Functional Theory for Excitations in Boson Systems

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We propose, work out and initiate a one-particle reduced density matrix functional theory (RDMFT) for targeting excitation energies of bosonic quantum systems. In analogy to its recently established fermionic counterpart, this bosonic $w$-ensemble RDMFT is based on a generalization of the Rayleigh-Ritz variational principle to ensemble states with spectrum $w$ used within the constrained search formalism. Most importantly, an exact convex relaxation is implemented to turn $w$-ensemble RDMFT into a practically feasibly method: The corresponding convex-relaxed 1-body $w$-ensemble $N$-representability problem can be solved, revealing a complete hierarchy of exclusion principle constraints in analogy to Pauli’s famous principle for fermions. To initiate $w$-ensemble RDMFT, we systematically derive universal functionals for the symmetric Bose-Hubbard dimer and for Bose-Einstein condensates in the Bogoliubov regime.

I. INTRODUCTION

A comprehensive understanding of quantum matter requires both the knowledge of the respective ground state properties and the accurate description of excitations. In the case of bosonic quantum systems, particularly prominent examples are collective phenomena in ultracold Bose gases such as excitations in Bose-Einstein condensates (BECs) [1–5], superfluids [6–9] or quantum magnetism [9–12]. Although wave function based methods allow for an exact treatment, at least in principle, they are only practically feasible for relatively small system sizes due to the exponential growth of the underlying Hilbert space. Conversely, Gross-Pitaevskii theory [9] and methods based on mean-field approximations in general fail to describe strong correlation effects. To this end, accurate approaches to strongly correlated many-particle systems with an affordable computational cost are of particular demand.

To motivate and put forth such an approach we recall the Penrose and Onsager criterion [13]: a system exhibits BEC whenever the maximum eigenvalue of the one-particle reduced density matrix (1RDM) is proportional to the total particle number. From a conceptual point of view, this clearly identifies one-particle reduced density matrix functional theory (RDMFT) as a promising novel approach to BEC. Actually, its scope would even extend to arbitrary strongly correlated bosonic quantum systems since the 1RDM provides direct insights into the correlation strength through its degree of mixedness. Despite all these points, the foundation for a bosonic ground state RDMFT was proposed only very recently [15, 16]. This is even more remarkable, since its fermionic counter part is enjoying an intensive ongoing development [17–37].

In order to describe many-body quantum systems comprehensively, however, an extension of ground RDMFT to excited states is imperative. A corresponding extended RDMFT was introduced for fermions in Ref. [38, 39], as a generalization of the well-known DFT for excited states (GOK-DFT) [40–42]. The bosonic counterpart of this so-called $w$-ensemble RDMFT is missing so far. It is this observation and the necessity to accurately describe excited states which urges us to propose in this paper an RDMFT for calculating bosonic excitations energies. In analogy to fermionic $w$-RDMFT and GOK-DFT, it is also based on a generalization of the Rayleigh-Ritz variational principle to $w$-ensembles employed within the constrained search formalism. Intriguingly, the bosonic exchange statistics leads to substantial differences compared to fermionic $w$-ensemble RDMFT: It is one of the key achievements of our work to solve the underlying one-body $w$-ensemble $N$-representability problem. This in turn reveals a hierarchy of bosonic exclusion principles, the bosonic counterpart of Pauli’s famous principle and its generalizations to mixed states [38, 39].

The paper is structured as follows. To keep our work self-contained, we recall in Sec. II foundational aspects of ground state RDMFT. In Sec. III, we introduce a bosonic RDMFT for excited states. A detailed discussion of the resulting exclusion principles for bosons is provided in Sec. IV. Moreover, we derive the respective constraints for several examples in Sec. V. We illustrate the bosonic $w$-ensemble RDMFT and derive the exact universal functionals for the symmetric Bose-Hubbard dimer in Sec. VI and homogeneous BECs in Sec. VII. Finally, we comment on the implications of the bosonic exclusion principles for bosonic lattice DFT in Sec. VIII.

II. RECAP OF GROUND STATE RDMFT

In this section, we recap the foundation of ground state RDMFT. These concepts will reappear in a modified form, adapted to the description of excited states, in Sec. III. Since we are interested in describing $N$-particle quantum systems, we first introduce the $N$-
particle Hilbert space $\mathcal{H}_N$ and denote by $\mathcal{H}_1$ the underlying $d$-dimensional one-particle Hilbert space. For identical fermions, all states in $\mathcal{H}_N$ are antisymmetric under the exchange of two particles, $\mathcal{H}_N \equiv \wedge^N[\mathcal{H}_1]$, whereas for bosons they are symmetric, $\mathcal{H}_N \equiv S^N[\mathcal{H}_1]$. Moreover, we denote by $\mathcal{E}^N$ the set of all ensemble $N$-particle density operators $\hat{\Gamma}$. Then, the extremal elements of $\mathcal{E}^N$ form the set $\mathcal{P}^N$ of pure states $\hat{\Gamma} = |\Psi\rangle\langle\Psi|$. By tracing out $N-1$ particles from a state $\hat{\Gamma} \in \mathcal{E}^N$, we obtain the one-particle reduced density operator $\hat{\gamma}$,

$$\hat{\gamma} \equiv N \text{Tr}_{N-1}[\hat{\Gamma}] = \sum_{i=1}^{d} \lambda_i |i\rangle\langle i| ,$$

where the eigenvalues $\lambda_i$ are the so-called natural occupation numbers and $|i\rangle$ the corresponding natural orbitals.

To motivate RDMFT from a general perspective, we observe that different fields in physics are typically characterized by the one-particle Hamiltonian $\hat{h}$ and an external potential $\hat{v}$. Then for a fixed interaction $\hat{W}$, the main motivation behind RDMFT is to solve the ground state problem for $\hat{H}(\hat{h})$ for the full class $\{\hat{h}\}$ of one-particle Hamiltonians $\hat{h}$. According to Levy [43] and Lieb [44], the ground state energy $E(\hat{h})$ and the ground state 1RDM then follow from the Rayleigh-Ritz variational principle as

$$E(\hat{h}) = \min_{\hat{\Gamma} \in \mathcal{P}^N} \text{Tr}_N \left[ (\hat{h} + \hat{W})\hat{\Gamma} \right]$$

$$\equiv \min_{\hat{\gamma} \in \mathcal{P}_1^N} \left[ \text{Tr}_1 [\hat{h}\hat{\gamma}] + \mathcal{F}(\hat{\gamma}) \right] .$$

The pure ground state universal functional $\mathcal{F}(\hat{\gamma})$ in this so-called constrained search formalism follows from the minimization of the expectation value $\text{Tr}_N[\hat{W}\hat{\Gamma}]$ over all $\hat{\Gamma} \in \mathcal{P}^N$,

$$\mathcal{F}(\hat{\gamma}) \equiv \min_{\hat{\Gamma} \in \mathcal{P}^N} \text{Tr}_N[\hat{W}\hat{\Gamma}] .$$

In its original formulation by Levy [43], the minimization in Eq. (3) is performed over all pure states $\hat{\Gamma} \in \mathcal{P}^N$ and thus the domain of $\mathcal{F}$ consists of all pure state $N$-representable 1RDMs $\hat{\gamma} \in \mathcal{P}_1^N = N \text{Tr}_{N-1}[\mathcal{P}^N]$. However, the non-convex set $\mathcal{P}_1^N$ is in general unknown for fermions, a fact which is due to the too complicated generalized Pauli constraints [45–47]. Justified by the observation that an exact convex relaxation [48] of the non-convex optimization problem [3] does not change the outcome of the minimization, Valone [49] proposed to extend the domain of $\mathcal{F}$ to all ensemble $N$-representable $\hat{\gamma} \in \mathcal{E}_1^N = N \text{Tr}_{N-1}[\mathcal{E}^N]$. Indeed, both sets $\mathcal{E}^N$ and $\mathcal{E}_1^N$ are convex and in particular $\mathcal{E}_1^N = \text{conv}(\mathcal{P}_1^N)$, where $\text{conv}(\cdot)$ denotes the convex hull (see Ref. [48]). Moreover, the corresponding ensemble ground state functional $\mathcal{F}$ is equal to the lower convex envelope of $\mathcal{F}$ [20],

$$\mathcal{F}(\hat{\gamma}) = \text{conv} (\mathcal{F}(\hat{\gamma})) .$$

For fermions, Valone’s [49] formulation of RDMFT is the crucial step which turned RDMFT into a practical method since the set $\mathcal{E}_1^N$ is only restricted through the well-known Pauli exclusion principle $0 \leq \lambda_i \leq 1$. In the case of bosons, the natural occupation numbers are only restricted through $0 \leq \lambda_i \leq N$ and there are no additional constraints as the generalized Pauli constraints for fermions. Therefore, the pure state $N$-representability constraints for bosons are known and, in particular, $\mathcal{E}_1^N = \mathcal{P}_1^N$ [15, 22]. Nevertheless, relaxing the minimization in Eq. (3) to a convex one is also advantageous for bosons since any local minimum of a convex functional is also a global minimum.

### III. RDMFT FOR EXCITED STATES

In this section, we propose an $w$-ensemble RDMFT for targeting excited states of bosonic quantum systems and provide a comprehensive foundation for it. To keep our work self-contained, we recall the required key concepts from its fermionic counterpart [38, 39], while focusing on the emerging challenges and modifications due to the bosonic exchange statistics.

#### A. Foundations of $w$-ensemble RDMFT

Similar to ground state RDMFT discussed in the section above, also the $w$-ensemble RDMFT for excited states is based on a suitable variational principle and a constrained search formalism as in Eq. (3). In 1988, Gross, Oliviera, and Kohn [40–42] proposed the following generalization of the Rayleigh-Ritz variational principle: Let $\hat{H}$ denote a Hamiltonian on a $D$-dimensional Hilbert space $\mathcal{H}$ with increasingly ordered eigenenergies $E_1 \leq E_2 \leq \ldots \leq E_D$ and corresponding eigenstates $|\Psi_j\rangle$. Furthermore, let $w \in \mathbb{R}^D$ be a vector with decreasingly ordered entries $w_1 \geq w_2 \geq \ldots \geq w_D$, and normalization $\sum_{j=1}^{D} w_j = 1$. $\mathcal{E}(w)$ shall denote the set of all density operators with fixed decreasingly ordered spectrum $\text{spec}^\uparrow(\hat{\Gamma}) = w$. Then, the weighted sum of the eigenvalues $E_j$ follows as

$$E_w \equiv \sum_{j=1}^{D} w_j E_j = \min_{\hat{\Gamma} \in \mathcal{E}(w)} \text{Tr} \left[ \hat{H}\hat{\Gamma} \right] .$$

where the minimizer reads

$$\hat{\Gamma}_w = \sum_{j=1}^{D} w_j |\Psi_j\rangle\langle\Psi_j| .$$

The GOK variational principle (6) applied to the N-particle Hamiltonian $H(\hat{h})$ in combination with the constrained search leads to an $w$-ensemble RDMFT in a straightforward manner:

$$E_w(\hat{h}) = \min_{\Gamma \in \mathcal{E}^N(w)} \text{Tr}_N[(\hat{h} + \hat{W})\hat{\Gamma}]$$

$$= \min_{\gamma \in \mathcal{E}_N^{\gamma}(w)} \min_{\Gamma \in \mathcal{E}^N(w)} \text{Tr}_N[(\hat{h} + \hat{W})\hat{\Gamma}]$$

$$= \min_{\gamma \in \mathcal{E}_N^{\gamma}(w)} \left[\text{Tr}_1[\hat{h}\gamma] + \mathcal{F}_w(\gamma)\right],$$

where

$$\mathcal{F}_w(\gamma) = \min_{\Gamma \in \mathcal{E}^N(w) \ni \gamma} \text{Tr}_N[\hat{W}\hat{\Gamma}].$$

It is worth noticing that for $w_0 \equiv (1,0,\ldots)$ we recover ground state RDMFT and (8) reduces indeed to (3). Moreover, Eq. (8) defines the universal $w$-ensemble functional $\mathcal{F}_w(\gamma)$ in a similar fashion as Eq. (3) defines the universal ground state functional $\mathcal{F}$. The domain of $\mathcal{F}_w$ is given by those 1RDMs $\gamma \in \mathcal{E}_N^{\gamma}(w)$ which follow from an $N$-boson density operator $\hat{\Gamma} \in \mathcal{E}^N(w)$ by tracing out $N-1$ particles. Thus, an $w$-ensemble $N$-representability problem arises. Due to the nonlinear spectral restriction of $\mathcal{E}^N$ to $\mathcal{E}_N(w)$, both sets $\mathcal{E}^N(w)$ and $\mathcal{E}_N^{\gamma}(w)$ are not convex in striking contrast to $\mathcal{E}^N$ and $\mathcal{E}_N$, respectively. In analogy to Levy’s ground state RDMFT, the description of $\mathcal{E}_N^{\gamma}(w)$ would involve for any $w$ highly involved additional constraints on the natural occupation numbers.

