Diverging exchange force and form of the exact density matrix functional

Christian Schilling$^{1,*}$ and Rolf Schilling$^{2,†}$

$^1$Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom
$^2$Institut für Physik, Johannes Gutenberg-Universität, D-55099 Mainz, Germany

(Dated: January 3, 2019)

For translationally invariant one-band lattice models, we exploit the \textit{ab initio} knowledge of the natural orbitals to simplify reduced density matrix functional theory (RDMFT). Striking underlying features are discovered: First, within each symmetry sector, the interaction functional $\mathcal{F}$ depends only on the natural occupation numbers $n$. The respective sets $\mathcal{P}_N^1$ and $\mathcal{E}_N^1$ of pure and ensemble $N$-representable one-matrices coincide. Second, and most importantly, the exact functional is strongly shaped by the geometry of the polytope $\mathcal{E}_N^1 \equiv \mathcal{P}_N^1$, described by linear constraints $D^{(i)}(n) \geq 0$. For smaller systems, it follows as $\mathcal{F}[n] = \sum_{i,i'} \bar{V}_{i,i'} \sqrt{D^{(i)}(n)D^{(i')}(n)}$. This generalizes to systems of arbitrary size by replacing each $D^{(i)}$ by a linear combination of $\{D^{(i)}(n)\}$ and adding a non-analytical term involving the interaction $\bar{V}$. Third, the gradient $d\mathcal{F}/dn$ is shown to diverge on the boundary $\partial \mathcal{E}_N^1$, suggesting that the fermionic exchange symmetry manifests itself within RDMFT in the form of an “exchange force”. All findings hold for systems with non-fixed particle number as well and $\bar{V}$ can be any $p$-particle interaction. As an illustration, we derive the exact functional for the Hubbard square.

\textbf{Introduction.} — Reduced density matrix functional theory (RDMFT) \cite{1–5} has the potential of overcoming the shortcomings and fundamental limitations of the widely used density functional theory (DFT) \cite{6–9}. Involving the full one-particle reduced density matrix (1RDM) $\gamma$ facilitates not only an exact description of the single particle potential energy, $U[\gamma] \equiv \text{Tr}[U\gamma]$, but also of the kinetic energy, $T[\gamma] \equiv \text{Tr}[T\gamma]$. It remains to derive accurate approximations to the interaction term $\mathcal{F}[\gamma]$. Moreover, RDMFT allows explicitly for fractional occupation numbers as it is required in the description of strongly correlated systems \cite{4}. At the same time, involving the full 1RDM lies, however, also at the heart of possible disadvantages of RDMFT relative to DFT: While both methods avoid the use of exponentially complex $N$-electron wave functions, the 1RDM involves $d^2$ degrees of freedom compared to $d$ for the spatial density used in DFT, where $d$ is the basis set size. To be more specific, one often uses the spectral representation $\gamma \equiv \sum_\nu n_\nu |\varphi_\nu\rangle \langle \varphi_\nu|$ and then minimizes the total energy functional $\mathcal{E}[\gamma] = T[\gamma] + U[\gamma] + \mathcal{F}[\gamma]$ with respect to the natural occupation numbers (NONs) $n_\nu$ and natural orbitals $|\varphi_\nu\rangle$, separately. The dependence on the latter makes the minimization of $\mathcal{E}$ particularly difficult and one often encounters slow convergence (see, e.g., \cite{10}).

The general situation drastically changes in favour of RDMFT for the important class of periodic one-band lattice systems as studied in solid state physics. The 1RDM inherits the translational symmetry of the ground state \cite{11} and the natural orbitals are known from the very beginning. They are given for all systems by plane waves (multiplied by some spin state). Thus, various possible disadvantages of RDMFT compared to DFT disappear and RDMFT simplifies \textit{de facto} to a NON-functional theory.

Based on this observation and the fact that in general the significance of symmetries in physics can hardly be overestimated, we will explore in this letter the role of the translational symmetry within RDMFT and reveal universal and far-reaching consequences. In that sense, our work complements previous studies of the homogeneous electron gas \cite{12–16}, periodic polymers \cite{17,18} and of lattice systems \cite{19–40} in which the crucial role of symmetries was not further explored. In particular, we determine the sets $\mathcal{P}_N^1$ and $\mathcal{E}_N^1$ of pure and ensemble $N$-representable 1RDMs and show that they coincide. Then, in the form of an analytic derivation, we discover the general form of the exact functional $\mathcal{F}$ which will illustrate the fundamental role of one-body $N$-representability constraints. Finally, we show that the fermionic exchange symmetry manifests itself within RDMFT in the form of an “exchange force” which diverges on the boundary $\partial \mathcal{E}_N^1$ of the polytope $\mathcal{E}_N^1 = \mathcal{P}_N^1$. All those universal features will be illustrated in two lattice cluster systems.

