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RESEARCH ARTICLE

Analysis of density matrix embedding theory around the non-interacting limit

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Abstract

This article provides the first mathematical analysis of the Density Matrix Embedding Theory (DMET) method. We prove that, under certain assumptions, (i) the exact ground-state density matrix is a fixed-point of the DMET map for non-interacting systems, (ii) there exists a unique physical solution in the weakly-interacting regime, and (iii) DMET is exact up to first order in the coupling parameter. We provide numerical simulations to support our results and comment on the physical meaning of the assumptions under which they hold true. We show that the violation of these assumptions may yield multiple solutions to the DMET equations. We moreover introduce and discuss a specific N-representability problem inherent to DMET.

CONTENTS

1 | INTRODUCTION

Electronic structure theory is a powerful quantum mechanical framework for investigating the intricate behavior of electrons within molecules and crystals. At the core lies the interaction between particles, specifically the electron-electron and electron-nuclei interactions. Embracing the essential quantum physical effects, this theory is the foundation for ab initio quantum chemistry and materials science calculations performed by many researchers in chemistry and related fields, complementing and supplementing painstaking laboratory work. With its diverse applications in chemistry and materials science, electronic structure theory holds vast implications for the mathematical sciences. Integrating mathematical doctrines into this field leads to the development of precise and scalable numerical methods, enabling extensive in silico studies of chemistry, for example, sustainable energy, green catalysis, and nanomaterials. The synergy between mathematics and electronic structure theory offers the potential for groundbreaking advancements in addressing these global challenges.

Within the realm of electronic structure theory, the treatment of *strongly correlated quantum systems* is a particularly difficult and long-standing challenge. Here, the application of high-accuracy quantum chemical methods that are able to capture the electronic correlation effects at chemical accuracy is inevitable. Unfortunately, the application of such high-accuracy methods is commonly stymied by a steep computational scaling with respect to the system's size. A potential remedy is provided by quantum embedding theories, that is, a paradigm for bootstrapping the success of highly accurate solvers at small scales up to significantly larger scales by decomposing the original system into smaller fragments, where each fragment is then solved individually and from which, a solution to the whole system is then obtained [17, 20, 44]. Such approaches include dynamical mean-field theory [15, 16, 26, 30, 32], or variational embedding theory [7, 22, 27].

The subject of this article is a widely-used quantum embedding theory, namely, density matrix embedding theory (DMET) [3, 9, 10, 23, 24, 48, 51, 56]. The general idea of DMET is to partition the global quantum system into several quantum "impurities", each impurity being treated accurately via a high-level theory (such as full configuration interaction (FCI) [25, 39, 52], coupled cluster theory [8], density matrix renormalization group (DMRG) [55], etc.). More precisely, the DMET methodology follows the procedure sketched out as: (1) fragment the system, (2) for each fragment, construct an interacting bath that describes the coupling between the fragment and the remaining system, thus giving rise to a so-called impurity problem, (3) solve an interacting problem for each impurity using a highly accurate method, (4) extract properties of the system, (5) perform steps (2)–(4) self-consistently in order to embed updated correlation effects back into the full system. Note that in step 2, one may also consider a non-interacting bath where Coulomb interactions are included only on the fragment orbitals [56]; however, in this work, we will focus solely on the interacting bath formulation. Over the past years, a large variety of methods following this general framework has been developed, including how the bath space is defined (including the choice of low-level theory) [13, 35, 36, 59], how the interacting cluster Hamiltonian is constructed and solved [14, 29, 38, 41, 43], and the choice of self-consistency requirements [12, 57, 58]. This variety of DMET flavors has been successfully applied to a wide range of systems such as Hubbard models [3, 6, 23, 45, 46, 54, 65–67], quantum spin models [11, 18, 42], and a number of strongly correlated molecular and periodic systems [1, 2, 9, 19, 24, 31, 33, 34, 37, 40, 49, 50, 56, 60–64]. Recently, the application of DMET variants on quantum computers has been explored [5, 28, 53].

In this article, we follow the computational procedure where the global information, at the level of the one-electron reduced density matrix (1-RDM), is made consistent between all the impurities with the help of a low-level Hartree–Fock (HF) type of theory. In the self-consistent-field DMET (SCF-DMET)[1], this global information is then used to update the impurity problems in the next self-consistent iteration, until a consistency condition of the 1-RDM is satisfied between the high-level and low-level theories.