B. Convex relaxation of $w$-ensemble RDMFT

Since the $w$-ensemble $N$-representability problem is too intricate, the excited state RDMFT introduced in Sec. III A is not feasible from a practical point of view: Without knowing the functional’s domain $\mathcal{E}_N^{\gamma}(w)$, the process of deriving functional approximations cannot be initiated. Yet, any non-convex minimization problem can be turned into a corresponding convex one, at least in principle [13]. This crucial observation from convex analysis allows us to circumvent the too involved $w$-ensemble $N$-representability constraints in the same way as Valone’s approach circumvented the generalized Pauli constraints (recall Sec. III A). Applied to the constrained search formalism in Eq. (8), this means to replace the universal $w$-ensemble functional $\mathcal{F}_w(\gamma)$ by its lower convex envelope,

$$\mathcal{F}_w(\gamma) \equiv \text{conv}(\mathcal{F}_w(\gamma)), \quad (10)$$

whose domain follows as the convex hull of $\mathcal{E}_N^{\gamma}(w)$,

$$\mathcal{E}_N^{\gamma}(w) \equiv \text{conv}(\mathcal{E}_N^{\gamma}(w)). \quad (11)$$

Since the partial trace map $\text{Tr}_{N-1}[\cdot]$ is linear, it commutes with the convex hull operation $\text{conv}(\cdot)$. This allows us to obtain a more concrete characterization of the convex hull of $\mathcal{E}_N^{\gamma}(w)$ (the proof is identical to the one of the same statement for fermions as presented in [39])

$$\mathcal{E}_N^{\gamma}(w) = N\text{Tr}_{N-1}[\mathcal{E}_N^{\gamma}(w)] = \bigcup_{w' \prec w} \mathcal{E}_N^{\gamma}(w'). \quad (12)$$

Here, a vector $w' \prec w \in \mathbb{R}^d$ is majorized by a vector $w \in \mathbb{R}^d$, $w' \prec w$, if and only if

$$w'_1 + \ldots + w'_k \leq w_1 + \ldots + w_k \forall 1 \leq k \leq d, \quad (13)$$

with equality for $k = d$. The arrow $\prec$ indicates that (13) refers to the vector entries arranged in decreasing order. In the following, we can always omit this superscript since the entries of weight vectors $w$ are already ordered decreasingly by definition. Moreover, the last equality in Eq. (12) leads to the following inclusion relation,

$$w' \prec w \Leftrightarrow \mathcal{E}_N^{\gamma}(w') \subset \mathcal{E}_N^{\gamma}(w). \quad (14)$$

Hence, the smaller the weight vector $w$ with respect to majorization, the smaller will be the set of relaxed $w$-ensemble $N$-representable 1RDMs, i.e. the domain of $\mathcal{F}_w$. We will illustrate this inclusion relation for several weight vectors $w$ in Fig. 4, where the shrinking volume of the respective domains is in evidence.

Clearly, the convex relaxation does not change the outcome for the energy $E_w$ and, in particular,

$$E_w = \min_{\gamma \in \mathcal{E}_N^{\gamma}(w)} \left[\text{Tr}_1[\hat{h}\gamma] + \mathcal{F}_w(\gamma)\right]. \quad (15)$$

It is worth noticing that the same relaxed $w$-ensemble RDMFT can be obtained by starting at the N-boson level, where $\mathcal{E}_N^{\gamma}(w) = \text{conv}\left(\mathcal{E}^N(w)\right)$. Thus, replacing the set $\mathcal{E}^N(w)$ by its convex hull in the constrained search formalism [13] immediately leads to a more concrete expression for the relaxed $w$-ensemble functional (using the same ideas as in [39])

$$\mathcal{F}_w(\gamma) = \min_{\mathcal{E}_N^{\gamma}(w) \ni \gamma} \text{Tr}_N[\hat{W}\hat{\Gamma}]. \quad (16)$$

The importance of the constrained search expression (16) of the relaxed $w$-ensemble functional $\mathcal{F}_w$ can hardly be overestimated. It will namely serve as the starting point for the construction of functional approximations. This and the general use of relaxed $w$-RDMFT are illustrated in Sec. VII and VIII for the symmetric Bose-Hubbard dimer and Bose-Einstein condensates in the regime of small depletion.

C. Determining the functional’s domain

A formal definition of the set $\mathcal{E}_N^{\gamma}(w)$ as in Sec. III B without a constructive approach to characterize it is apparently not sufficient for practical purposes. It would be unclear how to derive approximations of $\mathcal{F}_w$ and how to
minimize then the total energy functional $\text{Tr}_1[h\gamma] + \mathcal{F}_w$ over the set $\mathcal{E}_N^1(w)$. Hence, Eq. (16) only leads to a viable $w$-ensemble RDMFT if a compact description of the functional’s domain is found. In the following, we derive the vertex representation of the functional’s domain which takes effectively the form of a polytope. To this end, we will exploit a fruitful analogy between the mathematical problem of determining the set $\mathcal{E}_N^1(w)$ and the description of non-interacting bosons. In particular, this will allow us to resort to our physical intuition about non-interacting bosons, leading to a better understanding of the boundary of $\mathcal{E}_N^1(w)$.

**FIG. 1.** Illustration of the duality correspondence. Left: Schematic illustration of the minimization of $\text{Tr}_1[h\gamma]$ over a compact convex set $\mathcal{E}_N^1(w)$. Right: Performing the minimization for all possible “directions” $h$ determines the boundary of $\mathcal{E}_N^1(w)$.

According to a duality principle [38], every compact convex set can be characterized equivalently either through all its points or all its supporting hyperplanes. In the following, as it is illustrated in Fig. 1, we apply this to our compact convex set $\mathcal{E}_N^1(w)$. First, since $\text{Tr}_1[h\gamma] \equiv \langle h, \gamma \rangle_1$ defines an inner product, a notion of geometry exists on the underlying space of 1RDMs. Accordingly, any one-particle Hamiltonian $\hat{h}$ defines a direction within this space. Minimizing $\langle h, \gamma \rangle_1$ over all 1RDMs $\gamma \in \mathcal{E}_N^1(w)$ corresponds to shifting the hyperplane of constant value $\langle h, \gamma \rangle_1$ in direction $-\hat{h}$ until it touches the boundary of $\mathcal{E}_N^1(w)$. Performing this minimization for all one-particle Hamiltonians $\hat{h}$ determines $\mathcal{E}_N^1(w)$ entirely. Since the spectral vector $w$ refers primarily to the $N$-boson level, we lift the minimization to the $N$-boson level through

$$\min_{\gamma \in \mathcal{E}_N^1(w)} \text{Tr}_1[h\gamma] = \min_{\Gamma \in \mathcal{E}^1(w)} \text{Tr}_1[h\Gamma].$$

Furthermore, we observe that the set $\mathcal{E}_N^1(w)$ is invariant under unitary transformations,

$$\hat{u}\mathcal{E}_N^1(w)\hat{u}^\dagger = \mathcal{E}_N^1(w),$$

for all unitaries $\hat{u}$ acting on the one-particle Hilbert space $\mathcal{H}_1$. Accordingly, we can restrict our procedure of characterizing $\mathcal{E}_N^1(w)$ through supporting hyperplanes to one-particle Hamiltonians $\hat{h}$ with an arbitrary but fixed eigenbasis, $\hat{h} = \sum_{i=1}^d h_i|\gamma_i\rangle\langle \gamma_i|$, and $h_1 \leq h_2 \leq \ldots \leq h_d$. Equivalently, the knowledge of the spectrum of a 1RDM $\gamma$ is sufficient to determine whether $\gamma$ belongs to the set $\mathcal{E}_N^1(w)$ or not. Accordingly, we are interested in describing the set of all admissible spectra $\lambda \equiv \text{spec}(\gamma)$,

$$\Sigma(w) \equiv \text{spec} \left( \mathcal{E}_N^1(w) \right),$$

As our following derivation reveals, $\Sigma(w)$ takes the form of a polytope, i.e., it is the convex hull of a finite number of vertices $v^{(l)}$. Furthermore, in agreement with [18], the spectral polytope $\Sigma(w) \subset \mathbb{R}^d$ is invariant under permutations of the Cartesian coordinates. Without loss of generality we therefore focus for a moment on the set $\Sigma^i(w)$ of decreasingly ordered vectors

$$\Sigma^i(w) = \Sigma(w) \cap \Delta,$$

where $\Delta$ denotes the set

$$\Delta = \{ \lambda \in \mathbb{R}^d | N \geq \lambda_1 \geq \lambda_2 \geq \ldots \geq \lambda_d \geq 0 \}. $$

In contrast to the fermionic case [38], the natural occupation numbers $\lambda_i$ in the set $\Delta$ do not obey the Pauli exclusion principle. This in turn results in a different underlying combinatorial structure leading to a qualitative different characterization of $\Sigma^i(w)$ than in the fermionic case [39]. Before presenting the general procedure for calculating $\Sigma^i(w)$, we illustrate in the left panel of Fig. 2 relation (20) between the three sets $\Sigma(w)$ (grey), $\Sigma^i(w)$ (overlap between grey and blue) and $\Delta$ (blue) for $(N,d) = (2,3)$. Due to the normalization of $\lambda$ to the total particle number, we can omit the third natural occupation number $\lambda_3$, and focus solely on $\lambda_1$ and $\lambda_2$. The spectral polytope $\Sigma^i(w)$ is restricted through the two equalities on the natural occupation numbers $\lambda_1$ and $\lambda_2$ indicated by the dashed lines (see Sec. 1B for their derivation). The red point marks the position of the single independent vertex $v$ (the other five vertices follow from $v$ by permutation of its entries). We provide a systematic derivation of $\Sigma^i(w)$ and the vertex $v$ in the following. To illustrate the differences to the fermionic case discussed in Ref. [35, 39], we show in the right panel of Fig. 2 the spectral polytopes for $N = 2$ fermions in $d = 3$ orbitals and the same choice of the weight vector $w$.

To calculate the vertices $v^{(l)}$ of the bosonic spectral polytope $\Sigma^i(w)$, we start by determining for every set of energy levels $h \equiv h^i = (h_1, \ldots, h_d)$ the sequence

$$h \rightarrow \tilde{\Gamma} \rightarrow \tilde{\gamma} \rightarrow v^i.$$

Here, $\tilde{\Gamma}$ denotes the minimizer state of the right side of Eq. (17) and $v = \text{spec}(\tilde{\gamma})$ the spectrum of the resulting 1RDM $\tilde{\gamma}$. The eigenstates of a one-particle Hamiltonian $\hat{h}$, represented by the vector $h$, on the $N$-boson
Hilbert space are given by the configuration states $|i⟩ = |i_1, \ldots, i_N⟩$. To determine in a systematic manner all distinct vertices $v$ in Eq. (22), we first introduce a partial ordering of configurations $i$, where a configuration is an element of the set

$$I_{N,d} = \{ i = (i_1, \ldots, i_N) | 1 \leq i_1 \leq i_2 \leq \ldots \leq i_N \leq d \}.$$  

Furthermore, we can assign an energy to every configuration through $h$ and thus order different configurations according to their energy. Then, for any two configurations $i$ and $j$ we define the partial ordering

$$i \leq j \iff \sum_{k=1}^{N} h_{ik} \leq \sum_{k=1}^{N} h_{jk} \ \forall \ h \equiv h^\dagger$$

$$\iff i_k \leq j_k \ \forall \ 1 \leq k \leq N.$$  

Hence, the partial order of configurations in $I_{N,d}$ is completely characterized by the eigenenergies of a chosen one-particle Hamiltonian $\hat{h}$. The partial ordering (24) leads for each choice of $h$ to a corresponding lineup $l$,

$$i_1 \rightarrow i_2 \rightarrow \ldots \rightarrow i_r,$$  

of the energetically lowest configurations $i_j$. Due to reasons that become clear in the following, the length of those lineups is restricted to the number $r$ of non-vanishing weights $w_j$ in the weight vector $w$. As an illustration, we show in the left panel of Fig. 3 the structure of this “excitation spectrum” for $N = 3$ bosons following from the partial ordering of configurations in Eq. (24). For $r = 1$, the lineup (25) consists of only one configuration $(1,1,1)$. For $r = 2$, we have again a single lineup $(1,1,1) \rightarrow (1,1,2)$ now consisting of two configurations. Note that according to (6), $w$-RDMFT with $r = 2$ is already sufficient for calculating the ground state energy and its gap to the first excited state. For $r = 3$ there are now two choices of the second excitation leading to two lineups. Furthermore, for larger values $r \leq 4$, both configurations $(1,1,3)$ and $(1,2,2)$ must appear before $(1,2,3)$, a pattern which can be easily continued to derive all distinct lineups (25) required to calculate the vertex representation of $\Sigma(w)$. The right panel of Fig. 3 shows the structure of the excitation spectrum for $N = 3$ fermions discussed in detail in Ref. [38, 39] to highlight the differences between the fermionic and bosonic settings.