\textbf{One-body $N$-representability constraints.} — We consider translationally invariant systems of $N$ electrons on a one-band lattice in $D$ dimensions with periodic boundary conditions and $L$ sites in each direction. Due to the translational invariance, the symmetry-adapted “orbital” part of the one-electron states are plane waves with momenta $\vec{k} = (2\pi/L)(\nu_1,\ldots,\nu_D) \equiv (2\pi/L)\vec{\nu}$, where $\nu_i = 0, 1, \ldots, L - 1$. The spin-orbitals follow as $|\varphi_{m}\rangle$ ($m = \pm \frac{1}{2}$) and we introduce for the following the collective quantum number $q \equiv (i\vec{m})$. On the $N$-fermion level, a symmetry-adapted basis is then given by the Slater determinants $|q\rangle \equiv (q_1,\ldots,q_N)$. The translational and spin symmetries decompose the $N$-fermion Hilbert space $\mathcal{H}$ into irreducible sectors $\mathcal{H}^{(Q)}$, $Q \equiv (\vec{K},M_z)$, each of which is spanned by the Slater determinants $\{ |q\rangle \}_{q \in \mathcal{X}^{(Q)}}$ with total momentum $\vec{K} = \sum_{n=1}^{N} \vec{k}_n$ and magnetization $M_z$. 

\textbf{Acknowledgments.} — We are grateful to Christian Schilling and \textit{ab initio} methods for their invaluable discussions.

\textbf{References.}
\[ M_n = \sum_{q=1}^{N} n_m. \] The respective set of configurations \( q \) is denoted by \( \mathcal{T}(Q) \).

The crucial observation is now that any two Slater determinants belonging to the same symmetry sector \( Q \) differ in at least two entries \( q_n \). As a consequence the 1RDM \( \langle q|\gamma|q' \rangle = \text{Tr}[c_q^\dagger c_q \hat{\Gamma}] \) for an \( N \)-fermion density operator \( \hat{\Gamma} = \sum_{q,q' \in \mathcal{T}(Q)} \Gamma_{qq'} |q\rangle\langle q'| \) (including pure states \( \hat{\Gamma} \equiv |\Psi\rangle\langle \Psi| \), \( |\Psi\rangle = \sum_{q \in \mathcal{T}(Q)} \alpha_q |q\rangle \)) is diagonal. Its diagonal elements, the NONs \( n = (n_q) \), are given by

\[
 n = \sum_{q \in \mathcal{T}(Q)} \Gamma_{qq} v_q f(q) = \sum_{q \in \mathcal{T}(Q)} |\alpha_q|^2 v_q, \tag{1}
\]

where \( v_q \equiv \langle (q|c_q^\dagger c_q|q) \rangle \) is the vector of spin-momentum occupation numbers of the Slater determinant state \( |q\rangle \). Its entries are one whenever \( q \) is contained in \( q \) and zero otherwise. Since any \( n \) is given as the convex combination of the vectors \( \{v_q\}_{q \in \mathcal{T}(Q)} \), the respective sets \( \mathcal{E}_N^1(Q) \) and \( \mathcal{P}_N^1(Q) \) of ensemble and pure \( N \)-representable 1RDMs are given as the polytope with vertices \( \{v_q\}_{q \in \mathcal{T}(Q)} \) and in particular they do coincide (cf. Eq. (1)),

\[
 \mathcal{P}_N^1(Q) = \mathcal{E}_N^1(Q). \tag{2}
\]

Since not all vertices of the hypercube \([0,1]^d \) with particle number \( N \) contribute to those sets, the \( N \)-representability constraints for each sector \( Q \equiv (\vec{K}, M_z) \) are more restrictive than Pauli’s exclusion principle \( 0 \leq n_q \leq 1 \). Yet, it is important to notice that the calculation of those symmetry-adapted generalized Pauli constraints is considerably simpler than the calculation of the one-body pure \( N \)-representability constraints for systems without symmetries.

As an illustration, we consider three fully polarized electrons on a ring of six lattice sites with \( K = 0 \) (for details, see supporting information [41]). It is an elementary exercise to determine all \( (n_1, n_2, n_3) \) with \( \sum_{n=1}^{3} n_\nu = 0 \). One gets \((0,1,5), (0,2,4), (1,2,3), (3,4,5) \) and the respective polytope (2) is then given by the convex hull of the four vertices \((1,1,0,0,0,1), (0,1,1,1,0,0), (0,0,0,1,1,1) \). By solving linear equations this vertex representation of \( \mathcal{P}_N^1 = \mathcal{E}_N^1 \) can be transformed into a half space representation, \( \{D^{(j)}(n) \geq 0 \} \), with the following four \( N \)-representability constraints:

\[
\begin{align*}
 D^{(1)}(n) & = n_0 + n_1 - n_2 \geq 0 \\
 D^{(2)}(n) & = n_0 - n_1 + n_2 \geq 0 \\
 D^{(3)}(n) & = 2 - n_0 - n_1 - n_2 \geq 0 \\
 D^{(4)}(n) & = -n_0 + n_1 + n_2 \geq 0, \tag{3}
\end{align*}
\]

with the linearly dependent variables \( n_3 = 1 - n_0, n_4 = 1 - n_1 \) and \( n_5 = 1 - n_2 \). For larger settings, the easy-to-determine vertex representation of (2) can be transformed into a half space representation by resorting to standard softwares.

