Diverging Exchange Force and Form of the Exact Density Matrix Functional

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For translationally invariant one-band lattice models, we exploit the *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 $\boldsymbol{n}$. 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 $D^{(j)}(\boldsymbol{n}) \geq 0$. For smaller systems, it follows as $\mathcal{F}[\boldsymbol{n}] = \sum_{i,i'} \bar{V}_{i,i'} \sqrt{D^{(i)}(\boldsymbol{n}) D^{(i')}(\boldsymbol{n})}$. This generalizes to systems of arbitrary size by replacing each $D^{(i)}$ by a linear combination of $\{D^{(j)}(\boldsymbol{n})\}$ and adding a nonanalytical term involving the interaction $\hat{V}$. Third, the gradient $d\mathcal{F}/d\boldsymbol{n}$ 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 a nonfixed particle number as well and $\hat{V}$ can be *any* p-particle interaction. As an illustration, we derive the *exact* functional for the Hubbard square.

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Introduction.—Reduced density matrix functional theory (RDMFT) [1–5] has the potential of overcoming the shortcomings and fundamental limitations of the widely used density functional theory (DFT) [6–9]. Involving the full one-particle reduced density matrix (1RDM) γ facilitates not only an exact description of the single particle potential energy, $\mathcal{U}[\gamma] \equiv \mathrm{Tr}[\hat{U}\gamma]$, but also of the kinetic energy, $\mathcal{T}[\gamma] \equiv \mathrm{Tr}[\hat{T}\gamma]$. 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 [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 $\gamma \equiv \sum_j n_j |\varphi_j\rangle\langle\varphi_j|$ and then minimizes the total energy functional $\mathcal{E}[\gamma] = \mathcal{T}[\gamma] + \mathcal{U}[\gamma] + \mathcal{F}[\gamma]$ with respect to the natural occupation numbers (NONs) n_j *and* natural orbitals $|\varphi_j\rangle$, separately. The dependence on the latter makes the minimization of $\mathcal{E}$ particularly difficult and one often encounters slow convergence (see, e.g., Ref. [10]).

The general situation drastically changes in favor 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 [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 *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 [12–16], periodic polymers [17,18], and of lattice systems [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.

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 $\vec{k} = (2\pi/L)(\nu_1, \ldots, \nu_D)^t \equiv (2\pi/L)\vec{\nu}$, where $\nu_i = 0, 1, \ldots, L-1$. The spin-orbitals follow as $|\vec{\nu}m\rangle$ ($m = \pm\frac{1}{2}$) and we introduce for the following the collective quantum number $q \equiv (\vec{\nu}m)$. On the N-fermion level, a symmetry-adapted basis is then given by the Slater determinants $|\boldsymbol{q}\rangle \equiv |q_1, \ldots, q_N\rangle$. 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 $\{|\boldsymbol{q}\rangle\}_{\boldsymbol{q}\in\mathcal{I}^{(Q)}}$ with total momentum $\vec{K} = \sum_{n=1}^{N} \vec{k}_n$ and magnetization $M_z = \sum_{n=1}^{N} m_n$. The respective set of configurations $\boldsymbol{q}$ is denoted by $\mathcal{I}^{(Q)}$.

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 $\langle q|\gamma|q'\rangle = \mathrm{Tr}[c_{q'}^{\dagger} c_q \hat{\Gamma}]$ for an N-fermion density operator $\hat{\Gamma} = \sum_{\boldsymbol{q},\boldsymbol{q}'\in\mathcal{I}^{(Q)}} \Gamma_{\boldsymbol{q}\boldsymbol{q}'} |\boldsymbol{q}\rangle\langle\boldsymbol{q}'|$ (including pure states $\hat{\Gamma} \equiv |\Psi\rangle\langle\Psi|$, $|\Psi\rangle = \sum_{\boldsymbol{q}\in\mathcal{I}^{(Q)}} \alpha_{\boldsymbol{q}} |\boldsymbol{q}\rangle$) is diagonal. Its diagonal elements, the NONs $\boldsymbol{n} = (n_q)$, are given by

$$\boldsymbol{n} = \sum_{\boldsymbol{q}\in\mathcal{I}^{(Q)}} \Gamma_{\boldsymbol{q}\boldsymbol{q}} \boldsymbol{v}_{\boldsymbol{q}} \overset{\hat{\Gamma}\equiv|\Psi\rangle\langle\Psi|}{=} \sum_{\boldsymbol{q}\in\mathcal{I}^{(Q)}} |\alpha_{\boldsymbol{q}}|^2 \boldsymbol{v}_{\boldsymbol{q}}, \tag{1}$$