Determining all possible lineups $l$ (25) is absolutely essential because each of them defines one resulting $N$-boson minimizers $\hat{\Gamma}$ in (22) and in that sense gives rise to the excitation spectrum $\Sigma(w)$. According to the GOK variational principle (6), the density operator corresponding to (25) reads

$$\hat{\Gamma} = \sum_{j=1}^{r} w_j |i_j⟩⟨i_j|.$$  

Using the spectrum of the corresponding 1RDM $\hat{\gamma}$ in (22) we eventually obtain the $d$-dimensional vertex

$$v^{(l)} = \sum_{j=1}^{r} w_j n_{i_j},$$  

where $n_{i_j} = \text{spec}(NTr_{N-1} [|i_j⟩⟨i_j|])$ is an occupation number vector corresponding to the configuration $i_j$. The $k$-th entry of $n_{i_j}$ equals the number of $k$’s contained in $i_j$. Thus, by referring to the partial ordering (24), e.g., the lowest configuration $(1,1,\ldots,1)$ corresponds to the occupation number vector $n_{(1,1,\ldots,1)} = (N,0,\ldots,0)$.
Finally, the spectral polytope $\Sigma(w)$ follows as the convex hull of all possible permutations of the entries of all vertices $v(l)$ (recall (20)),

$$\Sigma(w) = \text{conv}\left( \{ \pi(v(l)) \mid l = 1, \ldots, R, \pi \in S^d \} \right), \quad (28)$$

where $S^d$ denotes the permutation group of a set of $d$ elements (here the entries $v(l)$) and $R$ the number of independent vertices $v(l)$. Hence, $\Sigma(w)$ takes indeed the form of a polytope as anticipated above.

In Fig. 4 as a further illustration of the spectral polytopes, we demonstrate the important inclusion relation Eq. (14) for the setting $(N, d) = (2, 3)$. Indeed the six spectral polytopes $\Sigma(w)$ (and $\Sigma'_N(w)$) are contained in each other since the six respective weight vectors $w$ are related by majorization.

So far, we developed a general strategy for determining the vertex representation of the spectral polytope $\Sigma(w)$ for a given total particle number $N$ and a $d$-dimensional one-particle Hilbert space $H_1$. Since experiments and theoretical studies are often performed for different particle numbers, we next comment on the relation between the spectral polytopes for different settings $(N, d)$. For $N \geq r - 1$, increasing the particle number to $N' > N$ just means to add to each configuration $i_j$ in the lineups (25) another $N' - N$ bosons in the lowest orbital $|1\rangle$. Consequently, this does not change the number of lineups and their structure. Instead, it only alters the first entry of the corresponding natural occupation number vectors $v(l)$ which indeed depends explicitly on $N$. This also implies that for fixed $d \geq r$ the vertices $v(l)$ for $N$ and $N' > N$ are related through

$$v(l)_{N'} = v(l)_{N} + \delta e_1, \quad (29)$$

where $\delta = N' - N$ and $e_1 = (1, 0, \ldots)$. Note that the polytope of a higher dimensional setting $(N', d')$ with $d' > d$ can be obtained from $\Sigma_N(w)$ by the following two steps. First, one extends the $d$-dimensional vector $v_N(l)$ to a $d'$-dimensional vector by adding zero entries and afterwards uses the relation (29) to increase the particle number. Yet, for the sake of simplicity we compare in the following spectral polytopes $\Sigma_N(w)$ and $\Sigma_N'(w)$ only for fixed $d$. By applying a generalization of Radó’s theorem (39) (see also Eq. (47)) and using Eq. (29), we find that

$$\Sigma_{N'}(w) \equiv \Sigma_N(w) + C, \quad (30)$$

where $C$ is a rescaled simplex with edge length $\delta$,

$$C \equiv \text{conv}\left( \{ \pi(\delta e_1) \mid \pi \in S^d \} \right). \quad (31)$$

The sum of the two sets in Eq. (30) is nothing else
than the Minkowski sum of two permutation invariant polytopes which means that \( \Sigma_N^r(w) = \Sigma_N(w) + \mathcal{C} = \{ \lambda + c | \lambda \in \Sigma_N(w), c \in \mathcal{C} \} \). Clearly, the sum of two convex sets is also convex. Also, every \( \mu \in \Sigma_N^r(w) \) is correctly normalized since all \( \lambda \in \Sigma_N(w) \) are normalized to \( N \) and all \( c \in \mathcal{C} \) are normalized to \( \delta \). In Fig. 5 we illustrate the Minkowski sum in Eq. (30) for \( d = 3 \) and the two particle numbers \( N' = 5 \) and \( N = 3 \). The spectral polytope \( \Sigma_5(w) \) (grey) is obtained by adding the elements of \( \Sigma_3(w) \) (green) and the rescaled simplex \( \mathcal{C} \) (dashed). Due to the normalization of all occupation number vectors in \( \Sigma_5(w) \) and \( \Sigma_3(w) \) to the respective total particle number \( N' \) or \( N \), we can omit the value of \( \lambda_3 \).

### IV. Bosonic Exclusion Principle

For practical purposes, using the vertex representation of the spectral polytope \( \Sigma(w) \) to check whether a natural occupation number vector \( \lambda \) belongs to \( \Sigma(w) \) or not is highly inefficient. Yet, every polytope can equivalently be described through its halfspace representation [48]. Then, testing the membership of \( \lambda \) reduces to checking only finitely many linear conditions \( D_k(\lambda^i) \geq 0 \). Actually, these linear constraints represent nothing else than an exclusion principle for bosonic quantum systems in mixed states, in conceptual analogy to Pauli’s exclusion principles for fermions. A simple procedure for translating the vertex representation of \( \Sigma(w) \) into a halfspace representation is presented for \( r \leq 3 \) in Sec. IV while a more general mathematical procedure applicable to arbitrary \( r \) can be found in Ref. [50].

Especially for bosons, the existence of those constraints might be surprising at first sight. This is due to the fact that the solution of the pure state \( N \)-representability problem for bosons is trivial, i.e. there are no constraints on occupation numbers beyond normalization and positivity [15, 22]. Furthermore, it is worth emphasizing here that the scope of these exclusion principle constraints is not restricted to the natural orbital basis. According to the Schur-Horn theorem [51, 52] they namely apply to the occupation numbers of any orthonormal basis. As it is explained in Sec. VII, they are thus potentially relevant in GOK-DFT applied to bosonic lattice models.

There are two particularly noteworthy additional structural aspects concerning the bosonic exclusion principle. First, the linear inequalities \( D_k(\lambda^i) \geq 0 \) are effectively independent of the total boson number \( N \) and the dimension \( d \) of the one-particle Hilbert space. The only requirement for this statement to be valid is that \( N \) and \( d \) are large enough such that

\[
N \geq r - 1, \quad d \geq r,
\]

which can generally be assumed. This independence of the inequalities of \( N \) and \( d \) is in striking contrast to the generalized Pauli constraints for fermions in ground state RDMFT for pure states [20]. There, different \( N \) and \( d \) give rise to different inequalities which in turn makes their derivation tremendously complicated [45–47, 53, 54] and their application practically impossible.

Second, there is even a hierarchy of bosonic exclusion principle constraints. To be more specific, all constraints derived for a value \( r \) are still contained in the minimal hyperplane representation for any \( r' > r \), i.e., they remain facet-defining. Or in other words, the constraints for \( r' \) are given by those for \( r' - 1 \), complemented by a few additional new ones. A concrete illustration of this hierarchy is presented in Sec. V for small \( r \). To demonstrate the above statement, we provide in Tab. I the number of vertices \( v^{(j)} \) and the corresponding number of inequalities for \( r \leq 12 \). The comprehensive derivation of those halfspace conditions from the vertex representation of \( \Sigma(w) \) for arbitrary \( r \) is presented in Ref. [50].

| \( r \) | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|----|----|----|
| \# v^{(j)} | 1 | 2 | 4 | 8 | 17 | 37 | 82 | 184 | 418 | 967 | 2278 |
| \# ineq. | 1 | 2 | 3 | 5 | 8 | 13 | 22 | 36 | 59 | 99 | 171 | 299 |

| TABLE I. Number of generating vertices \( v^{(j)} \) and number of exclusion principle constraints defining \( \Sigma(w) \) for \( r \leq 12 \). These numbers are independent of \( N \) and \( d \), provided [42] is respected. |

### V. Derivation of Inequalities for Small Numbers of Non-Vanishing Weights

In this section, also as an illustration of Secs. III C, IV, we derive for \( r \leq 3 \) the bosonic exclusion principle constraints. This means to first determine the vertex
representation of Σ(ν) and then to turn it into a halfspace representation by using the elegant concept of vector majorization. We also explain how possible redundant inequalities can be identified in order to obtain a minimal halfspace representation of Σ(ν). Throughout this section, we consider arbitrary N and d. This then also demonstrates how the resulting facet-defining inequalities can be extended to settings of larger N′ and d′, including the important complete basis set limit d′ → ∞.

A. r = 1 non-vanishing weight

We start by deriving the hyperplane representation of the spectral polytope Σ(ν) for r = 1 and verify as a consistency check that we recover the trivial solution of the bosonic one-body pure N-representability problem. As discussed in the context of Fig. 3 there is only one lineup [25] and it consists of the single configuration (1, . . . , 1). According to [22], this yields one ν-minimizer,

\[ \hat{v} = |1, \ldots, 1|1, \ldots, 1 \] \tag{33}

leading through [22] to the single vertex ν = (N, 0, . . .). We have thus obtained the vertex representation of Σ(ν) introduced in Eq. [28]. Hence, Σ(ν) is the permutohedron

\[ \Sigma(ν) = \text{conv}(\{π(ν) | π ∈ S^d\}) , \] \tag{34}

which is nothing else than the convex hull of all possible permutations of the entries of the vertex ν. By Rado’s theorem [55], [54] is equivalent to

\[ \Sigma(ν) = \{ λ ∈ ℝ^d | λ ≪ ν \} . \] \tag{35}

In particular, we have

\[ \Sigma_N^0(ν) = \{ \hat{γ} ∈ Σ_N^0 | \text{spec}(\hat{γ}) ≪ ν \} . \] \tag{36}

It is exactly the majorization condition λ ≪ ν which defines the sought-after hyperplane representation of Σ(ν), i.e., the bosonic exclusion constraints. Here, λ ≪ ν implies that

\[ \begin{align*} λ_i^1 & \leq N , \\
λ_i^1 + λ_i^2 & \leq N , \\
& \vdots \\
\sum_{i=1}^d λ_i^1 & = N . \end{align*} \tag{37} \]

Assuming λ_i^1 ≥ 0, all inequalities (37) are satisfied as a direct consequence of the normalization and are therefore redundant. Thus, we indeed recover the solution of the one-body pure N-representability problem with the well-known trivial constraints 0 ≤ λ_i ≤ N.

B. r = 2 non-vanishing weights

Next, we consider r = 2 non-vanishing weights with w_1 + w_2 = 1. By referring to the excitation spectrum in Fig. 3 we have only one lineup [25] which reads

\[ (1, \ldots, 1) → (1, \ldots, 1, 2) . \] \tag{38}

The corresponding minimizer \( \hat{v} \) is given by

\[ \hat{v} = w_1|1, \ldots, 1|1, \ldots, 1, 1 + w_2|1, \ldots, 1, 1, 1| \] \tag{39}

and according to [22] and [27] we obtain the vertex ν = (N − 1 + w_1, w_2, 0, . . .). Since there exists only one ν as for r = 1, we can again apply Rado’s theorem [55] to obtain a minimal hyperplane representation of Σ(ν) and it follows that

\[ \Sigma_N^1(ν) = \{ \hat{γ} ∈ Σ_N^1 | \text{spec}(\hat{γ}) ≪ ν \} . \] \tag{41}

Then, the majorization condition λ ≪ ν enforces the constraints

\[ \begin{align*} λ_1^1 & \leq N − 1 + w_1 , \\
λ_1^1 + λ_2^1 & ≤ N , \\
& \vdots \\
\sum_{i=1}^d λ_i^1 & = N . \end{align*} \tag{42} \]

Hence, all constraints except the first and last one are redundant, and we arrive at the minimal hyperplane representation

\[ \begin{align*} λ_1^1 & \leq N − 1 + w_1 , \\
\sum_{i=1}^d λ_i^1 & = N . \end{align*} \tag{43} \]

Moreover, in agreement with Tab. IV there is only one additional constraint in the first line of Eq. (43) compared to r = 1. In particular, the inequalities of a minimal hyperplane representation for r = 1 and r = 2 represent the first two levels of the hierarchy of bosonic exclusion principles.