**Interaction functional \( F \) and exchange force.**— To elaborate on the structure of the exact interaction functional \( F \), we resort to Levy’s construction [42] (see also Ref. [43]). For general systems (and by ignoring possible symmetries), the exact \( F[\gamma] \) follows as the minimization of the interaction energy over the set of all \( N \)-fermion pure states \( |\Psi\rangle \) with 1RDM \( \gamma \in \mathcal{P}_N^1 \), i.e.

\[ F_p[\gamma] = \min_{|\Psi\rangle} \langle \Psi|H|\Psi\rangle. \]

Yet, it is unclear why those based on pure state ansatzes with \( N \) fixed can be transformed into a half space representation by resorting to Levy’s construction [42] (see also Refs. [81, 82]) and introduce the respective set of configurations \( \{v_q\}_{q \in \mathcal{T}(Q)} \), the respective facet does not contain the vertex \( |\Psi\rangle \). This leads to a “pure RDMFT” on \( \mathcal{P}_N^1 \). In practice, one tries, however, to avoid the highly intricate generalized Pauli constraints [44–46] by relaxing the minimization to \( N \)-fermion ensemble states \( \hat{\Gamma} \). This then leads to an “ensemble RDMFT” with an interaction functional \( \mathcal{F}_\rho \) on the set \( \mathcal{E}_N^1 \) which is described by the simple Pauli exclusion principle constraints only [48].

In the following, we simplify the notation by enumerating all configurations \( q \in \mathcal{T}(Q) \), denote the respective Slater determinants by \( |r\rangle \), \( r = 1, \ldots, R \equiv \dim (\mathcal{H}(Q)) \) and introduce \( V_{r,r'} \equiv \langle r|V|r'\rangle \). Moreover, we will focus on \( \mathcal{F}_p \). As it is proven in the supporting information [41], the equivalence \( \mathcal{F}_p = \mathcal{F}_\rho \) holds, at least whenever there exists phase factors \( \eta_r \) such that \( V_{r,r'} = -\eta_r \eta_{r'} V_{r,r'} \). It is instructive to derive in a first step our main results for systems in which \( \mathcal{P}_N^1 \) takes the form of a simplex, i.e., each of its facets contains all vertices except one. Equivalently, it means that the number of independent coefficients, \( \{\alpha_q\} \), equals the number of independent NONs, \( n \). This condition is valid for several smaller systems, but also for systems of arbitrary size in case their underlying Hilbert space is restricted within \( \mathcal{H}(Q) \) to a subspace involving only \( O(d) \) CI coefficients (yielding an approximate functional). A prime example is the one of three fully polarized electrons on six sites as already discussed above (for details see [41]). We thus label the one-body \( N \)-representability constraints \( D^{(r)}(n) \geq 0 \) such that the respective facet does not contain the vertex \( v_r \), i.e. we have \( D^{(r)}(v_r) = 0 \) whenever \( r \neq r' \). Moreover, we “normalize” each \( D^{(r)} \geq 0 \) such that \( D^{(r)}(v_r) = 1 \). Using
Eq. (1) and the linearity of $D^{(r)}$, we find
\[ D^{(r)}(n) = |\alpha_r|^2. \tag{5} \]

It is exactly the simplicial structure of $\mathcal{P}_N^{1}$ which implies this crucial one-to-one relation between $\{D^{(r)}(n)\}$ and $\{|\alpha_r|^2\}$. Consequently, Levy’s construction (4) with the ansatz $|\Psi\rangle = \sum_r \eta_r |\alpha_r\rangle |r\rangle$ is trivial to carry out up to the phase factors $\eta_r$ of $\alpha_r$. Their minimization leads to some $\tilde{\eta}_r \equiv \eta_r (n, \hat{V})$ and eventually we obtain
\[ \mathcal{F}_p[n] = \sum_{r,r'} V_{rr'} \tilde{\eta}_r \tilde{\eta}_{r'} \sqrt{D^{(r)}(n) D^{(r')}(n)}. \tag{6} \]

The result (6) for the exact interaction functional valid for any symmetry-respecting interaction $\hat{V}$ could hardly be more striking: $\mathcal{F}_p$ is fully determined (up to phase factors $\eta_r (n, \hat{V})$) by the geometry of the simplex $\mathcal{P}_N^{1}$. Moreover, the presence of an exchange force, as we shall call it, follows immediately which diverges on the boundary of $\mathcal{P}_N^{1}$,
\[ \frac{\partial \mathcal{F}_p[n]}{\partial n} \sim \mathcal{G}^{(r)} \frac{1}{\sqrt{D^{(r)}(n)}}, \quad \text{as } D^{(r)}(n) \to 0. \tag{7} \]

Remarkably, the exchange force is always repulsive in the sense that it is repelling $n$ from the polytope boundary (see supporting information [41]).