This article is organized as follows. In Section 2.1, we introduce the many-body quantum model under investigation and its fragment decomposition, and set up some notation used in the sequel. In Section 2.2, we present a mathematical formulation of the DMET impurity problem and introduce (formally) the high-level DMET map. The low-level DMET map and the DMET fixed point problem are defined (still formally) in Sections 2.3 and 2.4 respectively. In Section 3, we state our main results:

1. in Proposition 1, we show that for non-interacting systems, the exact ground-state density matrix is a fixed-point of the DMET map if (i) the system is gapped (Assumption (A1)), and (ii) the fragment decomposition satisfies a natural and rather mild condition (Assumption (A2)). Although this result is well-known in the physics and chemistry community, a complete mathematical proof was still missing;
2. in Theorem 4, we prove that under two additional assumptions ((A3) and (A4)), the DMET fixed-point problem has a unique physical solution in the weakly-interacting regime, which is real-analytic in the coupling parameter α. Assumption (A3) is related to some specific N-representability condition inherent to the DMET approach, while Assumption (A4) has a physical interpretation in terms of linear response theory;
3. in Theorem 5, we prove that in the weakly-interacting regime, DMET is exact up to first order in α.

[1] Throughout the paper, DMET refers to SCF-DMET. This is in contrast to one-shot DMET, in which the impurity problem is only solved once without self-consistent updates.

The numerical simulations reported in Section 4 illustrate the above results and indicate that DMET does not seem to be exact at second order. Although, in the special case when there is only one site per fragment, Assumption (A4) is a consequence of Assumptions (A1)–(A3) (see Remark 3), the numerical simulations presented show that this is in general not the case. Further investigations using the H_6-model (vide infra) reveal the existence of a specific configuration (Θ_3) for which only Assumption (A4) is not satisfied. In the vicinity of this configuration, DMET has at least two distinct solutions that arise from a transcritical bifurcation at Θ_3. In Section 5, we formulate the impurity problem in more detail and discuss the domain of the high-level DMET map. In Section 6, we study the N-representability problem mentioned above and provide a simple criterion of local N-representability directly connected to Assumption (A3). In order to improve the readability of the paper, we postponed the technical proofs to Section 7. For the reader's convenience, the main notations used throughout this article are collected in Table A1 in Appendix A.

2 | THE DMET FORMALISM

2.1 | The quantum many-body problem and its fragment decomposition

We consider a physical system with L quantum sites, with one orbital per site, occupied by $1 \leq N < L$ electrons, and assume that magnetic effects (interaction with an external magnetic field, spin-orbit coupling, etc.) can be neglected. This allows us to work with real-valued wave-functions and density matrices. We set

$$\mathcal{H} := \mathbb{R}^L \quad \text{(one-particle state space)}, \quad \mathcal{B}_{\rm at} := \{e_\kappa\}_{\kappa\in[\![1,L]\!]} \quad \text{(canonical basis of } \mathbb{R}^L\text{)}, \tag{1}$$

$$\mathcal{H}_n := \bigwedge^n \mathcal{H} \quad (n\text{-particle state space}), \quad \mathrm{Fock}(\mathcal{H}) := \bigoplus_{n=0}^{L} \mathcal{H}_n \quad \text{(real fermionic Fock space)}.$$

We denote by $\widehat{a}_\kappa$ and $\widehat{a}_\kappa^\dagger$ the generators of the (real) CAR algebra associated with the canonical basis of $\mathcal{H}$, that is,

$$\widehat{a}_\kappa := \widehat{a}(e_\kappa) \quad \text{and} \quad \widehat{a}_\kappa^\dagger = \widehat{a}^\dagger(e_\kappa).$$

Recall that the maps

$$\mathbb{R}^L \ni f \mapsto \widehat{a}^\dagger(f) \in \mathcal{L}(\mathrm{Fock}(\mathcal{H})) \quad \text{and} \quad \mathbb{R}^L \ni f \mapsto \widehat{a}(f) \in \mathcal{L}(\mathrm{Fock}(\mathcal{H})),$$

are both linear in this setting since we work in a real Hilbert space framework. Here and below, $\mathcal{L}(E)$ is the space of linear operators from the finite-dimensional vector space E to itself. We also define the number operator $\widehat{N}$ by

$$\widehat{N} := \sum_{n=0}^{L} n\, \widehat{\mathbb{1}}_{\mathcal{H}_n} = \sum_{\kappa=1}^{L} \widehat{a}_\kappa^\dagger \widehat{a}_\kappa \qquad \text{(particle number operator)}.$$