C. r = 3 non-vanishing weights

According to the excitation spectrum in Fig. 3 there exist two lineups

\[ \begin{align*} (1): & \quad (1, \ldots, 1) → (1, \ldots, 1, 2) → (1, \ldots, 1, 3) , \\
(2): & \quad (1, \ldots, 1) → (1, \ldots, 1, 2) → (1, \ldots, 1, 2, 2) . \end{align*} \tag{44} \]


These two lineups correspond to the two vertices (recall that $w_1 + w_2 + w_3 = 1$

$$v^{(1)} = (N - 1 + w_1, w_2, 1 - w_1 - w_2, 0, \ldots),$$

$$v^{(2)} = (N - 2 + 2w_1 + w_2, 2 - 2w_1 - w_2, 0, \ldots).$$

(45)

Since there are now two vertices $v^{(1)}$ and $v^{(2)}$, Rado’s theorem used for $r = 1, 2$ to obtain the hyperplane representation of $\Sigma(w)$ in (28) does not apply anymore. Instead, we can make use of a generalization of Rado’s theorem introduced in Ref. [39]. It states that for vectors $v^{(1)}, \ldots, v^{(R)} \in \mathbb{R}^d$, the polytope

$$\mathcal{P} = \text{conv} \left( \left\{ \pi(v^{(j)}) \mid j = 1, \ldots, R, \pi \in S^d \right\} \right)$$

is equivalent to

$$\mathcal{P} = \left\{ \lambda \mid \exists \text{ conv. comb.} \sum_{j=1}^{R} p_j v^{(j)} = v : \lambda \prec v \right\}.$$  (47)

Applied to the spectral polytope $\Sigma(w)$ (28) with $R = 2$, this implies that $\lambda \in \Sigma(w)$ if and only if there exists a convex combination $u = q v^{(1)} + (1 - q) v^{(2)}$ such that $\lambda \prec u$. This majorization condition leads to the constraints

$$\lambda^1_1 \leq N - 2 + 2w_1 + w_2 + q(1 - w_1 - w_2),$$

$$\lambda^2_1 + \lambda^2_2 \leq N - q(1 - w_1 - w_2),$$

$$\lambda^2_1 + \lambda^2_2 + \lambda^3_1 \leq N,$$

$$\vdots$$

$$\sum_{i=1}^{d} \lambda^i_1 = N.$$  (48)

Here, all inequalities except the first two are redundant since they are automatically satisfied due to the normalization of $\lambda$. To derive a minimal hyperplane representation, we need to eliminate the parameter $q$. Therefore, we first notice that the upper bound on $\lambda^1_1$ can vary between $N - 2 + 2w_1 + w_2$ and $N - 1 + w_1$ since $q$ is restricted to $q \in [0, 1]$. Moreover, the upper bound on $\lambda^2_1 + \lambda^2_2$ can vary between $N - 1 + w_1 + w_2$. Thus, it must always hold that $\lambda^1_1 \leq N - 1 + w_1$ which requires to adjust the value of $q$ according to

$$q \geq \frac{\lambda^1_1 - N + 2 - 2w_1 - w_2}{1 - w_1 - w_2}.  \quad (49)$$

This inequality is then used to tighten the upper bound on $\lambda^1_1 + \lambda^2_1$ to $2(N - 1) + 2w_1 + w_2 - \lambda^3_1$. Thus, our linear constraints determining a minimal hyperplane representation of the spectral polytope are given by

$$\lambda^1_1 \leq N - 1 + w_1,$$

$$2\lambda^1_1 + \lambda^2_1 \leq 2(N - 1) + 2w_1 + w_2,$$

$$\sum_{i=1}^{d} \lambda^i_1 = N.$$  (50)

Thus, the second inequality is the only additional new one compared to $r = 2$. This illustrates again the hierarchy of exclusion principle constraints for bosons introduced and explained in Sec. [1].

The mathematical formalism to derive systematically the halfspace representation of the spectral polytopes for larger values of $r$ is presented in Ref. [50].

VI. APPLICATION TO SYMMETRIC BOSE-HUBBARD DIMER

In this section, we illustrate the $w$-ensemble RDMFT for bosons introduced in Sec. [1] for the symmetric Bose-Hubbard dimer. Understanding the $w$-ensemble functional and its domain for this model is particularly interesting since the Bose-Hubbard dimer constitutes the building block of the Bose-Hubbard model widely used in the field of ultracold quantum gases. The Hamiltonian for spinless bosons on two lattice sites reads

$$\hat{H} = -t \left( \hat{a}^\dagger_L \hat{a}_R + \hat{a}^\dagger_R \hat{a}_L \right) + U \sum_{j=L,R} \hat{n}_j (\hat{n}_j - 1),$$  (51)

where the first term describes hopping at a rate $t$ between the left ($L$) and right ($R$) lattice site. The second term describes the Hubbard on-site interaction with coupling strength $U$ and $\hat{n}_j = \hat{a}^\dagger_j \hat{a}_j$ is the occupation number operator. In the case of periodic boundary conditions, the Hamiltonian in Eq. (51) is translationally invariant. This implies that the total momentum $\hat{P}$ is conserved, i.e., $\hat{P}$ is a good quantum number. As a result, the minimization in the constrained search formalism in Eq. (5) can be restricted to all $\hat{P} \in \mathcal{E}^N(\omega, P)$ in the chosen symmetry sector with fixed $P$. Then, it is possible to establish a separate functional in each symmetry sector instead of a single more involved functional referring to all $\hat{P}$. Moreover, every 1RDM $\hat{\gamma}$ is diagonal in momentum representation. In the following, we consider the case of $N = 2$ spinless bosons and restrict to repulsive interactions, i.e., $U > 0$. Then, the natural occupation numbers are given by the momentum occupation numbers $n_p \geq 0$ restricted through the normalization $\sum_p n_p = 2$.

In the following, we consider the $P = 0$ momentum sector. For two lattice sites, the single particle momentum can take the discrete values $p_v = \pi v$ with $v = 0, 1$. We denote the creation (annihilation) operator referring to the momentum $\nu$ by $\hat{a}^\dagger_v, (\hat{a}_v)$ and the occupation number operators by $\hat{n}_v = \hat{a}^\dagger_v \hat{a}_v$. The only two configurations satisfying $\sum_{v=1,2} \nu \bmod 2 = 0$ are $(0, 0)$ and $(1, 1)$ corresponding to the two basis states $|1\rangle = \frac{1}{\sqrt{2}}(\hat{a}^\dagger_0)^2|0\rangle$ and $|2\rangle = \frac{1}{\sqrt{2}}(\hat{a}^\dagger_1)^2|0\rangle$, where $|0\rangle$ denotes the vacuum state. Since $(0, 0)$ and $(1, 1)$ are the only allowed configurations in the $P = 0$ sector, it follows from Sec. [14] that the larger value of $n_0$ and $n_1 = 2 - n_0$ is bounded from above by $2w_1$ and the lower one from below by $2w_2$. In particular, this means that the domain $\mathcal{E}^N_\lambda(\omega)$ of the $w$-
ensemble functional $F_w$ is already convex,

$$\Sigma(w, P = 0) = \left\{ n_0 \mid 0 \leq 2w_2 \leq n_0 \leq 2w_1 \leq 2 \right\}. \quad (52)$$

Then, minimizing the expectation value $\text{Tr}_2[\hat{W}^\dagger]\hat{W}$ according to the constrained search formalism, where $\hat{W}$ is the second term in the Hamiltonian [51], leads to (see Appendix B)

$$F_w(n_0) = U \left( 1 - \sqrt{n_0(2 - n_0)} - 4w_1w_2 \right)$$
$$= U \left( 1 - \sqrt{(n_0 - 2w_2)(2w_1 - n_0)} \right). \quad (53)$$

Since $F_w(n_0)$ is already convex, it is equal to the relaxed functional $\overline{F}_w(n_0)$.

It is also worth noticing that for the symmetric Bose-Hubbard dimer the translational invariance is equivalent to the inversion symmetry. To explain this, we now skip the periodic boundary conditions and instead restrict to the even symmetry sector. The corresponding symmetry-adapted one-boson basis consists of the two states $|e\rangle = (|L\rangle + |R\rangle)/\sqrt{2}$ and $|o\rangle = (|L\rangle - |R\rangle)/\sqrt{2}$ which actually coincide with the two one-particle momentum states. The two-dimensional subspace with even inversion-symmetry is then spanned by the two basis states $|e, e\rangle$ and $|o, o\rangle$. This implies that the 1RDM $\hat{\gamma}$ is diagonal and thus depends on only one free parameter $n_e$, the occupation number of $|e\rangle$. Thus, the resulting $w$-ensemble functional $F_w$ is equivalent to $\overline{F}_w$ in the symmetric Bose-Hubbard dimer with periodic boundary conditions and restricted to $P = 0$ in (53) with $n_0$ replaced by $n_e$.

The two equivalent expressions of $F_w(n_0)$ in Eq. (53) illustrate two different properties of the universal functional. From the first line together with (52) it follows immediately that $F_w(n_0)$ is symmetric around $n_0 = 1$. The second expression in (53) emphasizes the diverging behaviour of the gradient of $F_w(n_0)$ at the boundary of its domain for each weight $w_1 = 1 - w_2$. Indeed, the derivative of $F_w(n_0)$ with respect to $n_0$ diverges at the boundary $\partial \Sigma$ of the domain of $F_w$ as

$$\left| \frac{\partial F_w(n_0)}{\partial n_0} \right| \sim \frac{1}{\text{dist}(n_0, \partial \Sigma)}. \quad (54)$$

The sign of the gradient reveals that the corresponding force is repulsive, i.e., it prevents $n_0$ from ever reaching the boundary $\partial \Sigma$.

The universal functional $\overline{F}_w(n_0)$ is illustrated for several values of $w_1$ in Fig. 6. This also demonstrates the inclusion relation [14], i.e., for $w' < w$ (corresponding here to $w'_1 \leq w_1$) we have $\Sigma(w', P = 0) \subset \Sigma(w, P = 0)$.

To further illustrate $w$-ensemble RDMFT we calculate the energy $E_w$ by minimizing the total energy functional $\text{Tr}_2[\hat{\gamma}^\dagger] + F_w(n_0)$, where $\hat{\gamma}$ is given by the first term in (51) and $\text{Tr}_2[\hat{\gamma}^\dagger] = -4t(n_0 - 1)$. Solving

$$\frac{\partial}{\partial n_0} (-4t(n_0 - 1) + F_w(n_0)) \bigg|_{n_0 = n_0} = 0 \quad (55)$$

for the minimizer $\tilde{n}_0$ and substituting the result into the energy functional yields for the weighted sum $E_w$ of the two eigenenergies $E_1, E_2$ according to Eqs. (6) and (8),

$$E_w = w_1 \left( U - \sqrt{16t^2 + U^2} \right) + w_2 \left( U + \sqrt{16t^2 + U^2} \right). \quad (56)$$

Note that this result is in agreement with the eigenenergies $E_1$ and $E_2$ obtained from an exact diagonalization of the Hamiltonian $\hat{H}$ in Eq. (51) (see Appendix B for further details).

VII. APPLICATION TO BOSE-EINSTEIN CONDENSATES

The application of $w$-ensemble RDMFT to Bose-Einstein condensates (BECs) is appealing due to a number of reasons. First, as already stressed in the introduction the Penrose and Onsager criterion [14, 16] for BEC identifies RDMFT as a particularly suitable approach to BECs. Second, recent analyses of their ground states have revealed an intriguing new concept, namely the existence of a BEC-force [15, 16, 56]. The question arises whether this force based on the one-particle picture is also present in excited BECs. Third, the comprehensive understanding of the regime of small quantum depletion through Bogoliubov theory provides excellent prospects for deriving a corresponding approximation of the universal functional. This actually allows us to provide three conceptually different derivations of the $w$-ensemble universal functional in the following.