Generalizing the results (6) and (7) to systems with arbitrary underlying polytope $\mathcal{P}_N^{1} = \mathcal{E}_N^{1}$ is quite intricate: Relation (5) takes the form (see supporting information [41])
\[ D^{(j)}(n) = \sum_{r=1}^{R} D^{(j)}(v_r) |\alpha_r|^2, \tag{8} \]

for all $j = 1, \ldots, J$, where typically $D^{(j)}(v_r) > 0$ for more than one $r$. We also introduced $J$, the number of N-representability constraints. As a consequence, $n$ does not uniquely determine $\{|\alpha_r|\}$ anymore and instead a set of $d$ linear equations with $R > d$ variables has to be solved. The constrained search in (4) then amounts to a non-trivial minimization over the $R - d$ remaining variables. This purely technical and less informative derivation (see supporting information [41]) leads to the general final form
\[ \mathcal{F}_p[n] = \sum_{r,r'=1}^{R} V_{rr'} \tilde{\eta}_r \tilde{\eta}_{r'} \sqrt{\tilde{D}_r(n, \hat{V}) \sqrt{\tilde{D}_{r'}(n, \hat{V})}}, \tag{9} \]
\[ \tilde{D}_r(n, \hat{V}) = \sum_{j=1}^{J} b^{(j)} \tilde{D}^{(j)}(n) + \pi_r (\{D^{(j)}(n)\}, \hat{V}). \]

The coefficients $b^{(j)}$ are solely determined by the geometry of the polytope $\mathcal{P}_N^{1}$ and $\pi_r (\{D^{(j)}(n)\}, \hat{V})$ follow from the minimization of the degrees of freedom not fixed by $n$. This highly involved minimization, as discussed in the supporting information [41], leads to an implicit additional dependence of $\mathcal{F}_p$ on $n$ and the interaction $\hat{V}$.

At the same time, the general form (9) offers excellent prospects for a perturbation theoretical approach by expanding $\pi_r (\{D^{(j)}(n)\}, \hat{V})$ (see Hubbard square below). Whenever $n$ approaches the facet described by $D^{(j)} = 0$, it follows from Eq. (8) that $|\alpha_r| \to 0$ for all $r$ whose vertices $v_r$ do not belong to that facet. This fact must reflect itself in the $n$-dependence of $\mathcal{F}_p$. Indeed, one obtains for each $j$ the singular $n$-dependence [41]
\[ \mathcal{F}_p[n] = \mathcal{F}_p^{(j)} + \mathcal{G}_p^{(j)} \sqrt{\tilde{D}^{(j)}(n) + \mathcal{O}(D^{(j)}(n))}. \tag{10} \]

This result presents in a particularly striking form the crucial role of the $N$-representability constraints $D^{(j)}(n) \geq 0$. In particular, as an extension of (7), it confirms that the fermionic exchange symmetry manifests itself within RDMFT in the form of an exchange force diverging on the boundary of the polytope $\mathcal{P}_N^{1} = \mathcal{E}_N^{1}$.

**Hubbard square.**— Now, as an illustration, we apply the general framework from above to the one-dimensional one-band Hubbard model with $N = 4$ electrons, $L = 4$ sites (half filling) and nearest neighbor hopping with hopping rate $t > 0$. This will emphasize from a different perspective the drastic simplification of RDMFT in case all symmetries are fully exploited: The boundaries of exact functional calculation are extended from the commonly studied Hubbard dimer [21, 23, 24, 32–34, 36] with an underlying six-dimensional Hilbert space to the Hubbard square with a Hilbert space of dimension $70 = (6)^2$.

The kinetic energy functional for the Hubbard square reads $\mathcal{T}[n] = -4t \sum_{\nu=0}^{3} \cos (2\pi \nu / 4) (n_{\nu\uparrow} + n_{\nu\downarrow})$ and the Hubbard on-site interaction has strength $U \geq 0$ (Coulombic repulsion). We will present only the essential steps and refer to the supporting information [41], where all details of the following discussion are presented.

![FIG. 1. Weak and strong coupling asymptotes (11) (dashed lines) and exact functional $\mathcal{F}$ (solid line).](image)
Hence there is only one independent variable \( (n_2) \) (which is identified with \( n \)) constrained by Pauli’s exclusion principle \( 0 \leq n_2 \leq 1 \), only. This is a particular incidence and in larger systems in a singlet state, the translational symmetry implies constraints which are more restrictive than Pauli’s exclusion principle.

For given \( n \), Levy’s construction (4) cannot be fully carried out by analytical means since it involves the root of a polynomial of degree six. The exact functional \( F \equiv F_p = F \) as function of \( n_2 \) is determined numerically instead and we depict it in Figure 1. Its graph demonstrates the divergence of the slope on the “facets” \( n_2 = 0, 1 \), as predicted by (10). Also the particle-hole duality \( F[n_2] = F[1 - n_2] \) is obvious and the convexity of \( F \) is consistent with the fact that “ensemble functionals” \( F_e \) are always convex [43, 83].

Using a perturbative approach for (9), the functional \( F \) simplifies in the asymptotic regimes of weak \((0 \leq U \ll t)\) and strong \((U \gg t)\) coupling [41],

\[
F[n] = U\left[\frac{3}{4} - \frac{\sqrt{13}}{2}\sqrt{n_2} + O(n_2)\right], \quad 0 \leq U \ll t \quad (11)
\]

\[
F[n] = U\left[\frac{4}{3}(1 - n_2)^2 + 40\frac{1}{27}(1 - n_2)^4 + \ldots\right], \quad U \gg t.
\]

FIG. 2. Left: Exact result for the ground state energy \( E_0(u) \) (blue solid line) from the exact functional. The weak and strong coupling result from the functionals (11) is shown by the blue dashed lines. The result from PNOF5 and PNOF7(-) is presented by orange and red dots, respectively. Right: Relative error \( \Delta E/E_0 \) as a function of \( u \).

