbf{r},\mathbf{p})$. Let us define the one-particle Hamiltonian $h(\mathbf{r},\mathbf{p}) = \mathbf{p}^2/2m + v(\mathbf{r})$, with $v(\mathbf{r})$ being the external potential. Using the inverse of the Wigner transformation, the Hartree-Fock energy reads

$$\mathcal{E}_{\mathrm{HF}}[\omega] = \int h(\mathbf{r},\mathbf{p})\,\omega(\mathbf{r},\mathbf{p})\,d\Omega + \frac{1}{2}\int \frac{\omega(\mathbf{r},\mathbf{p})\,\omega(\mathbf{r}',\mathbf{p}')}{|\mathbf{r}-\mathbf{r}'|}d\Omega\,d\Omega' - \frac{1}{2}\int e^{i(\mathbf{p}-\mathbf{p}')\cdot(\mathbf{r}-\mathbf{r}')}\frac{\omega((\mathbf{r}+\mathbf{r}')/2,\mathbf{p})\,\omega[(\mathbf{r}+\mathbf{r}')/2,\mathbf{p}']}{|\mathbf{r}-\mathbf{r}'|}d\Omega\,d\Omega'.$$

A straightforward calculation finally gives

$$\begin{aligned} f_{\mathrm{HF}}(\mathbf{r},\mathbf{p}) &= h(\mathbf{r},\mathbf{p}) + \int \frac{\omega(\mathbf{r}',\mathbf{p}')}{|\mathbf{r}-\mathbf{r}'|}\,d\Omega' - \int \frac{e^{i(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}'}}{|\mathbf{r}'|}\omega(\mathbf{r},\mathbf{p}')\,d\Omega' \\ &= h(\mathbf{r},\mathbf{p}) + \int \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\,d\mathbf{r}' - \int \frac{\omega(\mathbf{r},\mathbf{p}')}{|\mathbf{p}-\mathbf{p}'|^2}\,d\mathbf{p}'. \end{aligned} \tag{B6}$$

---

[1] J. R. Friedman, V. Patel, W. Chen, S. K. Tolpygo, and J. E. Lukens, Quantum superposition of distinct macroscopic states, Nature (London) **406**, 43 (2000).

[2] Y. Y. Fein, P. Geyer, P. Zwick, F. Kiałka, S. Pedalino, M. Mayor, S. Gerlich, and M. Arndt, Quantum superposition of molecules beyond 25 kDa, Nat. Phys. **15**, 1242 (2019).

[3] E. A. Wollack, A. Y. Cleland, R. G. Gruenke, Z. Wang, P. Arrangoiz-Arriola, and A. H. Safavi-Naeini, Quantum state preparation and tomography of entangled mechanical resonators, Nature (London) **604**, 463 (2022).

[4] B. Schrinski, Y. Yang, U. von Lüpke, M. Bild, Y. Chu, K. Hornberger, S. Nimmrichter, and M. Fadel, Macroscopic quantum test with bulk acoustic wave resonators, Phys. Rev. Lett. **130**, 133604 (2023).

[5] M. Walschaers, C. Fabre, V. Parigi, and N. Treps, Entanglement and Wigner function negativity of multimode non-Gaussian states, Phys. Rev. Lett. **119**, 183601 (2017).

[6] J. P. Dahl, H. Mack, A. Wolf, and W. P. Schleich, Entanglement versus negative domains of Wigner functions, Phys. Rev. A **74**, 042323 (2006).

[7] A. Kenfack and K. Życzkowski, Negativity of the Wigner function as an indicator of non-classicality, J. Opt. B: Quantum Semiclassical Opt. **6**, 396 (2004).

[8] R. W. Spekkens, Negativity and contextuality are equivalent notions of nonclassicality, Phys. Rev. Lett. **101**, 020401 (2008).

[9] R. Raussendorf, D. E. Browne, N. Delfosse, C. Okay, and J. Bermejo-Vega, Contextuality and Wigner-function negativity in qubit quantum computation, Phys. Rev. A **95**, 052334 (2017).

[10] M. Howard, J. Wallman, V. Veitch, and J. Emerson, Contextuality supplies the ‘magic’ for quantum computation, Nature (London) **510**, 351 (2014).

[11] V. Veitch, C. Ferrie, D. Gross, and J. Emerson, Negative quasi-probability as a resource for quantum computation, New J. Phys. **14**, 113011 (2012).

[12] M. Walschaers, V. Parigi, and N. Treps, Practical framework for conditional non-Gaussian quantum state preparation, PRX Quantum **1**, 020305 (2020).