To commence, we consider a dilute homogeneous BEC in a three-dimensional box with volume $V$. In second quantization, the general Hamiltonian $\hat{H} = \hat{h} + \hat{W}$ for interacting bosons in momentum representation reads

$$\hat{H} = \sum_p t_p \hat{a}_p^\dagger \hat{a}_p + \frac{1}{2V} \sum_{p, q, k} W_p \hat{a}_{q+p-k}^\dagger \hat{a}_{q-k} \hat{a}_q \hat{a}_k, \quad (57)$$

where $\hat{a}_p^\dagger$ and $\hat{a}_p$ are the bosonic creation and annihilation operators, $W_p$ the Fourier coefficients of the pair
interaction between the bosons and $t_p$ denotes the dispersion. By assuming a macroscopic occupation of the $p = 0$ momentum state in the homogeneous BEC under consideration, the well-known Bogoliubov approximation simplifies the general Hamiltonian in \[ (57) \] to \[ (58) \]

\[
\hat{H}_B = \sum_p t_p \hat{n}_p + \frac{1}{2V} \sum_{p \neq 0} W_p \left[ 2\hat{n}_0 \hat{n}_p + \left( \hat{a}_p^\dagger \hat{a}_p^\dagger \hat{a}_p^2 + \text{h.c.} \right) \right]
\]

where $\hat{n}_p = \hat{a}_p^\dagger \hat{a}_p$ is the occupation number operator and we omit the constant energy shift $\frac{N(N-1)\omega_0}{2V}$. Moreover, the Fourier coefficients satisfy $W_p = W_{-p}$ and we restrict to purely repulsive interactions such that $W_p \geq 0 \forall p$. Furthermore, the kinetic energy dispersion satisfies $t_p = t_{-p}$ with $t_0 = 0$. This implies in particular that the Bogoliubov approximated Hamiltonian \[ (58) \] is invariant under $p \to -p$. We therefore introduce a new index $p'$ which labels all pairs $(-p,p), p \neq 0$ and for each such pair $p'$ can be chosen to be either $p' = -p$ or $p' = p$ without loss of generality. Then, the Hamiltonian $\hat{H}_B$ in Eq. \[ (58) \] is equivalent to

\[
\hat{H}_B = \sum_{p'} t_{p'} \hat{n}_{p'} + \frac{1}{2V} \sum_{p'} W_{p'} \left[ \hat{n}_0 \hat{n}_{p'} + \left( \hat{a}_{p'}^\dagger \hat{a}_{p'}^\dagger \hat{a}_{p'}^2 + \text{h.c.} \right) \right]
\]

where we introduced the operator

\[
\hat{n}_{p'} \equiv \hat{n}_{p'} + \hat{n}_{-p'}.
\]

This notation emphasizes that the expectation value of the kinetic energy operator

\[
\text{Tr}_1[\hat{\tilde{H}}] = \sum_p t_p \hat{n}_p
\]

is completely determined by the pairs $(t_{p'}, \eta_{p'})$ for all $p'$ since $t_p = t_{-p}$. In particular, this implies that the vectors $t \equiv (t_{p'})_{p'}$ and $\eta \equiv (\eta_{p'})_{p'}$ constitute the conjugate variables in our functional theory, denoted by $t \leftrightarrow \eta$. Furthermore, it follows that the ground state functional $\mathcal{F}_{w_0}$ (recall that $w_0 = (1,0,\ldots)$) and the excited state functional $\mathcal{F}_w$ can be written as functionals of $\eta$ only, i.e. $\mathcal{F}_{w_0} = \mathcal{F}(\eta)$ and $\mathcal{F}_w = \mathcal{F}_w(\eta)$.

\section{Recap of ground state universal functional}

In this section, we derive the ground state universal functional $\mathcal{F}_{w_0}$. The calculation shown in the following uses the same concepts as in Ref. \[ 16 \] but derives $\mathcal{F}_{w_0}$ as a functional of $\eta$ rather than of the full occupation number vector $n$.

In the Bogoliubov theory, the interacting ground state of a BEC has the form $|\Psi_0\rangle = U|N\rangle$, where $|N\rangle = (N!)^{-1/2}(\hat{a}_0^\dagger)^N|0\rangle$ and $[58],[59]$

\[
\hat{U} = \exp \left\{ \frac{1}{2} \sum_{p \neq 0} \theta_p \left[ (\hat{\beta}_0^\dagger)^2 \hat{a}_p \hat{a}_{-p} - \hat{\beta}_0^2 \hat{a}_p^\dagger \hat{a}_{-p}^\dagger \right] \right\}
\]

is a unitary operator with variational parameters $\theta_p \in \mathbb{R}$. Here, the operator $\hat{\beta}_0 \equiv (\hat{n}_0 + 1)^{-1/2} \hat{a}_0$ [59] annihilates a boson with momentum $p = 0$ without creating a prefactor in front of the new state. In particular, $\hat{U}$ commutes with the particle number operator, $[\hat{U}, N] = 0$. Then, the $\mathbf{w}$-minimizer for $r = 1$ (referring to ground state RDMFT) is given by $\hat{\Gamma}_{w_0} = |\Psi_0\rangle|\Psi_0\rangle$ according to the GOK variational principle in Eq. \[ (6) \]. Usually it is assumed that $\theta_p = \theta_{-p}$. If we allowed, however, for $\theta_p \neq \theta_{-p}$ one could show that

\[
\hat{U}^\dagger \hat{a}_p \hat{U} \approx \frac{1}{\sqrt{1 - \phi_p^2}} \left( \hat{a}_p - \phi_p \hat{\beta}_0 \hat{a}_p^\dagger \right)
\]

with variational parameters

\[
\phi_p = \tanh \left( \frac{\theta_p + \theta_{-p}}{2} \right)
\]

satisfying also $\phi_p = \phi_{-p}$. To proceed, the ground state functional $\mathcal{F}_{w_0}$ is obtained by minimizing the expectation value $\langle \Psi_0 | \hat{W}_B | \Psi_0 \rangle = \langle N | \hat{U}^\dagger \hat{W}_B \hat{U} | N \rangle$ over the variational parameters $\phi_{p'}$. Here, $\hat{W}_B$ denotes the Bogoliubov approximated interaction $W_B = \hat{H}_B - \hat{\tilde{H}}$, where $\hat{\tilde{H}} = \sum_{p'} t_{p'} \hat{n}_{p'}$ is the kinetic energy operator. From Eq. \[ (64) \] and the definition of the ground state $|\Psi_0\rangle$ it follows that $\eta_{p'}$ and $\phi_{p'}$ are related through

\[
\eta_{p'} = \langle \Psi_0 | \hat{n}_{p'} | \Psi_0 \rangle \approx \frac{2 \phi_{p'}^2}{1 - \phi_{p'}^2}.
\]

The expression on the right hand side holds only approximately due to the approximation in Eq. \[ (64) \] and an estimate of its accuracy can be deduced from Ref. \[ 60 \]. Inverting this expression for the variational parameters $\phi_{p'}$ shows that the occupation numbers $\eta_{p'}$ determine the variational parameters $\phi_{p'}$ up to phases $\sigma_{p'} = \pm 1$. This simplifies the minimization in Levy’s constrained search over all $\phi_{p'}$ to a minimization over the phases $\sigma_{p'}$ according to

\[
\mathcal{F}_{w_0}(\eta) = \min_{\{\sigma_{p'} = \pm 1\}} \left\{ \frac{n \sum_{p'} W_{p'} \eta_{p'} - \sigma_{p'} W_{p'} \sqrt{\eta_{p'} (\eta_{p'} + 2)}}{n \sum_{p'} W_{p'} \left( \eta_{p'} - \sqrt{\eta_{p'} (\eta_{p'} + 2)} \right)} \right\}
\]

where we used $W_{p'} \geq 0 \forall p'$ in the last line and $n = N/V$ denotes the particle density. As a consistency check, we note that the universal functional \[ (66) \] is indeed equivalent to the one derived in Ref. \[ 16 \] after replacing in the latter the momentum occupation numbers $\eta_{p'}$ by $\eta_{p'}/2$. 


B. Excitations within Bogoliubov theory

In the following we recall the most important aspects of the excitation spectrum of a homogeneous Bose gas within the Bogoliubov approximation. This serves as a preliminary for the derivation of the excited state functional \( \mathcal{F}_w \) in Sec. VII E.

The ground state energy of the Bogoliubov approximated Hamiltonian \( \hat{H}_B \) is given by [58, 60]

\[
E_0 = -\frac{1}{2} \sum_{\mu \neq 0} \left( nW_p + t_p - \sqrt{t_p(t_p + 2nW_p)} \right),
\]

and the same result also holds approximately within the particle number conserving Bogoliubov theory up to a controllable error [58, 60]. Moreover, the energy spectrum consists of elementary excitations of the ground state and takes the form [62, 58]

\[
E = E_0 + \sum_{\mu > 0} \omega_p \mu_p.
\]

Here, \( \mu_p \) counts the number of quasiparticles with momentum \( p \) created by acting with the quasiparticle operator [58]

\[
\hat{c}^\dagger_p = \hat{U} \hat{a}^\dagger_p \hat{U}^{-1} \hat{\beta}_0
\]

\[
\approx \frac{1}{\sqrt{1 - \phi_p}} \left( \hat{a}^\dagger_p \hat{\beta}_0 + \phi_p \hat{\beta}_0 \hat{a}^\dagger_p \right),
\]

on the interacting ground state \( |\Psi_0\rangle \), and \( \omega_p \) denotes the quasiparticle dispersion

\[
\omega_p = \sqrt{t_p(t_p + 2nW_p)}.
\]

For small enough quantum depletion and low-lying excited states, (68) holds in good approximation also for the particle number conserving Bogoliubov Hamiltonian with [58] and, in particular, \( \mu_p \approx n_p \) in Eq. (68). Furthermore, \( \omega_p = \omega_p \) and we replace accordingly \( p \) by \( p' \) in the derivation of the \( w \)-ensemble functional.

Before we can present three different instructive derivations of the \( w \)-ensemble functional \( \mathcal{F}_w \) for targeting the ground state and the first excited state, we discuss two critical conceptual aspects of \( w \)-ensemble RDMFT. Both Secs. VII C and VII D and their conclusions are not restricted to BECs but are valid for \( w \)-RDMFT applied to arbitrary quantum systems of bosons or fermions.

C. Crossing of energy levels

The energy levels of many-body quantum systems can cross as one varies system parameters such as the coupling constants of two-body interactions or the strength of an external field. A particularly prominent example is given by quantum phase transitions for which the ground state and first excited state cross. This in turn manifests itself in the context of functional theories in the form of nonanalyticities of the universal functional: By referring to the constrained search formalism, the \( \gamma \)-fermion minimizer for 1RDMs belonging to different quantum phases are not necessarily analytically connected anymore. As a consequence, the functional’s domain would split into different cells (subdomains) and one would need to derive an analytical functional for each them separately. At the borders of these cells those different functionals would be “glued” together continuously.

In the context of excited state RDMFT this reasoning would apply to various energy levels of interest, i.e., the lowest \( r \) ones in \( w \)-RDMFT. Accordingly, there will be many more relevant crossings and the functional’s domain would divide into even more cells than in case of ground state RDMFT. These consequences of crossing energy levels make the calculation of the universal functional in the following more involved. From a general perspective, this highlights that the commonly pursued strategy to write down smooth ansatzes for the universal functional is rather problematic. At the same time, it also questions the importance and meaning of universality in functional theories.

D. \( w \)-ensemble \( \gamma \)-representability problem

The original formulation of ground state RDMFT by Gilbert [61] was hampered by the so-called \( \gamma \)-representability problem which for most quantum systems is impossible to solve. A 1RDM \( \gamma \in \mathcal{P}_N \) is called \( \gamma \)-representable if there exists some one-particle Hamiltonian \( \hat{h} \) yielding \( \gamma_{\hat{h}} \) as the ground state 1RDM according to

\[
\hat{h} \mapsto \hat{H}(\hat{h}) \mapsto |\Psi_{\hat{h}}\rangle \mapsto \gamma_{\hat{h}},
\]

where \( |\Psi_{\hat{h}}\rangle \) denotes the \( N \)-particle ground state of \( \hat{H}(\hat{h}) \). The significance of this definition rests upon the following relation between the ground state energy \( E \) and the universal ground state functional \( \mathcal{F} \) for \( \gamma \)-representable 1RDMs,

\[
\mathcal{F}(\gamma_{\hat{h}}) = E(\gamma_{\hat{h}}) - \text{Tr} \left[ \gamma_{\hat{h}} \hat{h} \right].
\]

Because of its fruitful consequence (72), we now establish an extension of \( \gamma \)-representability to \( w \)-ensemble RDMFT. A 1RDM \( \gamma \in \mathcal{E}_N^w(\hat{w}) \) shall be called \( w \)-ensemble \( \gamma \)-representable if \( \gamma \) emerges as the 1RDM of the minimizer in the GOK variational principle (6) applied to the Hamiltonian \( \hat{H}(\hat{h}) \) for some \( \hat{h} \). Note that in Sec. III this \( w \)-ensemble \( \gamma \)-representability problem was circumvented by the constrained search formalism (8) from the very beginning. Yet, if there was given a compact solution of the \( w \)-ensemble \( \gamma \)-representability problem, the universal functional could be determined more directly. In analogy to (72), for any \( w \)-ensemble
in Sec. VII E 2, we illustrate the derivation of \( \mathcal{F}_w \) for all \( w \)-ensemble \( \psi \)-representable 1RDMs for a homogeneous BEC.