where $\boldsymbol{v}_{\boldsymbol{q}} \equiv (\langle\boldsymbol{q}|c_q^{\dagger} c_q|\boldsymbol{q}\rangle)$ is the vector of spin-momentum occupation numbers of the Slater determinant state $|\boldsymbol{q}\rangle\langle\boldsymbol{q}|$. Its entries are one whenever q is contained in $\boldsymbol{q}$ and zero otherwise. Since any $\boldsymbol{n}$ is given as the convex combination of the vectors $\{\boldsymbol{v}_{\boldsymbol{q}}\}_{\boldsymbol{q}\in\mathcal{I}^{(Q)}}$, 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 $\{\boldsymbol{v}_{\boldsymbol{q}}\}_{\boldsymbol{q}\in\mathcal{I}^{(Q)}}$ 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 $0 \le n_q \le 1$. 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 the Supplemental Material [41]). It is an elementary exercise to determine all (ν_1, ν_2, ν_3) with $\sum_{n=1}^{3} \nu_n (\mathrm{mod}\ 6) = 0$. 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), (1,0,1,0,1,0), (0,1,1,1,0,0), and (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, $\{D^{(j)}(\boldsymbol{n}) \ge 0\}$, with the following four N-representability constraints:

$$\begin{aligned} D^{(1)}(\boldsymbol{n}) &= n_0 + n_1 - n_2 \ge 0, \\ D^{(2)}(\boldsymbol{n}) &= n_0 - n_1 + n_2 \ge 0, \\ D^{(3)}(\boldsymbol{n}) &= 2 - n_0 - n_1 - n_2 \ge 0, \\ D^{(4)}(\boldsymbol{n}) &= -n_0 + n_1 + n_2 \ge 0, \end{aligned} \tag{3}$$

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 Eq. (2) can be transformed into a half-space representation by resorting to standard softwares.

Interaction functional $\mathcal{F}$ and exchange force.—To elaborate on the structure of the *exact* interaction functional $\mathcal{F}$, we resort to Levy's construction [47] (see also Ref. [48]). For general systems (and by ignoring possible symmetries), the *exact* $\mathcal{F}[\gamma]$ 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., $\mathcal{F}_p[\gamma] = \min_{\Psi\mapsto\gamma} \langle\Psi|\hat{V}|\Psi\rangle$. This leads to a "pure RDMFT" on $\mathcal{P}_N^1$. In practice, one tries, however, to avoid the highly intricate generalized Pauli constraints [44,49,50] by relaxing the minimization to N-fermion *ensemble* states $\hat{\Gamma}$ [51]. This then leads to an "ensemble RDMFT" with an interaction functional $\mathcal{F}_e$ on the set $\mathcal{E}_N^1$, which is described by the simple Pauli exclusion principle constraints only [52]. Yet, this cannot allow one to "circumvent" the mathematically proven complexity of the ground state problem [53,54] and the complexity is just shifted from the set of underlying 1RDMs to the derivation of the functional $\mathcal{F}_e$ and/or its minimization [55]. In that context, with regard to approximated functionals such as those found in Refs. [5,12–16,40,56–84], it is unclear why those based on pure state *Ansätze* with fixed N are treated within "ensemble RDMFT," as well. For more details, the reader is referred to the reviews [4,5] and references therein.

As already stressed above, for periodic one-band lattice systems the interaction functionals simplify drastically to functionals (or more precisely to functions) of the spin-momentum occupation numbers $\boldsymbol{n}$. For each $Q \equiv (\vec{K}, M_z)$, Levy's construction [47] is restricted to $|\Psi\rangle$ in the respective symmetry-sector (see also Refs. [85,86])

$$\mathcal{F}_p[\boldsymbol{n}] = \min_{\mathcal{H}^{(Q)}\ni\Psi\mapsto\boldsymbol{n}} \langle\Psi|\hat{V}|\Psi\rangle. \tag{4}$$

In the following, we simplify the notation by enumerating all configurations $\boldsymbol{q} \in \mathcal{I}^{(Q)}$, denote the respective Slater determinants by $|r\rangle$, $r = 1, \ldots, R \equiv \dim[\mathcal{H}^{(Q)}]$, and introduce $V_{rr'} \equiv \langle r|\hat{V}|r'\rangle$. Moreover, we will focus on $\mathcal{F}_p$. As it is proven in the supporting information [41], the

equivalence $\mathcal{F}_e \equiv \mathcal{F}_p$ holds, at least whenever there exists phase factors η_r such that $V_{rr'} \equiv -\eta_r \eta_{r'} |V_{rr'}|$.