[13] M. Walschaers, On quantum steering and Wigner negativity, Quantum **7**, 1038 (2023).

[14] R. Howl, V. Vedral, D. Naik, M. Christodoulou, C. Rovelli, and A. Iyer, Non-Gaussianity as a signature of a quantum theory of gravity, PRX Quantum **2**, 010325 (2021).

[15] P. Hohenberg and W. Kohn, Inhomogeneous electron gas, Phys. Rev. **136**, B864 (1964).

[16] R. O. Jones, Density functional theory: Its origins, rise to prominence, and future, Rev. Mod. Phys. **87**, 897 (2015).

[17] A. M. Teale *et al.*, DFT exchange: sharing perspectives on the workhorse of quantum chemistry and materials science, Phys. Chem. Chem. Phys. **24**, 28700 (2022).

[18] B. Huang, G. F. von Rudorff, and O. A. von Lilienfeld, The central role of density functional theory in the AI age, Science **381**, 170 (2023).

[19] F. Aikebaier, T. Ojanen, and J. Lado, Extracting electronic many-body correlations from local measurements with artificial neural networks, SciPost Phys. Core **6**, 030 (2023).

[20] J. R. Moreno, G. Carleo, and A. Georges, Deep learning the Hohenberg-Kohn maps of density functional theory, Phys. Rev. Lett. **125**, 076402 (2020).

[21] P. Blanchard, J. M. Gracia-Bondía, and J. C. Várilly, Density functional theory on phase space, Int. J. Quantum Chem. **112**, 1134 (2012).

[22] K. Ryczko, S. J. Wetzel, R. G. Melko, and I. Tamblyn, Toward orbital-free density functional theory with small data sets and deep learning, J. Chem. Theory Comput. **18**, 1122 (2022).

[23] K. Jiang, X. Shao, and M. Pavanello, Efficient time-dependent orbital-free density functional theory: Semilocal adiabatic response, Phys. Rev. B **106**, 115153 (2022).

[24] W. Kohn and L. J. Sham, Self-consistent equations including exchange and correlation effects, Phys. Rev. **140**, A1133 (1965).

[25] E. Engel and R.M. Dreizler, *Density Functional Theory: An Advanced Course*, Theoretical and Mathematical Physics (Springer, Berlin, 2011).

[26] T. L. Gilbert, Hohenberg-Kohn theorem for nonlocal external potentials, Phys. Rev. B **12**, 2111 (1975).

[27] R. A. Donnelly and R. G. Parr, Elementary properties of an energy functional of the first-order reduced density matrix, J. Chem. Phys. **69**, 4431 (1978).

[28] K. Pernal and K. J. H. Giesbertz, Reduced density matrix functional theory (RDMFT) and linear response time-dependent RDMFT (TD-RDMFT), in *Density-Functional Methods for Excited States*, edited by Nicolas Ferré, Michael Filatov, and Miquel Huix-Rotllant (Springer, Cham, 2016), p. 125.

[29] C. Schilling, Communication: Relating the pure and ensemble density matrix functional, J. Chem. Phys. **149**, 231102 (2018).

[30] M. Piris, Global method for electron correlation, Phys. Rev. Lett. **119**, 063002 (2017).

[31] J. Schmidt, C. L. Benavides-Riveros, and M. A. L. Marques, Reduced density matrix functional theory for superconductors, Phys. Rev. B **99**, 224502 (2019).

[32] C. L. Benavides-Riveros, J. Wolff, M. A. L. Marques, and C. Schilling, Reduced density matrix functional theory for Bosons, Phys. Rev. Lett. **124**, 180603 (2020).

[33] J. Liebert and C. Schilling, Functional theory for Bose-Einstein condensates, Phys. Rev. Res. **3**, 013282 (2021).

[34] T. Maciażek, Repulsively diverging gradient of the density functional in the reduced density matrix functional theory, New J. Phys. **23**, 113006 (2021).

[35] J. Liebert and C. Schilling, An exact one-particle theory of bosonic excitations: from a generalized Hohenberg-Kohn theorem to convexified N-representability, New J. Phys. **25**, 013009 (2023).

[36] M. Rodríguez-Mayorga, K. J.H. Giesbertz, and L. Visscher, Relativistic reduced density matrix functional theory, SciPost Chem. **1**, 004 (2022).

[37] S. Sharma, J. K. Dewhurst, S. Shallcross, and E. K. U. Gross, Spectral density and metal-insulator phase transition in Mott insulators within reduced density matrix functional theory, Phys. Rev. Lett. **110**, 116403 (2013).