The two \( \psi \)-ensemble functionals \( \mathcal{F}_w \) and \( \mathcal{F}_w^\ast \) defined in Eqs. (9) and (10) are equal for a given 1RDM \( \psi \) whenever \( \psi \) is \( \psi \)-ensemble \( \psi \)-representable. In case \( \mathcal{F}_w \) is convex, every \( \psi \in \mathcal{E}_\lambda^1(w) \) is \( \psi \)-ensemble \( \psi \)-representable, a statement that is well known in the context of ground state functional theory (see, e.g., Ref. [20]). Since the Legendre-Fenchel transform of \( \mathcal{F}_w \) is the energy \( E_w \) up to minus signs, this implies that for a convex functional \( \mathcal{F}_w \) the biconjugate \( \mathcal{F}_w^\ast \) of \( \mathcal{F}_w \) is equal to the functional \( \mathcal{F}_w \) itself. Moreover, as an alternative to the constrained search formalism [11], we can then derive not only \( \mathcal{F}_w \), but also \( \mathcal{F}_w \) through a Legendre-Fenchel transformation. Conversely, if \( \mathcal{F}_w \) turns out to be non-convex, the set \( \mathcal{E}_\lambda^1(w) \) has to contain 1RDMs \( \psi \) which are \( \psi \)-ensemble \( \psi \)-representable. In that case, calculating the biconjugate \( \mathcal{F}_w^{\ast\ast} \) would just yield the lower convex envelope of \( \mathcal{F}_w \).

We will exploit the Legendre-Fenchel transformation in Sec. VII E 1 to derive the \( \psi \)-ensemble functional \( \mathcal{F}_w \) for a homogeneous BEC.

### E. Derivation of \( \psi \)-ensemble functional for \( r = 2 \)

In order to apply the \( \psi \)-ensemble RDMFT for bosons to a homogeneous BEC, we restrict in the following to finite but large enough systems such that the \( p = 0 \) momentum state is macroscopically occupied and there exists a finite gap between the energy levels. Due to \( W_p = W_{-p} \) and \( t_p = t_{-p} \), the excited energy states are degenerate. In the following, we restrict to \( r = 2 \), such that the corresponding \( \psi \)-ensemble functional \( \mathcal{F}_w \) in [3] allows one to determine the ground state and the first excited state. Thus, we consider weight vectors of the form

\[
\nu = (w, 1 - w, 0, \ldots)
\]

with \( w \geq \frac{1}{2} \). According to Eq. (68), for each \( p' \) the weighted sum of the ground state energy \( E_0 \) and a single excitation with momentum \( p' \) reads

\[
E_{w, p'} = w E_0 + (1 - w) E_1 = E_0 + (1 - w) \omega p'.
\]

This implies that the sought-after energy \( E_w \) follows as

\[
E_w = \min_{p'} E_{w, p'}.
\]

Furthermore, we recall from Sec. VII D that the domain of the relaxed \( \psi \)-ensemble functional \( \mathcal{F}_w \) is given by the spectral polytope

\[
\Sigma(w) = \text{conv} \left\{ \{ \nu(v) \mid \nu \in \mathcal{S}^1 \} \right\},
\]

where \( \nu \) is the natural occupation number vector (see Eq. (40))

\[
\nu = (N - 1 + w, 1 - w, 0, \ldots)
\]

In the following, we present three different approaches for deriving the universal \( \psi \)-ensemble functional for \( r = 2 \) in the context of Bogoliubov theory, i.e., in the regime of small quantum depletion. This also allows us to illustrate various aspects of \( \psi \)-ensemble RDMFT discussed in the previous sections.

#### 1. Legendre-Fenchel transformation

In the following, we derive the relaxed \( \psi \)-ensemble functional \( \mathcal{F}_w \) for \( r = 2 \) through a Legendre-Fenchel transformation of the energy in Eq. (75). As introduced in Ref. [20] for the ground state functional and anticipated in Sec. VII D, the energy \( \mathcal{E}_w \) and the \( \psi \)-ensemble functional \( \mathcal{F}_w \) are related through the Legendre-Fenchel transform by

\[
\mathcal{F}_w(\hat{h}) = \max_{\psi \in \mathcal{E}_\lambda^1(w)} \left[ \text{Tr}_1(\hat{h} \hat{\gamma}) - \mathcal{F}_w(\hat{\gamma}) \right] = -E_w(\hat{h}) \cdot \hat{h}.
\]

Consequently, the biconjugate \( \mathcal{F}_w^{\ast\ast} \) of \( \mathcal{F}_w \) can be expressed as

\[
\mathcal{F}_w^{\ast\ast}(\hat{\gamma}) = \max_{\hat{h}} \left[ E_w(\hat{h}) - \text{Tr}_1(\hat{h} \hat{\gamma}) \right].
\]

Since the energy \( E_w \) is related to the energies \( E_{w, p'} \) (see Eq. (76)), auxiliary functionals \( \mathcal{F}_{w, p'} \) are introduced as the Legendre-Fenchel transforms (up to the common minus signs) of the (concave) \( \mathcal{F}_{w, p'} \) for all \( p' \). They allow us to rewrite the energy \( E_w \) using Eq. (76) as

\[
E_w = \min_{p'} \min_{\hat{\gamma} \in \mathcal{E}_\lambda^1(w)} \left[ \text{Tr}_1(\hat{h} \hat{\gamma}) + \mathcal{F}_{w, p'}(\hat{\gamma}) \right]
\]

\[
= \min_{\hat{\gamma} \in \mathcal{E}_\lambda^1(w)} \left[ \text{Tr}_1(\hat{h} \hat{\gamma}) + \min_{p'} \mathcal{F}_{w, p'}(\hat{\gamma}) \right].
\]

From the above equation it immediately follows that

\[
\mathcal{F}_w(\hat{\gamma}) = \min_{p'} \mathcal{F}_{w, p'}(\hat{\gamma}).
\]

This reflects very well the curse of universality outlined in Sec. VII C since no closed analytical form exists for \( \mathcal{F}_w \).

To proceed, we first recall that we restrict to \( \hat{h} \equiv \hat{t} \) with \( t_p = t_{-p} \) which implies that the inner product \( \langle \gamma, \tilde{t} \rangle \) for a fixed \( \tilde{t} \) is completely determined through the vector \( \eta \) defined in Eq. (60). Then, the maximum in Eq. (80) is obtained by solving for all \( p' \)

\[
\eta_{p'} \equiv n_{\tilde{p}} + n_{-\tilde{p}} = \frac{\partial E_{w, p'}(\tilde{t})}{\partial t_{p'}}.
\]
Its solution $t_{p'}(\eta_{p'})$ corresponding to a maximum and reads

$$
t_{p'}(\eta_{p'}) = \begin{cases} 
nW_{p'} \left( \frac{1+\eta_{p'}}{\sqrt{\eta_{p'}(\eta_{p'}+2)}} - 1 \right) & \text{if } \tilde{p}' \neq p', \\
nW_{p'} \left( \frac{1+\eta_{p'}}{\sqrt{(\eta_{p'}+1)(\eta_{p'}+1)}} - 1 \right) & \text{if } \tilde{p}' = p'. 
\end{cases} \quad \text{(85)}
$$

Combining (80) and (85) eventually leads to

$$
\mathcal{F}_{w,p'}(\eta) = \mathcal{F}_{w,0}(\eta) + nW_{p'} \left( \sqrt{\eta_{p'}}(\eta_{p'}+2) - \sqrt{\eta_{p'}+3-w}(\eta_{p'}+1-w) \right). \quad \text{(86)}
$$

It worth recalling here that for a given $p'$ the functional $\mathcal{F}_{w,p'}(\eta)$ equals the universal functional $\mathcal{F}_{w}(\eta)$ only for those $\eta$ whose minimizers in (16) involve as first excited state the respective $p'$-excitation. In general, the functional $\mathcal{F}_{w}$ then follows from Eq. (83).

We close this section by observing the following intriguing relation. $\mathcal{F}_{w}$ (through $\mathcal{F}_{w,p'}$) consists of the convex ground state functional $\mathcal{F}_{w,0}$ given by Eq. (66) plus an additional positive term which always increases the energy due to a single elementary excitation $p' = \hat{p}'(\eta)$ of the ground state. Moreover, one can easily check that $\mathcal{F}_{w,p'}(\eta)$ in Eq. (80) and thus $\mathcal{F}_{w}$ in Eq. (83) reduces to $\mathcal{F}_{w,0}$ for $w = 1$, as required.

2. Derivation of $\mathcal{F}_{w}$ and $\mathcal{F}_{w}$ for all $w$-ensemble $v$-representable 1RDMs

Once the energy $E_{w}$ is known, we can derive for all $w$-ensemble $v$-representable 1RDMs $\hat{\gamma}$ the value of the $w$-ensemble functionals $\mathcal{F}_{w}(\hat{\gamma}) = \mathcal{F}_{w}(\hat{\gamma})$ through Eq. (73).

In order to apply this approach to the Bogoliubov approximated interaction $\hat{W}_{B}$, we first define the momentum $q'$ corresponding to the first excitation, which is determined through the lowest value of the quasiparticle dispersion $\omega_{p} = \omega_{-p}$. Moreover, the degenerate subspace of the first excited state $|\Psi_{1}\rangle$ is spanned the two orthonormal states $\hat{c}_{q}|\Psi_{0}\rangle$ and $\hat{c}_{-q}|\Psi_{0}\rangle$. Thus, any superposition state

$$
|\Psi_{1}\rangle = \alpha \hat{c}_{q}^{\dagger}|\Psi_{0}\rangle + \beta \hat{c}_{-q}^{\dagger}|\Psi_{0}\rangle \quad \text{(87)}
$$

with $\alpha, \beta \in \mathbb{C}$ and normalization $|\alpha|^{2} + |\beta|^{2} = 1$ corresponds to a single excitation on top of the interacting ground state $|\Psi_{0}\rangle$. Therefore, within the Bogoliubov approximation, we restrict the set $\mathcal{E}^{N}$ of all $N$-boson density operators $\hat{\Gamma}$ to the subset of all variational states of the form

$$
\hat{\Gamma}_{w} = w|\Psi_{0}\rangle\langle\Psi_{0}| + (1-w)|\Psi_{1}\rangle\langle\Psi_{1}| \quad \text{(88)}
$$

with $|\Psi_{1}\rangle$ given by Eq. (87). This variational ansatz reduces the minimization on the right hand side of the GOK variational principle (6) applied to the Bogoliubov Hamiltonian $\hat{H}_{B}$ to a minimization of the energy

$$
\mathrm{Tr}_{N}[\hat{H}_{B}\hat{\Gamma}_{w}] = \sum_{p'} 2 \left( (nW_{p'} + t_{p'}) \frac{\phi_{p'}^{2}}{1 - \phi_{p'}^{2}} - nW_{p'} \phi_{p'}^{2} \right) + (1-w) \left( nW_{q'} + t_{q'} \frac{1+\phi_{q'}^{2}}{1 - \phi_{q'}^{2}} - nW_{q'} \frac{2\phi_{q'}^{2}}{1 - \phi_{q'}^{2}} \right) \quad \text{(89)}
$$

over the variational parameters $\{\phi_{p'}, \eta_{p'}\}$ defined in Sec. VII A. Performing the minimization in (89) for all momenta $p'$ separately leads to the solution

$$
\tilde{\phi}_{p'} = \frac{1}{nW_{p'}} \left( t_{p'} + nW_{p'} - \sqrt{t_{p'}^{2} + 2nW_{p'}} \right), \quad \text{(90)}
$$

in agreement with Ref. [58] [59]. As a consistency check one can show that Eqs. (89) and (90) indeed lead to $E_{w}$ in (73). Furthermore, the expectation value of the operator $\eta_{p'}$ (60) is given by

$$
\eta_{p'} = \mathrm{Tr}_{N}[\hat{\eta}_{p'}\hat{\Gamma}_{w}] = \begin{cases} 
\frac{2\phi_{p'}^{2}}{1 - \phi_{p'}^{2}} & \text{if } \tilde{p}' \neq q', \\
2\phi_{p'}^{2} + (1-w) \frac{1+3\phi_{q'}^{2}}{1 - \phi_{q'}^{2}} & \text{if } \tilde{p}' = q'.
\end{cases} \quad \text{(91)}
$$

Inserting the minimizers $\tilde{\phi}_{p'}$ (90) into (91) leads for all momenta $p'$ to the same solution for the dispersion $t_{p'}$ as in (85). This allows us to use (73),

$$
\mathcal{F}_{w,q}(\eta) = E_{w,q}(t) - \sum_{p'} t_{p'} \eta_{p'} \quad \text{(92)}
$$

to calculate the $w$-ensemble functional $\mathcal{F}_{w,q}$ for all $w$-ensemble $v$-representable $\eta$. Evaluating (92) eventually leads to the same expression for $\mathcal{F}_{w,q}$ as the biconjugate $\mathcal{F}_{w}^{*} = \mathcal{F}_{w}^{*}(\eta)$ derived through the Legendre-Fenchel transformation in Sec. VII E 1. In particular, this confirms for all $v$-representable $\eta$

$$
\mathcal{F}_{w}(\eta) = \mathcal{F}_{w}(\eta). \quad \text{(93)}
$$
Thus, for the Bogoliubov Hamiltonian \[ \mathcal{H}_\text{B} \] the functional \( \mathcal{F}_w \) is convex and therefore every occupation number vector \( \eta \) which is \( w \)-ensemble \( N \)-representable is also \( w \)-ensemble \( v \)-representable.