Using \( T[n] = -8t(\frac{1}{2} - n_2) \) and the results from Eq. (11), one obtains from the minimization of \( E'[n] \) the ground state energy \( E_0 \) and the corresponding NON \( n_2 \) in the weak coupling regime as a function of \( u = U/t \)

\[
E_0(u)/t = -4 + \frac{3}{4}u - \frac{13}{128}u^2 + O(u^3)
\]

\[
n_2(u) = \frac{13}{1024}u^2 + O(u^3)
\]

and for strong coupling

\[
E_0(u)/t = -12u^{-1} + 120u^{-3} + O(u^{-5})
\]

\[
n_2(u) = 1 - 3u^{-1} - 60u^{-3} + O(u^{-5}).
\]

The asymptotically exact results (12),(13) are shown in Figure 2 (left). This figure also contains the exact result and those of PNOF5 [72, 76] and PNOF7(-) [40], the best approximate functionals among all used in Ref. [37, 38]. Result (13) fits perfectly the exact result for all \( u > 10 \). The convergence to zero for \( u \to \infty \) (a general property of the Hubbard model at half filling in any dimension [84]) is reproduced also by PNOF5 and PNOF7(-). In order to check the quality of the approximate functionals more, we have also plotted the relative error \( \Delta E/E_0 \) in Figure 2 (right). We observe that this error is about 60% and 10% for PNOF5 and PNOF7(-), respectively, and practically zero for our approximate result (13) for all \( u > 10 \).

Summary and conclusions.— We have demonstrated how the ab initio knowledge of the natural orbitals for translationally invariant one-band lattice models significantly simplifies reduced density matrix functional theory (RDMFT). For each symmetry sector, the sets \( P \) and \( E_\alpha \) of pure and ensemble \( N \)-representable one-matrices coincide, the interaction functionals \( F_{p,e} \) depend only on the natural occupation numbers \( n \) and RDMFT therefore reduces de facto to a natural occupation number “functional” theory.

Those insights have tremendous consequences. Based on Levy’s construction [42] they allowed us, to discover the form of the exact functional \( F_p[n] \) (cf. (9)) which differs considerably from the approximate functionals proposed so far [4, 5]. Intriguingly, \( F_p[n] \) is given by a bilinear form of square roots (generalizing the two-electron result [85]), whose radicants contain two terms. The first one is linear in the one-body \( N \)-representability constraints \( \{D^{(j)}(n)\} \), while the second summand depends nonlinearly on \( \{D^{(j)}(n)\} \) and on the interaction \( V \) (cf. Eq. (9)). This summand deserves particular attention: First, it arises in the constrained-search (4) from those degrees of freedom of \( \Psi \) which are not determined by the one-matrix. Therefore, it represents within RDMFT irreducible correlations, a crucial concept recently established in quantum information theory [86, 87]. Second, its dependence on \( V \) emphasizes that the construction of highly accurate functionals based, e.g., on tensor properties [54, 59] or \( N \)-representability conditions for the 2RDM [67, 78] would necessitate information on the interaction \( V \), as well. Third a finite series expansion of that term, \( \sum_{\pi} \{D^{(j)}(n)\}, \hat{V} \), with respect to \( \{D^{(j)}(n)\} \) in conjunction with a fitting scheme would allow one to establish a hierarchy of approximate functionals similar to Jacob’s ladder in DFT [88].

Another potentially transformative key result of our work is the discovery of an “exchange force” emerging from the fermionic exchange symmetry: The gradient of the exact functional diverves, \( |dF_p/dn| \sim c_1/\sqrt{D^{(i)}(n)} \), as \( n \) approaches a facet of the polytope \( P_1^N = E_\alpha \), defined by \( D^{(i)} \equiv 0 \). This repulsive divergence on the boundary of \( E_\alpha \) also explains why fermionic occupation numbers \( n_k \) typically cannot take the extremal values 0 or 1.
turn, studying the equation $c_i \hat{V} = 0$ would allow one to systematically identify all (highly non-generic) systems (such as [89]) for which occupation numbers can be pinned to 0 or 1. It will be one of the crucial future challenges to generalize those new concepts to systems without translational symmetry, with particular focus on ensemble RDMFT (i.e., $\mathcal{F}_e$ on $E_{\mathcal{F}}^L$).

Finally, we would like to stress that all our findings hold for systems with non-fixed particle number, as well and $\hat{V}$ can be any (spin-dependent) $p$-particle interaction obeying translational symmetry.

We are grateful to M. Piris and coworkers for sharing their data concerning the Hubbard square. We also thank P.G.J. van Dongen, K.J.H. Giesbertz, I. Mitxelena, T.S. Müller, M. Piris and R. Schade for helpful comments on the manuscript. C.S. acknowledges financial support from the UK Engineering and Physical Sciences Research Council (Grant EP/P007155/1).