[38] X. Shao, L. Paetow, M. E. Tuckerman, and M. Pavanello, Machine learning electronic structure methods based on the one-electron reduced density matrix, Nat. Commun. **14**, 6281 (2023).

[39] M. Piris and K. Pernal, Comment on "Generalization of the Kohn-Sham system that can represent arbitrary one-electron density matrices," Phys. Rev. A **96**, 046501 (2017).

[40] R. Gebauer, M. H. Cohen, and R. Car, A well-scaling natural orbital theory, Proc. Natl. Acad. Sci. USA **113**, 12913 (2016).

[41] M. Piris and J. M. Ugalde, Iterative diagonalization for orbital optimization in natural orbital functional theory, J. Comput. Chem. **30**, 2078 (2009).

[42] K. Pernal, Effective potential for natural spin orbitals, Phys. Rev. Lett. **94**, 233002 (2005).

[43] P.-O. Löwdin and H. Shull, Natural orbitals in the quantum theory of two-electron systems, Phys. Rev. **101**, 1730 (1956).

[44] M. Rodríguez-Mayorga, I. Mitxelena, F. Bruneval, and M. Piris, Coupling natural orbital functional theory and many-body perturbation theory by using nondynamically correlated canonical orbitals, J. Chem. Theory Comput. **17**, 7562 (2021).

[45] E. Wigner, On the quantum correction for thermodynamic equilibrium, Phys. Rev. **40**, 749 (1932).

[46] J. E. Moyal, Quantum mechanics as a statistical theory, Math. Proc. Cambridge Philos. Soc. **45**, 99 (1949).

[47] H. Weyl, Quantenmechanik und gruppentheorie, Z. Phys. **46**, 1 (1927).

[48] To ease the notation we skip from now on the spin indices in ω.

[49] A. J. Coleman, Structure of fermion density matrices, Rev. Mod. Phys. **35**, 668 (1963).

[50] In the literature, there are other representability conditions for the spin coordinates [86].

[51] J.-D. Chai and J. D. Weeks, Orbital-free density functional theory: Kinetic potentials and *ab initio* local pseudopotentials, Phys. Rev. B **75**, 205122 (2007).

[52] S. S. Iyengar, M. Ernzerhof, S. N. Maximoff, and G. E. Scuseria, Challenge of creating accurate and effective kinetic-energy functionals, Phys. Rev. A **63**, 052508 (2001).

[53] K. Finzel, Local conditions for the Pauli potential in order to yield self-consistent electron densities exhibiting proper atomic shell structure, J. Chem. Phys. **144**, 034108 (2016).

[54] E. Kraisler and A. Schild, Discontinuous behavior of the Pauli potential in density functional theory as a function of the electron number, Phys. Rev. Res. **2**, 013159 (2020).

[55] M. Levy, J. P. Perdew, and V. Sahni, Exact differential equation for the density and ionization energy of a many-particle system, Phys. Rev. A **30**, 2745 (1984).

[56] N. H. March, The local potential determining the square root of the ground-state electron density of atoms and molecules from the Schrödinger equation, Phys. Lett. A **113**, 476 (1986).

[57] A. Polkovnikov, Phase space representation of quantum dynamics, Ann. Phys. **325**, 1790 (2010).

[58] M. Piris, A new approach for the two-electron cumulant in natural orbital functional theory, Int. J. Quantum Chem. **106**, 1093 (2006).

[59] J. Cioslowski, C. Schilling, and R. Schilling, 1-Matrix functional for long-range interaction energy of two hydrogen atoms, J. Chem. Phys. **158**, 084106 (2023).

[60] C. L. Benavides-Riveros and M. A. L. Marques, On the time evolution of fermionic occupation numbers, J. Chem. Phys. **151**, 044112 (2019).

[61] M. Piris, Global natural orbital functional: Towards the complete description of the electron correlation, Phys. Rev. Lett. **127**, 233001 (2021).

[62] A. M. K. Müller, Explicit approximate relation between reduced two- and one-particle density matrices, Phys. Lett. A **105**, 446 (1984).

[63] K. J. H. Giesbertz and E. J. Baerends, Aufbau derived from a unified treatment of occupation numbers in Hartree-Fock, Kohn-Sham, and natural orbital theories with the Karush-Kuhn-Tucker conditions for the inequality constraints $n_i \leqslant 1$ and $n_i \geqslant 0$, J. Chem. Phys. **132**, 194108 (2010).

[64] J. Wang and E. J. Baerends, Self-consistent-field method for correlated many-electron systems with an entropic cumulant energy, Phys. Rev. Lett. **128**, 013001 (2022).