3. Constrained search formalism

In contrast to the Bogoliubov approximated interaction \( \hat{W}_{\text{B}} \), the domain \( \mathcal{E}_N^1(w) \) of \( \mathcal{F}_w \) for a generic interaction \( \hat{W} \) typically contains 1RDMs \( \hat{\gamma} \) which are not \( w \)-ensemble \( v \)-representable. Due to the complexity of the \( v \)-representability problem it would then not be possible anymore to determine a universal functional through Eq. (92). More promisingly, the constrained search formalism \[8\] defines \( \mathcal{F}_w \) for all \( \hat{\gamma} \in \mathcal{E}_N^1(w) \), or equivalently \( \mathcal{F}_w \) for all \( \gamma \in \mathcal{E}_N^1(w) \). Therefore, the approach to derive \( \mathcal{F}_w \) via \( w \)-ensemble \( v \)-representability in Sec. VII E 2 and the constrained search formalism discussed below are in general quite different from a conceptual point of view. However, the restriction in the constrained search \[8\] to the variational states \[88\] and the convexity of \( \mathcal{F}_w \) reduces the differences between the approaches to derive \( \mathcal{F}_w \) in this section and Sec. VII E 2 as we will observe in the following.

To illustrate the constrained search \[8\] for a homogeneous BEC, we first need to calculate the expectation value \( \text{Tr}_N [ \hat{W}_B \hat{\Gamma}_w ] = \text{Tr}_N [ \hat{H}_B \hat{\Gamma}_w ] - \text{Tr}_N [ \hat{\Gamma}_w ] \). Since \( \text{Tr}_N [ \hat{H}_B \hat{\Gamma}_w ] \) is given by Eq. (89) we immediately arrive at

\[ \text{Tr}_N [ \hat{W}_B \hat{\Gamma}_w ] = \sum_{p'} 2 n W_{p'} \left( \frac{\phi^2_{p'}}{1 - \phi^2_{p'}} - \frac{\phi_{p'}}{1 - \phi^2_{p'}} \right) \]

\[ + (1 - w) n W_q \left( 1 + \frac{\phi^2_q}{1 - \phi^2_q} - 2 \phi_q \right). \]

Furthermore, the occupation numbers \( \eta_{p'} \) in Eq. (91) determine the variational parameters \( \phi_{p'} \),

\[ \phi_{p'} = \begin{cases} \sigma_{p'} \sqrt{\eta_{p'}} - \frac{\eta_{p'}}{2 + \eta_{p'}} & \text{if } p' \neq q', \\ \sigma_{p'} \sqrt{\eta_{p'}} + w - 1 & \text{if } p' = q', \end{cases} \]

up to a phase \( \sigma_{p'} = \pm 1 \). As a result, the constrained search formalism \[8\] simplifies to a minimization over the phases \( \sigma_{p'} \) of \( \phi_{p'} \).

\( F_{w,q} (\eta) = \min_{\{\sigma_p = \pm 1\}} \left[ \sum_{p' \neq q'} n W_{p'} \left( \eta_{p'} - \sigma_{p'} \sqrt{\eta_{p'} (\eta_{p'} + 2)} \right) + n W_{q'} \left( \eta_{q'} - \sigma_{q'} \sqrt{\eta_{q'} + 3 - w} \right) \right] \),

which can be solved independent of the sign of the Fourier coefficients \( W_{p'} \). Here, we have \( W_{p'} \geq 0 \) for all momenta \( p' \) such that the minimization in (96) leads indeed to the functional presented in Eq. (86) and (83), respectively.

F. Bose-Einstein condensation force

For ground state RDMFT a remarkable property has recently been discovered: the gradient of the universal functional diverges repulsively on the boundary of the allowed regime. This BEC force for bosons \[15\] \[16\] \[56\] and exchange force for fermions \[21\] is a consequence of the geometry of quantum states and thus independent of the microscopic properties of the system. In the following, based on the result \[86\], \[83\] we confirm the existence of this BEC force also in the context of excited state RDMFT. This demonstrates that the boundary of the functional’s domain \( \mathcal{E}_N^1(w) \) and effectively \( \Sigma^1(w) \) contains crucial information about the excitation structure of \( N \)-boson quantum systems.

In the following we therefore consider the boundary of \( \Sigma^1(w) \), with a particular emphasis on the neighborhood of the generating vertex \( v \) \[78\]. From Eq. (86) we obtain for the derivative of \( \mathcal{F}_w \) with respect to the occupation numbers \( \eta_{p'} \) close to the vertex \( v = (N + 1, 1, 0, \ldots) \),

\[ \frac{\partial \mathcal{F}_w}{\partial \eta_{p'}} (\eta) \propto \begin{cases} -\frac{1}{\sqrt{\eta_{p'}}} & \text{if } p' \neq q', \\ -\frac{1}{\sqrt{\eta_{p'} + w - 1}} & \text{if } p' = q'. \end{cases} \]

The above equation already reveals that the gradient of \( \mathcal{F}_w \) diverges repulsively whenever one of the occupation numbers \( \eta_{p'} \) tends to zero. Consequently, whenever \( \eta \) approaches \( v \) or any other point on the boundary, the corresponding gradient force is collectively diverging. This BEC force namely contains individual contributions from various polytope facets that are reached. In this context, the zero momentum state requires a separate treatment since we assumed that this state would be macroscopically occupied. To be more specific, our derivation of \( \mathcal{F}_w \) assumed \( n_0 \approx N \) and we used the normalization to substitute \( n_0 \). Hence, the information about \( n_0 \) close to the boundary of \( \Sigma^1(w) \) is hidden in all the other occupation numbers. Let us now assume that the upper bound on \( n_0 \) is saturated, i.e., \( n_0 \rightarrow N - 1 + w \). Then, for a large dimension \( d \) of the one-particle Hilbert space, i.e. a large number of different \( p' \), all \( N - n_0 \) bosons not occupying the \( p = 0 \) state can distribute over the remaining orbitals. As a result, either some of the occupation num-
bers $\eta p$ are equal to zero or they are all very close to zero. According to Eq. (97) this leads again to a collective repulsive force at the boundary of the domain of $F_w$. To elaborate a bit more on these findings, we investigate in the following the divergence of the gradient of $F_w$ as a function of the distance

$$ D = \frac{1}{N} \sum_{p} \eta p - D_0 $$

of a momentum occupation number vector $\eta$ to the vertex $v$ of $\Sigma^i(w)$. Here, $D_0 = (1 - w)/N$ denotes the fraction of non-condensed bosons at the vertex $v$. For this purpose, let us consider a straight path from a starting point $v$ towards $v$ according to

$$ \eta(t) = \eta + t(v - \eta) $$

with $t \in [0,1]$. In particular, we obtain for the distance $D$ along this straight path $D(t) = (1 - t)D(0) \equiv (1 - t)D$, where $D$ denotes the fraction of non-condensed bosons at the occupation number vector $\eta$. Taking the derivative of $F_w$ with respect to the distance $D$ defined in Eq. (98) eventually yields after an elementary calculation

$$ \frac{dF_w}{dD}(\eta) \propto -\frac{1}{\sqrt{D}}. $$

Thus, $dF_w/dD$ diverges as $1/\sqrt{D}$ in the limit $D \to 0$ in analogy to the fermionic exchange force [21] and the BEC force for ground states [15, 16]. Moreover, the corresponding prefactor is always negative and contains all information about the system’s specific properties of the interaction.

We thus succeeded in generalizing the concept of a BEC force also to excitations in homogeneous BECs. Motivated by the significance of the energy gap between the ground state and the first excited state for most physical systems, we discussed above the case $r = 2$. Of course, the derivation of the $w$-ensemble universal functional $F_w$ and its gradient can be extended in a similar fashion to larger values of $r$ in order to provide access to a larger number of excitation energies. Yet, this would require more mathematical effort which is also due to the degeneracy of the excited states.

Each $\eta p$ is a function of the distance $\hat{r}$ from the vertex $v$ to the vertex $v$.

## VIII. DFT FOR EXCITATIONS IN LATTICE BOSON SYSTEMS

In this section we explain how and why the bosonic exclusion principle constraints discussed in Sec. [11] apply also to bosonic lattice DFT for excitations (GOK-DFT). Let us start by recalling from a general perspective the relation and main difference between RDMFT and DFT: Compared to RDMFT, DFT restricts the class of Hamiltonians of interest further to $\hat{H}(\hat{r}) \equiv \hat{r} + \hat{t} + \hat{W}$ by fixing also the kinetic energy, i.e., only the external potential $\hat{t}$ is variable. Thus, the resulting universal functional $\tilde{G}_w$ in DFT depends on both the fixed kinetic energy $\hat{t}$ and fixed interaction $\hat{W}$ and it is universal in the sense that it is independent of $\hat{v}$. For discrete lattice systems, $\tilde{G}_w$ is a functional of the vector $n \equiv (n_i)$ of the lattice site occupancies $n_i$, $\tilde{G}_w \equiv \tilde{G}_w(n)$. Then, the domain of $\tilde{G}_w(n)$ is the set of all vectors $n$ which follow from a 1RDM $\hat{\gamma}$ which is (relaxed) $w$-ensemble $N$-representable. Since the elements of the vector $n$ are nothing else than the diagonal elements of the 1RDM $\hat{\gamma}$ in the lattice site basis $\{|i\rangle\}$, all concepts presented in Sec. [11] can easily be translated to define the universal $w$-functional $\tilde{G}_w(n)$ and determine its domain. According to a fundamental theorem by Schur and Horn [51, 52], the vector of diagonal elements of a matrix is majorized by the vector of its eigenvalues. Hence, the occupation number vector $n$ is majorized by the natural occupation number vector $\lambda$, i.e.,

$$ n \equiv (\langle i|\hat{\gamma}|i\rangle) \prec \lambda \equiv \text{spec}(\hat{\gamma}). $$

Then, using a generalization of Rado’s theorem [39] (see also Eq. (47)) it follows immediately from the transitivity of the majorization that $n$ must obey the same non-trivial constraints as $\lambda$. In summary, our work in particular provides the first constructive derivation of the weight dependence of the functional’s domain in $w$-ensemble lattice DFT. Despite the combinatorial differences in the derivation of the bosonic exclusion principle constraints, this result on lattice DFT is not specific to bosons and also applies to the fermionic case as already emphasized in Ref. [39].

## IX. SUMMARY AND CONCLUSIONS

We proposed and worked out a novel method for calculating excitation energies in quantum systems of correlated bosons. Motivated by the Penrose-Onsager criterion for BEC, the exponentially complex many-boson wave function was substituted by the simpler one-particle reduced density matrix (1RDM) $\hat{\gamma}$: By employing a variational principle for $N$-boson density matrices with spectrum $w$ [40, 41] within the Levy-Lieb constrained search formalism [43, 44] we established a universal functional $F_w(\hat{\gamma})$. This functional theory provides access to the energies of low-lying states and their differences — including the important ground state gap — through the variation of the weight distribution $w$ of the excited states. Accordingly, $w$-ensemble RDMFT constitutes a potentially ideal framework for describing quantum phase transitions. In particular, it will be an instructive challenge to explore the nonanalytical structural features that the universal functional needs to possess in order to correctly describe quantum phase transitions. Yet, to first establish $w$-RDMFT as a practically useful method we had to master two critical challenges related to the universal functional: How can one construct meaningful approximations and how does even its domain look like?
Determining the functional’s domain would mean to solve efficiently the corresponding one-body $w$-ensemble $N$-representability problem. This is, however, impossible for realistic systems sizes \cite{Hartree1928,Poplawski2011,Sei2014}. It is therefore one of the main achievements of our work (see Secs. III \textbf{V}) to circumvent the computational complexity of the respective $N$-representability problem. Inspired by the seminal work \cite{Valone2011} by Valone on fermionic ground state RDMFT and in analogy to fermionic $w$-RDMFT \cite{Sei2014,Nic2017}, we achieved this by applying an exact convex relaxation to $w$-RDMFT. Then, in a second step we resorted to concepts from convex analysis to derive a systematic and efficient characterization of the functional’s domain $\mathcal{E}_N^1(w)$. Eventually, this revealed a hierarchy of bosonic exclusion principle constraints parameterized by the number $r$ of finite weights $w_j$. In analogy to Pauli’s famous exclusion principle for fermionic pure states, these new constraints are effectively independent of the total particle number $N$ and the dimension $d$ of the one-particle Hilbert space. Our comprehensive derivation also demonstrates that the boundary of the prescribed set of admissible 1RDMs contains the entire information about the excitation spectrum of non-interacting bosons. It follows from perturbation theoretical arguments that this statement remains approximately true for weakly interacting systems. Moreover, the bosonic exclusion principle constraints may have some broad physical relevance beyond functional theory. For instance, they could be used to dissect the statistical uncertainty of bosonic occupation numbers according to its origin to entropic thermal contributions and the interaction between the particles. An alternative prospective application is to monitor the time-evolution of open-quantum systems with an emphasis on the effect of thermalization.