[1] T. L. Gilbert, “Hohenberg-Kohn theorem for nonlocal external potentials,” Phys. Rev. B 12, 2111 (1975).
[2] J. Cioslowski, Many-electron densities and reduced density matrices (Springer Science & Business Media, 2000).
[3] M. Piris, “Natural orbital functional theory,” in Reduced-Density-Matrix Mechanics: With Application to Many-Atom Systems and Molecules, edited by D. A. Mazziotto (Wiley-Blackwell, 2007) Chap. 14, p. 387.
[4] 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, M. Filatov, and M. Huix-Rotllant (Springer International Publishing, Cham, 2016) p. 125.
[5] R. Schade, E. Kamil, and P.E. Blöchl, “Reduced density-matrix functionals from many-particle theory,” Eur. Phys. J. Special Topics 226, 2677 (2017).
[6] P. Hohenberg and W. Kohn, “Inhomogeneous electron gas,” Phys. Rev. 136, B864 (1964).
[7] R. G. Parr and W. Yang, “Density-functional theory of the electronic structure of molecules,” Annu. Rev. Phys. Chem. 46, 701 (1995).
[8] E.K.U. Gross and R.M. Dreizler, Density functional theory, Vol. 337 (Springer Science & Business Media, 2013).
[9] R. O. Jones, “Density functional theory: Its origins, rise to prominence, and future,” Rev. Mod. Phys. 87, 897 (2015).
[10] K. Pernal, “Effective potential for natural spin orbitals,” Phys. Rev. Lett. 94, 233002 (2005).
[11] E. Davidson, Reduced density matrices in quantum chemistry, Vol. 6 (Elsevier, 2012).
[12] J. Cioslowski and K. Pernal, “Constraints upon natural spin orbital functionals imposed by properties of a homogeneous electron gas,” J. Chem. Phys. 111, 3396 (1999).
[13] N. N. Lathiotakis, N. Helbig, and E. K. U. Gross, “Performance of one-body reduced density-matrix functionals for the homogeneous electron gas,” Phys. Rev. B 75, 195120 (2007).
[14] S. Sharma, J.K. Dewhurst, N.N. Lathiotakis, and E.K.U. Gross, “Reduced density matrix functional for many-electron systems,” Phys. Rev. B 78, 201103 (2008).
[15] N. N. Lathiotakis, N. Helbig, A. Zacarias, and E. K. U. Gross, “A functional of the one-body-reduced density matrix derived from the homogeneous electron gas: Performance for finite systems,” J. Chem. Phys. 130, 064109 (2009).
[16] N. N. Lathiotakis, S. Sharma, J. K. Dewhurst, F. G. Eich, M. A. L. Marques, and E. K. U. Gross, “Density-matrix-power functional: Performance for finite systems and the homogeneous electron gas,” Phys. Rev. A 79, 040501 (2009).
[17] M. Piris and P. Otto, “The improved Bardeen-Cooper-Schrieffer method in polymers,” J. Chem. Phys. 112, 8187 (2000).
[18] M. Piris and P. Otto, “Natural orbital functional for correlation in polymers,” Int. J. Quant. Chem. 102, 90 (2005).
[19] A. Schindlmayr and R. W. Godby, “Density-functional theory and the $\gamma$-representability problem for model strongly correlated electron systems,” Phys. Rev. B 51, 10427 (1995).
[20] A. E. Carlsson, “Exchange-correlation functional based on the density matrix,” Phys. Rev. B 56, 12058 (1997).
[21] R. López-Sandoval and G. M. Pastor, “Density-matrix functional theory of the Hubbard model: An exact numerical study,” Phys. Rev. B 61, 1764 (2000).
[22] R. G. Hennig and A. E. Carlsson, “Density-matrix functional method for electronic properties of impurities,” Phys. Rev. B 63, 115116 (2001).
[23] R. López-Sandoval and G. M. Pastor, “Density-matrix functional theory of strongly correlated lattice fermions,” Phys. Rev. B 66, 155118 (2002).
[24] R. López-Sandoval and G. M. Pastor, “Interaction-energy functional for lattice density functional theory: Applications to one-, two-, and three-dimensional Hubbard models,” Phys. Rev. B 69, 085101 (2004).
[25] M.F. Silva, N.A. Lima, A.L. Malvezzi, and K. Capelle, “Effects of nanoscale spatial inhomogeneity in strongly correlated systems,” Phys. Rev. B 71, 125130 (2005).
[26] W. Töws and G. M. Pastor, “Lattice density functional theory of the single-impurity Anderson model: Development and applications,” Phys. Rev. B 83, 235101 (2011).
[27] M. Saubanère and G. M. Pastor, “Density-matrix functional study of the Hubbard model on one- and two-dimensional bipartite lattices,” Phys. Rev. B 84, 035111 (2011).
[28] W. Töws and G. M. Pastor, “Spin-polarized density-matrix functional theory of the single-impurity Anderson model,” Phys. Rev. B 86, 245123 (2012).
[29] K. Capelle and V. L. Campo Jr, “Density functionals and model Hamiltonians: Pillars of many-particle physics,” Phys. Rep. 528, 91 (2013).
[30] W. Töws, M. Saubanère, and G. M. Pastor, “Density-matrix functional theory of strongly correlated fermions on lattice models and minimal-basis Hamiltonians,” Theor. Chem. Acc. 133, 1422 (2013).
[31] M. Saubanère and G. M. Pastor, “Lattice density-functional theory of the attractive Hubbard model,” Phys. Rev. B 90, 125128 (2014).