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, $\boldsymbol{n}$. This condition is valid for several smaller systems, but also for systems of arbitrary size in case their underlying Hilbert space is restricted within Eq. (4) to a subspace involving only $\mathcal{O}(d)$ 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 the Supplemental Material [41]). We thus label the one-body N-representability constraints $D^{(r)}(\boldsymbol{n}) \geq 0$ such that the respective facet does not contain the vertex $\boldsymbol{v}_r$; i.e., we have $D^{(r)}(\boldsymbol{v}_{r'}) = 0$ whenever $r \neq r'$. Moreover, we "normalize" each $D^{(r)} \geq 0$ such that $D^{(r)}(\boldsymbol{v}_r) = 1$. Using Eq. (1) and the linearity of $D^{(r)}$, we find

$$D^{(r)}(\boldsymbol{n}) = |\alpha_r|^2. \tag{5}$$

It is exactly the simplicial structure of $\mathcal{P}_N^1$ that implies this crucial one-to-one relation between $\{D^{(r)}(\boldsymbol{n})\}$ and $\{|\alpha_r|^2\}$. Consequently, Levy's construction [Eq. (4)] with the *Ansatz* $|\Psi\rangle = \sum_r \eta_r |\alpha_r| |r\rangle$ is trivial to carry out up to the phase factors η_r of α_r. Their minimization leads to some $\bar{\eta}_r \equiv \bar{\eta}_r(\boldsymbol{n}, \hat{V})$ and eventually we obtain

$$\mathcal{F}_p[\boldsymbol{n}] = \sum_{r,r'} V_{rr'} \bar{\eta}_r^* \bar{\eta}_{r'} \sqrt{D^{(r)}(\boldsymbol{n}) D^{(r')}(\boldsymbol{n})}. \tag{6}$$

The result [Eq. (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 $\bar{\eta}_r(\boldsymbol{n}, \hat{V})$] 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 $\mathcal{P}_N^1$,

$$\left| \frac{\mathrm{d}\mathcal{F}_p}{\mathrm{d}\boldsymbol{n}}[\boldsymbol{n}] \right| \sim \mathcal{G}^{(r)} \frac{1}{\sqrt{D^{(r)}(\boldsymbol{n})}}, \quad \text{as } D^{(r)}(\boldsymbol{n}) \to 0. \tag{7}$$

Remarkably, the exchange force is always repulsive in the sense that it is repelling $\boldsymbol{n}$ from the polytope boundary (see Supplemental Material [41]).

Generalizing the results [Eqs. (6) and (7)] to systems with *arbitrary* underlying polytope $\mathcal{P}_N^1 \equiv \mathcal{E}_N^1$ is quite intricate: relation [Eq. (5)] takes the form (see Supplemental Material [41])

$$D^{(j)}(\boldsymbol{n}) = \sum_{r=1}^{R} D^{(j)}(\boldsymbol{v}_r) |\alpha_r|^2, \tag{8}$$

for all $j = 1, \ldots, J$, where typically $D^{(j)}(\boldsymbol{v}_r) > 0$ for more than one r. We also introduced J, the number of N-representability constraints. As a consequence, $\boldsymbol{n}$ 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 Eq. (4) then amounts to a nontrivial minimization over the $R - d$ remaining variables. This purely technical and less informative derivation (see Supplemental Material [41]) leads to the general final *form*

$$\mathcal{F}_p[\boldsymbol{n}] = \sum_{r,r'=1}^{R} V_{rr'} \bar{\eta}_r^* \bar{\eta}_{r'} \sqrt{\tilde{D}_r(\boldsymbol{n}, \hat{V})} \sqrt{\tilde{D}_{r'}(\boldsymbol{n}, \hat{V})},$$

$$\tilde{D}_r(\boldsymbol{n}, \hat{V}) \equiv \sum_{j=1}^{J} b_r^{(j)} D^{(j)}(\boldsymbol{n}) + \bar{a}_r(\{D^{(i)}(\boldsymbol{n})\}, \hat{V}). \tag{9}$$

The coefficients $b_r^{(j)}$ are solely determined by the geometry of the polytope $\mathcal{P}_1^N$ and $\bar{a}_r(\{D^{(j)}(\boldsymbol{n})\}, \hat{V})$ follow from the minimization of the degrees of freedom not fixed by $\boldsymbol{n}$. This highly involved minimization, as discussed in the Supplemental Material [41], leads to an implicit additional dependence of $\mathcal{F}_p$ on $\boldsymbol{n}$ and the interaction $\hat{V}$.