To initiate $w$-RDMFT, we derived analytically the functionals for the symmetric Bose-Hubbard dimer (Sec. \textbf{V}) and the homogeneous Bose gas within the Bogoliubov approximation (Sec. \textbf{V}) for $r = 2$ finite weights $w_j$. Indeed, these two systems provide ideal starting points for the future development of more sophisticated functional approximations: The Bose-Hubbard dimer constitutes the building block of the Bose-Hubbard model, one of the most important models in condensed matter physics. In turn, the Bogoliubov functional represents the bosonic analogue of the pivotal Hartree-Fock functional for fermionic systems \cite{Sei2014} since it refers to the regime of small quantum depletion. Last but not least, these two systems highlight that the boundary $\partial\mathcal{E}_N^1(w)$ of the functional’s domain $\mathcal{E}_N^1(w)$ has a particular relevance. To be more specific, the gradient of each functional was found to diverge repulsively as the 1RDM approaches $\partial\mathcal{E}_N^1(w)$. This remarkable result generalizes the recently discovered exchange force \cite{Bra2015} for fermions and BEC force for bosons \cite{Lan2015,Sei2016,Sei2017} in their ground states to mixed states. The existence of those forces does not depend on any microscopic details but has a solely geometrical origin. In that sense, these novel concepts emphasize the prominent role that the geometry of reduced quantum states can play in general in advancing functional theories.

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**Appendix A: Relating spectral polytope for different particle numbers**

In this section, we prove that every natural occupation number vector $\mu \in \Sigma_N(w)$ corresponding to a system with $N'$ bosons can be related to a $\lambda \in \Sigma_N(w)$ with $N' > N \geq r - 1$ in a unique way. A main tool to establish the sought-after relation between the spectral polytopes for $N$ and $N' > N$ is the following generalization of Radó’s theorem \cite{Rado1925} stating that for finitely many vectors $v^{(1)}, \ldots, v^{(R)} \in \mathbb{R}^d$, the permutation-invariant polytope

$$\mathcal{P} = \text{conv}\left(\left\{\pi(v^{(j)}) \mid j = 1, \ldots, R, \pi \in \mathcal{S}^d\right\}\right) \quad (A1)$$

is equivalent to

$$\mathcal{P} = \left\{\lambda \mid \exists \text{ conv. comb.} \sum_{j=1}^R p_j v^{(j)} = v : \lambda \prec v\right\}.$$ \quad (A2)

Also recall that the vertices $v^{(j)}$ of the two spectral polytopes $\Sigma_N(w)$ and $\Sigma_{N'}(w)$ are related through

$$v^{(j)}_{N'} = v^{(j)}_N + \delta e_1,$$ \quad (A3)

where $\delta = N' - N$ and $e_1 = (1, 0, 0, \ldots, 0)$ is a unit vector.

In the following, we consider an arbitrary but fixed number $R$ of vertices $v^{(j)}$. According to two main theorems in convex analysis by Hardy, Littlewood, and Polya \cite{Hardy1952} and Birkhoff, and von Neumann \cite{Neumann1936,Neumann1946}, every $\mu \in \Sigma_N(w)$ can be written as

$$\mu = \sum_{i=1}^R \sum_{\pi \in \mathcal{S}^d} q_i, \pi(v^{(i)}_N),$$ \quad (A4)

where $\sum_{i=1}^R \sum_{\pi \in \mathcal{S}^d} q_i, \pi = 1$ and $\mathcal{S}^d$ denotes the set of all permutations $\pi$ of $d$ elements. Combining this with \textbf{A2}...
and (A3) leads to
\[ \mu = \sum_{i=1}^{R} \sum_{\pi \in S^d} q_{i,\pi} (\nu_{N}^{(i)}) \]
\[ = \sum_{i=1}^{R} \sum_{\pi \in S^d} q_{i,\pi} (\nu_{N}^{(i)} + \delta e_1) \]
\[ = \sum_{i=1}^{R} \sum_{\pi \in S^d} q_{i,\pi} \left( \pi(\nu_{N}^{(i)}) + \pi(\delta e_1) \right) \]
\[ = \lambda + \delta \sum_{i=1}^{R} \sum_{\pi \in S^d} q_{i,\pi} (e_1) , \quad \text{(A5)} \]

where \( \lambda \in \Sigma_N(w) \). The argument in (A3) can be simply reverted to derive for any \( \lambda \in \Sigma_N(w) \) a unique corresponding \( \mu \in \Sigma_N(w) \). It follows that
\[ \mu \in \Sigma_{N'}(w) = \Sigma_N(w) + C , \]
\[ \{ \lambda + c \mid \lambda \in \Sigma_N(w) , c \in C \} \quad \text{(A6)} \]
where the set
\[ C \equiv \text{conv}(\{ \pi(\delta e_1) \mid \pi \in S^d \}) \quad \text{(A7)} \]
is a rescaled simplex with edge length \( \delta = N' - N \). Thus, according to Eq. (A6), \( \Sigma_{N'}(w) \) is given by the Minkowski sum of \( \Sigma_N(w) \) and \( C \).

Appendix B: Derivation of the \( w \)-ensemble functional in the symmetric Bose-Hubbard dimer

In this section, we derive the \( w \)-ensemble functional for the symmetric Bose-Hubbard dimer in the total momentum sector with \( \hat{P} = 0 \). The allowed values for the discrete momentum \( p \) are given by \( p_{\nu} = \pi \nu \) with \( \nu = 0, 1 \). We denote the operator creating a boson with momentum \( p_{\nu} \) by \( a_{p_{\nu}}^\dagger \) (see also Sec. [V]). The two-dimensional subspace \( \mathcal{H}_{1/2} = 0 \) of the two boson Hilbert space \( \mathcal{H}_2 \) is spanned by the basis states
\[ |1\rangle = \frac{1}{\sqrt{2}} (a_{0}^\dagger)^2 |0\rangle , \quad \text{(B1)} \]
\[ |2\rangle = \frac{1}{\sqrt{2}} (a_{1}^\dagger)^2 |0\rangle , \quad \text{(B2)} \]
where \( |0\rangle \) denotes the vacuum state. In this basis, every two-boson density operator \( \hat{\Gamma} \) with spectrum \( w = (w_1, 1 \rangle \) can be expressed as
\[ \hat{\Gamma}_w = \sum_{i,j=1}^{2} \Gamma_{ij}^{(w)} |i\rangle \langle j| . \quad \text{(B3)} \]

However, the minimizer states in the GOK variational principle (6) take the form \( \hat{\Gamma}_w = \sum_{j=1}^{2} w_j |\Psi_j\rangle \langle \Psi_j| \), where \( |\Psi_j\rangle \) are the eigenstates of the Hamiltonian \( \hat{H} = \hat{h} + \hat{W} \). Here, \( \hat{W} \) denotes the Hubbard on-site interaction term in Eq. (51). To determine \( \Gamma^{(w)} \) in terms of the weights \( w_j \), we expand the eigenstates \( |\Psi_j\rangle \) as follows,
\[ |w_1\rangle = \alpha_1 |1\rangle + \alpha_2 |2\rangle , \quad |\Psi_2\rangle = \beta_1 |1\rangle + \beta_2 |2\rangle \quad \text{(B4)} \]
with \( \alpha_i, \beta_i \in \mathbb{R} \). These expansion coefficients \( \alpha_i, \beta_i \) must further satisfy the orthonormality conditions
\[ \alpha_1^2 + \alpha_2^2 = 1 , \quad \beta_1^2 + \beta_2^2 = 1 , \quad \alpha_2 \beta_2 = 0 . \quad \text{(B5)-(B7)} \]
Combining Eq. (B3) with Eq. (B4) leads to
\[ \Gamma_{11}^{(w)} = 1 - \Gamma_{22}^{(w)} = w_1 \alpha_1^2 + w_2 \beta_1^2 \]
\[ \Gamma_{21}^{(w)} = \Gamma_{12}^{(w)} = w_1 \alpha_1 \alpha_2 + w_2 \beta_1 \beta_2 . \quad \text{(B8)} \]

To express the 1RDM \( \hat{\gamma} \) in terms of the matrix elements \( \Gamma_{ij}^{(w)} \), in the next step, we first recall that \( \hat{\gamma} \) is diagonal in momentum representation. Thus, in our case \( \hat{\gamma} \) depends only on a single independent parameter due to the normalisation \( n_0 + n_2 = 2 \), where \( n_\nu = \text{Tr}_2 [a_{\nu}^\dagger a_\nu \hat{\Gamma}_w] \). Together with Eq. (B3) we arrive at
\[ n_0 = \text{Tr}_2 [a_{0}^\dagger a_0 \hat{\Gamma}_w] = 2 \Gamma_{11}^{(w)} = 2 (w_1 \alpha_1^2 + w_2 \beta_1^2) . \quad \text{(B9)} \]

Moreover, to determine the \( w \)-ensemble functional \( F_w(\gamma) \), we need to minimize
\[ \text{Tr}_2 [\hat{W} \hat{\Gamma}_w] = U \left( 1 + 2 \Gamma_{12}^{(w)} \right) \]
\[ = U (1 + 2 (w_1 \alpha_1 \alpha_2 + w_2 \beta_1 \beta_2)) \quad \text{(B10)} \]
according to the constrained search formalism with respect to the coefficients \( \alpha_i, \beta_i \). Since their three orthonormality conditions together with Eq. (B9) constitute four conditions for four free variables, this minimization can be carried out analytically without much effort. Solving the resulting system of equations (B3), (B5), (B6) and (B7) and (B9) leads in a straightforward manner to
\[ \text{Tr}_2 [\hat{W} \hat{\Gamma}_w] = U \left( 1 \pm \sqrt{(n_0 - 2w_2) (2w_1 - n_0)} \right) . \quad \text{(B11)} \]

Choosing the minus sign which minimizes the expectation value in Eq. (B10) eventually yields
\[ F_w(n_0) = U \left( 1 - \sqrt{(n_0 - 2w_2) (2w_1 - n_0)} \right) \]
\[ = U (1 - \sqrt{n_0 (2 - n_0) - 4 w_1 w_2}) , \quad \text{(B12)} \]
which is a functional of the momentum occupation number \( n_0 \) only. Since the functional \( F_w \) and its domain are both convex for every \( w \) this functional coincides with its relaxed variant \( F_w \).

Next, we minimize the energy functional \( \text{Tr}_1 [\hat{\gamma}^2] + F_w(n_0) \), where \( \hat{\gamma} = -2t \sum_{\nu = 0,1} \cos(\pi \nu) \hat{n}_\nu \), to verify that the result for \( E_w \) in (4) is in agreement with the eigenenergies of \( \hat{H} = \hat{h} + \hat{W} \) obtained from an exact diagonalization. The kinetic energy in terms of \( n_0 \) is given by (recall that \( n_1 = 2 - n_0 \))
\[ \text{Tr}_1 [\hat{\gamma}^2] = -4t (n_0 - 1) . \quad \text{(B13)} \]
Thus, to calculate $E_w$ we need to solve
\[
\frac{\partial}{\partial n_0} \left( -4t(n_0 - 1) + F_w(n_0) \right) \bigg|_{n_0 = \tilde{n}_0} = 0
\]  
for the momentum occupation number $\tilde{n}_0$. This leads to
\[
\tilde{n}_0 = 1 + \frac{8(t_1 - t_2)}{\sqrt{64t^2 + 4U^2}}.
\]
Then, the energy $E_w$ follows as
\[
E_w = -4t(\tilde{n}_0 - 1) + U \left( 1 - \sqrt{\tilde{n}_0(2 - \tilde{n}_0)} - 4t_1t_2 \right)
\equiv w_1E_1 + w_2E_2,
\]
where
\[
E_1 = U - \sqrt{16t^2 + U^2}, \quad (B17)
\]
\[
E_2 = U + \sqrt{16t^2 + U^2}, \quad (B18)
\]
The Hamiltonian $\hat{H}$ in Eq. (51) in the basis spanned by the states $|1\rangle$ and $|2\rangle$ defined in Eqs. (B1) and (B2), can be represented by the matrix
\[
H = \begin{pmatrix} U - 4t & U \\ U & U + 4t \end{pmatrix}, \quad (B19)
\]
which has the two eigenvalues $E_1$ and $E_2$ introduced in Eq. (B17) and (B18). Hence, the result for the energy $E_w$ in Eq. (B16) is in agreement with the eigenenergies obtained from diagonalizing the matrix $H$. 

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