[32] D.J. Carrascal, J. Ferrer, J.C. Smith, and K. Burke, “The Hubbard dimer: A density functional case study
of a many-body problem,” J. Phys. Condens. Matter 27, 393001 (2015).
[33] S. Di Sabatino, J. A. Berger, L. Reining, and P. Romaniello, “Reduced density-matrix functional theory: Correlation and spectroscopy,” J. Chem. Phys. 143, 024108 (2015).
[34] E. Kamil, R. Schade, T. Pruschke, and P. E. Blöchl, “Reduced density-matrix functionals applied to the Hubbard dimer,” Phys. Rev. B 93, 085141 (2016).
[35] M. Saubanère, M. B. Lepetit, and G. M. Pastor, “Interaction-energy functional of the Hubbard model: Local formulation and application to low-dimensional lattices,” Phys. Rev. B 94, 045102 (2016).
[36] A. J. Cohen and P. Mori-Sánchez, “Landscape of an exact energy functional,” Phys. Rev. A 93, 042511 (2016).
[37] I. Mitxelena, M. Piris, and M. Rodríguez-Mayorga, “On the performance of natural orbital functional approximations in the Hubbard model,” J. Phys. Condens. Matter 29, 425602 (2017).
[38] I. Mitxelena, M. Piris, and M. Rodríguez-Mayorga, “Corrigendum: On the performance of natural orbital functional approximations in the Hubbard model,” J. Phys. Condens. Matter 30, 089501 (2018).
[39] T. S. Müller, W. Töws, and G. M. Pastor, “Exploiting the links between ground-state correlations and independent-fermion entropy in the Hubbard model,” Phys. Rev. B 98, 045135 (2018).
[40] I. Mitxelena, M. Rodríguez-Mayorga, and M. Piris, “Phase dilemma in natural orbital functional theory from the N-representability perspective,” Eur. Phys. J. B 91, 109 (2018).
[41] See the Supplemental Material at url for technical details on the derivation of the exact functional, the diverging exchange force and the solution of lattice cluster systems, which includes Refs. [44, 84, 90–92].
[42] M. Levy, “Universal variational functionals of electron densities, first-order density matrices, and natural spin-orbitals and solution of the v-representability problem,” Proc. Natl. Acad. Sci. U.S.A 76, 6062 (1979).
[43] E. H. Lieb, “Density functionals for coulomb systems,” Int. J. Quantum Chem. 24, 243 (1983).
[44] R. E. Borland and K. Dennis, “The conditions on the one-matrix for three-body fermion wavefunctions with one-rank equal to six,” J. Phys. B 5, 7 (1972).
[45] A. Klyachko, “Quantum marginal problem and N-representability,” J. Phys. Conf. Ser. 36, 72 (2006).
[46] M. Altunbulak and A. Klyachko, “The Pauli principle revisited,” Commun. Math. Phys. 282, 287 (2008).
[47] S. M. Valone, “Consequences of extending 1-matrix energy functionals from pure–state representable to all ensemble representable 1-matrices,” J. Chem. Phys. 73, 1344 (1980).
[48] A. J. Coleman, “Structure of fermion density matrices,” Rev. Mod. Phys. 35, 668 (1963).
[49] Y. K. Liu, M. Christian, and F. Verstraete, “Quantum computational complexity of the n-representability problem: QMA complete,” Phys. Rev. Lett. 98, 110503 (2007).
[50] N. Schuch and F. Verstraete, “Computational complexity of interacting electrons and fundamental limitations of density functional theory,” Nat. Phys. 5, 732 (2009).
[51] Christian Schilling, “Communication: Relating the pure and ensemble density matrix functional,” J. Chem. Phys. 149, 231102 (2018).
[52] A. M. K. Müller, “Explicit approximate relation between reduced two- and one-particle density matrices,” Phys. Lett. A 105, 446 (1984).
[53] S. Goedecker and C. J. Umrigar, “Natural orbital functional for the many-electron problem,” Phys. Rev. Lett. 81, 866 (1998).
[54] G. Csányi and T. A. Arias, “Tensor product expansions for correlation in quantum many-body systems,” Phys. Rev. B 61, 7348 (2000).
[55] E. J. Baerends, “Exact exchange-correlation treatment of dissociated H₂ in density functional theory,” Phys. Rev. Lett. 87, 133004 (2001).
[56] K. Yasuda, “Correlation energy functional in the density-matrix functional theory,” Phys. Rev. A 63, 032517 (2001).
[57] K. Yasuda, “Local approximation of the correlation energy functional in the density matrix functional theory,” Phys. Rev. Lett. 88, 053001 (2002).
[58] M. A. Buijse and E. J. Baerends, “An approximate exchange-correlation hole density as a functional of the natural orbitals,” Mol. Phys. 100, 401 (2002).
[59] G. Csányi, S. Goedecker, and T. A. Arias, “Improved tensor-product expansions for the two-particle density matrix,” Phys. Rev. A 65, 032510 (2002).
[60] J. M. Herbert and J. E. Harriman, “N-representability and variational stability in natural orbital functional theory,” J. Chem. Phys. 118, 10835 (2003).