At the same time, the general form [Eq. (9)] offers excellent prospects for a perturbation theoretical approach by expanding $\bar{a}_r(\{D^{(j)}(\boldsymbol{n})\}, \hat{V})$ (see Hubbard square below).

Whenever $\boldsymbol{n}$ approaches the facet described by $D^{(j)} \equiv 0$, it follows from Eq. (8) that $|\alpha_r| \to 0$ for all r whose vertices $\boldsymbol{v}^{(r)}$ do not belong to that facet. This fact must reflect itself in the $\boldsymbol{n}$ dependence of $\mathcal{F}_p$. Indeed, one obtains for each j the singular $\boldsymbol{n}$ dependence [41]

$$\mathcal{F}_p[\boldsymbol{n}] = \mathcal{F}_p^{(j)} + \mathcal{G}_p^{(j)} \sqrt{D^{(j)}(\boldsymbol{n})} + \mathcal{O}[D^{(j)}(\boldsymbol{n})]. \tag{10}$$

This result presents in a particularly striking form the crucial role of the N-representability constraints $D^{(j)}(\boldsymbol{n}) \geq 0$. In particular, as an extension of Eq. (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 $70 = \binom{8}{4}$.

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

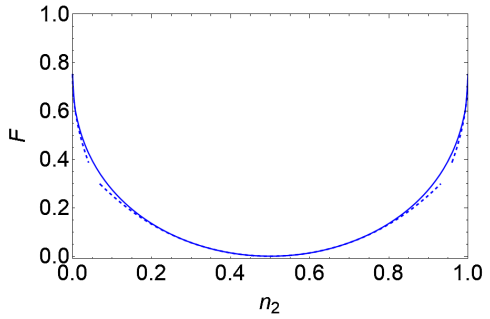

FIG. 1. Weak and strong coupling asymptotes [Eq. (11)] (dashed lines) and exact functional $\mathcal{F}$ (solid line).

The ground state for $U \geq 0$ is a singlet state with total momentum $K = (2\pi/4)2 = \pi$ and parity $p = -1$. Taking all these symmetries into account leads to a rather simple polytope $\mathcal{P}_N^1 = \mathcal{E}_N^1 \cong [0,1]$ of N-representable 1RDMs: it is $n_{\nu\uparrow} = n_{\nu\downarrow} \equiv n_\nu$, $n_1 = n_3 = 1/2$, and $n_0 = 1 - n_2$. Hence there is only one independent variable (n_2) (which is identified with $\boldsymbol{n}$) 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 $\boldsymbol{n}$, Levy's construction [Eq. (4)] cannot be fully carried out by analytical means since it involves the root of a polynomial of degree six. The exact functional $\mathcal{F} \equiv \mathcal{F}_p = \mathcal{F}_e$ [41] as a function of n_2 is determined numerically instead and we depict it in Fig. 1. Its graph demonstrates the divergence of the slope on the "facets" $n_2 = 0$, 1, as predicted by Eq. (10). Also the particle-hole duality $\mathcal{F}[n_2] = \mathcal{F}[1 - n_2]$ [60] is obvious and the convexity of $\mathcal{F}$ is consistent with the fact that "ensemble functionals" $\mathcal{F}_e$ are always convex [48,87].

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

$$\mathcal{F}[\boldsymbol{n}] = U\left(\frac{3}{4} - \frac{\sqrt{13}}{2}\sqrt{n_2} + \mathcal{O}(n_2)\right), \qquad 0 \leq U \ll t,$$
$$\mathcal{F}[\boldsymbol{n}] = U\left[\frac{4}{3}\left(\frac{1}{2} - n_2\right)^2 + \frac{40}{27}\left(\frac{1}{2} - n_2\right)^4 + \ldots\right], \qquad U \gg t. \tag{11}$$

Using $\mathcal{T}[\boldsymbol{n}] = -8t(\frac{1}{2} - n_2)$ and the results from Eq. (11), one obtains from the minimization of $\mathcal{E}[\boldsymbol{n}]$ the ground state energy E_0 and the corresponding NON n_2 in the weak coupling regime as a function of $u = U/t$

$$E_0(u)/t = -4 + \frac{3}{4}u - \frac{13}{128}u^2 + \mathcal{O}(u^3),$$
$$n_2(u) = \frac{13}{1024}u^2 + \mathcal{O}(u^3) \tag{12}$$