[65] J. Schmidt, M. Fadel, and C. L. Benavides-Riveros, Machine learning universal bosonic functionals, Phys. Rev. Res. **3**, L032063 (2021).

[66] D. Gibney, J.-N. Boyn, and D. A. Mazziotti, Density functional theory transformed into a one-electron reduced-density-matrix functional theory for the capture of static correlation, J. Phys. Chem. Lett. **13**, 1382 (2022).

[67] M. Piris, Dynamic electron-correlation energy in the natural-orbital-functional second-order-Møller-Plesset method from the orbital-invariant perturbation theory, Phys. Rev. A **98**, 022504 (2018).

[68] A. J. Cohen and P. Mori-Sánchez, Landscape of an exact energy functional, Phys. Rev. A **93**, 042511 (2016).

[69] C. Schilling and R. Schilling, Diverging exchange force and form of the exact density matrix functional, Phys. Rev. Lett. **122**, 013001 (2019).

[70] K. Burke and J. Kozlowski, Lies my teacher told me about density functional theory: Seeing through them with the Hubbard dimer, in *Simulating Correlations with Computers Lecture Notes of the Autumn School on Correlated Electrons 2021*, edited

by E. Pavarini and E. Koch (Forschungszentrum Jülich GmbH, Institute for Advanced Simulation, 2021).

[71] S. Di Sabatino, P.-F. Loos, and P. Romaniello, Scrutinizing GW-based methods using the hubbard dimer, Front. Chem. **9**, 751054 (2021).

[72] J. Liebert, A. Y. Chaou, and C. Schilling, Refining and relating fundamentals of functional theory, J. Chem. Phys. **158**, 214108 (2023).

[73] R. Feynman, Negative probabilities, in *Quantum Implications: Essays in Honour of David Bohm*, edited by B. Hiley and F. Peat (Routledge, 1987), p. 235.

[74] W. K. Wootters, A Wigner-function formulation of finite-state quantum mechanics, Ann. Phys. **176**, 1 (1987).

[75] J. Schachenmayer, A. Pikovski, and A. M. Rey, Many-body quantum spin dynamics with Monte Carlo trajectories on a discrete phase space, Phys. Rev. X **5**, 011022 (2015).

[76] M. Przanowski and J. Tosiek, From the discrete Weyl-Wigner formalism for symmetric ordering to a number-phase Wigner function, J. Math. Phys. **58**, 102106 (2017).

[77] A. Grisafi, A. M. Lewis, M. Rossi, and M. Ceriotti, Electronic-structure properties from atom-centered predictions of the electron density, J. Chem. Theory Comput. **19**, 4451 (2022).

[78] J. Kirkpatrick *et al.*, Pushing the frontiers of density functionals by solving the fractional electron problem, Science **374**, 1385 (2021).

[79] H. Ma, A. Narayanaswamy, P. Riley, and L. Li, Evolving symbolic density functionals, Sci. Adv. **8**, eabq0279 (2022).

[80] J. Gedeon, J. Schmidt, M. J. P. Hodgson, J. Wetherell, C. L. Benavides-Riveros, and M. A. L. Marques, Machine learning the derivative discontinuity of density-functional theory, Mach. Learn.: Sci. Technol. **3**, 015011 (2022).

[81] R. Izsák, A. V. Ivanov, N. S. Blunt, N. Holzmann, and F. Neese, Measuring electron correlation: The impact of symmetry and orbital transformations, J. Chem. Theory Comput. **19**, 2703 (2023).

[82] D. Zhang and D. G. Truhlar, Decomposition of the electronic energy in terms of density, density coherence, and the connected part of the two-body reduced density matrix, J. Chem. Theory Comput. **17**, 5733 (2021).

[83] K. Ptaszyński and M. Esposito, Fermionic one-body entanglement as a thermodynamic resource, Phys. Rev. Lett. **130**, 150201 (2023).

[84] C. L. Benavides-Riveros, L. Chen, C. Schilling, S. Mantilla, and S. Pittalis, Excitations of quantum many-body systems via purified ensembles: A unitary-coupled-cluster-based approach, Phys. Rev. Lett. **129**, 066401 (2022).

[85] C. Schilling and S. Pittalis, Ensemble reduced density matrix functional theory for excited states and hierarchical generalization of Pauli's exclusion principle, Phys. Rev. Lett. **127**, 023001 (2021).

[86] C. L. Benavides-Riveros and J. M. Gracia-Bondía, Physical Wigner functions, Phys. Rev. A **87**, 022118 (2013).