[61] J. Cioslowski, M. Buchowiecki, and P. Ziesche, “Density matrix functional theory of four-electron systems,” J. Chem. Phys. 119, 11570 (2003).
[62] C. Kollmar and B. A. Heß, “A new approach to density matrix functional theory,” J. Chem. Phys. 119, 4655 (2003).
[63] C. Kollmar and B. A. Heß, “The structure of the second-order reduced density matrix in density matrix functional theory and its construction from formal criteria,” J. Chem. Phys. 120, 3158 (2004).
[64] K. Pernal and J. Cioslowski, “Phase dilemma in density matrix functional theory,” J. Chem. Phys. 120, 5987 (2004).
[65] C. Kollmar, “The “JK-only” approximation in density matrix functional and wave function theory,” J. Chem. Phys. 121, 11581 (2004).
[66] O. Gritsenko, K. Pernal, and E. J. Baerends, “An improved density matrix functional by physically motivated repulsive corrections,” J. Chem. Phys. 122, 204102 (2005).
[67] M. Piris, “A new approach for the two-electron cumulant in natural orbital functional theory,” Int. J. Quantum Chem. 106, 1093 (2005).
[68] C. Kollmar, “A size extensive energy functional derived from a double configuration interaction approach: The role of N representability conditions,” J. Chem. Phys. 125, 084108 (2006).
[69] D. R. Rohr, K. Pernal, O. V. Gritsenko, and E. J. Baerends, “A density matrix functional with occupation number driven treatment of dynamical and nondynamical correlation,” J. Chem. Phys. 129, 164105 (2008).
[70] N. N. Lathiotakis and M. A. L. Marques, “Benchmark calculations for reduced-density-matrix functional theory,” J. Chem. Phys. 128, 184103 (2008).
[71] M. A. L. Marques and N. N. Lathiotakis, “Empirical functionals for reduced-density-matrix-functional theory,” Phys. Rev. A 77, 032509 (2008).
[72] M. Piris, X. Lopez, F. Ruipérez, J. M. Matxain, and J. M. Ugalde, “A natural orbital functional for multiconfigurational states,” J. Chem. Phys. 134, 164102 (2011).
[73] C. L. Benavides-Riveros and J. C. Várilly, “Testing one-body density functionals on a solvable model,” Eur. Phys. J. D 66, 274 (2012).
[74] M. Piris, “A natural orbital functional based on an explicit approach of the two-electron cumulant,” Int. J. Quantum Chem. 113, 620 (2012).
[75] K. Pernal, “The equivalence of the Piris natural orbital functional 5 (PNOF5) and the antisymmetrized product of strongly orthogonal geminal theory,” Comput. Theor. Chem. 1003, 127 (2013).
[76] M. Piris, J.M. Matxain, and X. Lopez, “The intrapair electron correlation in natural orbital functional theory,” J. Chem. Phys. 139, 234109 (2013).
[77] M. Piris and J. M. Ugalde, “Perspective on natural orbital functional theory,” Int. J. Quantum Chem. 114, 1169 (2014).
[78] M. Piris, “Global method for electron correlation,” Phys. Rev. Lett. 119, 063002 (2017).
[79] M. Piris, “Dynamic electron-correlation energy in the natural-orbital-functional second-order-Møller-Plesset perturbation theory,” Phys. Rev. A 98, 022504 (2018).
[80] C. L. Benavides-Riveros and M. A. L. Marques, “Static correlated functionals for reduced density matrix functional theory,” Eur. Phys. J. B 91, 133 (2018).
[81] O. V. Gritsenko, “Comment on “Nonuniqueness of algebraic first-order density-matrix functionals” ,” Phys. Rev. A 97, 026501 (2018).
[82] J. Wang and P. J. Knowles, “Reply to “Comment on “Nonuniqueness of algebraic first-order density-matrix functionals” ,” Phys. Rev. A 97, 026502 (2018).
[83] G. Zundel and K. Maschke, “Density matrix functional theory for the n-particle ground state,” J. Chem. Phys. 82, 5604 (1985).
[84] E. Fradkin, Field theories of condensed matter physics (Cambridge University Press, 2013).
[85] P.-O. Löwdin and H. Shull, “Natural orbitals in the quantum theory of two-electron systems,” Phys. Rev. 101, 1730 (1956).
[86] N. Linden, S. Popescu, and W. K. Wootters, “Almost every pure state of three qubits is completely determined by its two-particle reduced density matrices,” Phys. Rev. Lett. 89, 207901 (2002).
[87] N. Linden and W. K. Wootters, “The parts determine the whole in a generic pure quantum state,” Phys. Rev. Lett. 89, 277906 (2002).
[88] J. P. Perdew and K. Schmidt, “Jacob’s ladder of density functional approximations for the exchange-correlation energy,” in AIP Conference Proceedings, Vol. 577 (2001) p. 1.
[89] J. Cioslowski, “Solitonic natural orbitals,” J. Chem. Phys. 148, 134120 (2018).
[90] A. Klyachko, “The Pauli exclusion principle and beyond,” arXiv:0904.2009 (2009).
[91] C. Schilling, C. L. Benavides-Riveros, and P. Vrana, “Reconstructing quantum states from single-party information,” Phys. Rev. A 96, 052312 (2017).
[92] M. B. Ruskai, “Connecting N-representability to Weyl’s problem: the one-particle density matrix for n = 3 and r = 6,” J. Phys. A 40, F961 (2007).