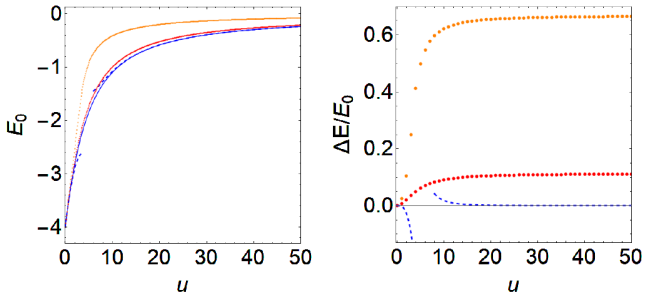

FIG. 2. Left: Exact result for the ground state energy $E_0(u)$ (blue solid line) from the exact functional. The weak and strong coupling result from the functionals [Eq. (11)] is shown by the blue dashed lines. The result from PNOF5 and PNOF7(-) is presented by orange and red dots, respectively. Right: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

and for strong coupling

$$E_0(u)/t = -12u^{-1} + 120u^{-3} + \mathcal{O}(u^{-5}),$$
$$n_2(u) = \frac{1}{2} - 3u^{-1} - 60u^{-3} + \mathcal{O}(u^{-5}). \tag{13}$$

The asymptotically exact results [Eqs. (12) and (13)] are shown in Fig. 2(left). This figure also contains the exact result and those of PNOF5 [76,80] and PNOF7(-) [40], the best approximate functionals among all used in Refs. [37,38]. The result of Eq. (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 [46]) 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 Fig. 2(right). We observe that this error is about 60% and 10% for PNOF5 and PNOF7(-), respectively, and practically zero for our approximate result [Eq. (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 $\mathcal{P}_N^1$ and $\mathcal{E}_N^1$ of pure and ensemble N-representable one-matrices coincide, the interaction functionals $\mathcal{F}_{p/e}$ depend only on the natural occupation numbers $\boldsymbol{n}$ and RDMFT therefore reduces *de facto* to a natural occupation number "functional" theory.

Those insights have tremendous consequences. Based on Levy's construction [47], they allowed us to discover the *form* of the *exact* functional $\mathcal{F}_p[\boldsymbol{n}]$ [cf. Eq. (9)] which differs considerably from the approximate functionals proposed so far [4,5]. Intriguingly, $\mathcal{F}_p[\boldsymbol{n}]$ is given by a bilinear form of square roots (generalizing the two-electron result [88]), whose radicants contain *two* terms. The first one is linear in the one-body N-representability constraints

$\{D^{(j)}(\boldsymbol{n})\}$, while the second summand depends nonlinearly on $\{D^{(j)}(\boldsymbol{n})\}$ and on the interaction $\hat{V}$ [cf. Eq. (9)]. This summand deserves particular attention. First, it arises in the constrained search [Eq. (4)] from those degrees of freedom of Ψ which are *not* determined by the one-matrix. Therefore, it represents within RDMFT *irreducible correlations*, a crucial concept recently established in quantum information theory [89,90]. Second, its dependence on $\hat{V}$ emphasizes that the construction of highly accurate functionals based, e.g., on tensor properties [58,63] or N-representability conditions for the 2RDM [71,82] would necessitate information on the interaction $\hat{V}$, as well. Third, a finite series expansion of that term, $\bar{a}_r(\{D^{(i)}(\boldsymbol{n})\}, \hat{V})$, with respect to $\{D^{(i)}(\boldsymbol{n})\}$ in conjunction with a fitting scheme would allow one to establish a hierarchy of approximate functionals similar to Jacob's ladder in DFT [91].

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 diverges, $|\mathrm{d}\mathcal{F}_p/\mathrm{d}\boldsymbol{n}| \sim c_i/\sqrt{D^{(i)}(\boldsymbol{n})}$, as $\boldsymbol{n}$ approaches a facet of the polytope $\mathcal{P}_N^1 = \mathcal{E}_N^1$, defined by $D^{(i)} \equiv 0$. This repulsive divergence on the boundary of $\mathcal{E}_N^1$ also explains why fermionic occupation numbers n_k *typically* cannot take the extremal values 0 or 1. In turn, studying the equation $c_i(\hat{V}) = 0$ would allow one to systematically identify all (highly nongeneric) systems (such as Ref. [92]) 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 $\mathcal{E}_N^1$).

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

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