For each linear subspace E of $\mathcal{H}$, we denote the orthogonal projector on E by $\Pi_E \in \mathcal{L}(\mathcal{H})$. We assume that the Hamiltonian of the system in the second-quantized formulation reads

$$\widehat{H} := \sum_{\kappa,\lambda=1}^{L} h_{\kappa\lambda} \widehat{a}_\kappa^\dagger \widehat{a}_\lambda + \frac{1}{2} \sum_{\kappa,\lambda,\nu,\xi=1}^{L} V_{\kappa\lambda\nu\xi} \widehat{a}_\kappa^\dagger \widehat{a}_\lambda^\dagger \widehat{a}_\xi \widehat{a}_\nu, \tag{2}$$

where the matrix $h \in \mathbb{R}^{L\times L}$ and the fourth-order tensor $V \in \mathbb{R}^{L\times L\times L\times L}$ satisfy the following symmetry properties:

$$h_{\kappa\lambda} = h_{\lambda\kappa} \quad \text{and} \quad V_{\kappa\lambda\nu\xi} = V_{\nu\xi\kappa\lambda}.$$

We denote by $\mathcal{D}$ the Grassmannian of rank-N orthogonal projectors in $\mathbb{R}^L$:

$$\mathcal{D} := \mathrm{Gr}_{\mathbb{R}}(N, L) = \{D \in \mathbb{R}^{L\times L}_{\mathrm{sym}} \mid D^2 = D,\ \mathrm{Tr}(D) = N\}, \tag{3}$$

and by $\mathrm{CH}(\mathcal{D})$ the convex hull of $\mathcal{D}$, that is,

$$\mathrm{CH}(\mathcal{D}) = \{D \in \mathbb{R}^{L\times L}_{\mathrm{sym}} \mid 0 \le D \le 1,\ \mathrm{Tr}(D) = N\}. \tag{4}$$

Physically, the set $\mathrm{CH}(\mathcal{D})$ corresponds to the set of (real-valued, mixed-state) N-representable one-body density matrices with N electrons, and $\mathcal{D}$ is the set of one-body density matrices generated by (real-valued) Slater determinants in $\mathcal{H}_N$.

We consider a fixed partition of the L sites into N_f non-overlapping fragments $\{\mathcal{I}_x\}_{x\in[\![1,N_f]\!]}$ of sizes $\{L_x\}_{x\in[\![1,N_f]\!]}$ such that $L_x < N$ for all x. Up to reordering the sites, we can assume that the partition is the following:

$$[\![1, L]\!] = \left\{ \underbrace{(1, \dots, L_1)}_{\mathcal{I}_1}, \underbrace{(1 + L_1, \dots, L_1 + L_2)}_{\mathcal{I}_2}, \dots, \underbrace{(1 + L_1 + \cdots + L_{N_f-1}, \dots, L)}_{\mathcal{I}_{N_f}} \right\}. \tag{5}$$

This partition corresponds to a decomposition of the space into N_f fragment subspaces fulfilling

$$\mathcal{H} = X_1 \oplus \cdots \oplus X_{N_f} \quad \text{with} \quad X_x := \mathrm{Span}(e_\kappa,\ \kappa \in \mathcal{I}_x). \tag{6}$$

For $M \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$, we set

$$\mathrm{Bd}(M) := \sum_{x=1}^{N_f} \Pi_x M \Pi_x, \tag{7}$$

where $\Pi_x := \Pi_{X_x}$ is the orthogonal projector on X_x. The operator $\mathrm{Bd} \in \mathcal{L}(\mathbb{R}^{L\times L}_{\mathrm{sym}})$ is the orthogonal projector onto the set of block-diagonal matrices for the partition (5) (endowed with the Frobenius inner product).

As we will see, a central intermediary in DMET is the diagonal blocks of the density matrix, $P = \mathrm{Bd}(D) \in \mathrm{Bd}(\mathcal{D})$. It is clear that these blocks must satisfy $0 \le P_x \le 1$ and $\sum_{x=1}^{N_f} \mathrm{Tr}(P_x) = N$. Conversely, it is easy to see that grouping these blocks together into a block-diagonal matrix produces a matrix in $\mathrm{CH}(\mathcal{D})$; therefore, we have

$$\mathcal{P} := \mathrm{Bd}(\mathrm{CH}(\mathcal{D})) = \left\{ P = \begin{pmatrix} P_1 & 0 & \cdots & 0 \\ 0 & P_2 & \cdots & 0 \\ \vdots & & \ddots & \vdots \\ 0 & 0 & \cdots & P_{N_f} \end{pmatrix} \right.$$
$$\left. \text{s.t. } \forall 1 \le x \le N_f,\ P_x \in \mathbb{R}^{L_x\times L_x}_{\mathrm{sym}},\ 0 \le P_x \le 1,\ \sum_{x=1}^{N_f} \mathrm{Tr}(P_x) = N \right\}. \tag{8}$$

From a geometrical viewpoint, $\mathcal{P}$ is a non-empty, compact, convex subset of an affine vector subspace of $\mathbb{R}^{L\times L}_{\mathrm{sym}}$ with base vector space

$$\mathcal{Y} := \left\{ Y = \begin{pmatrix} Y_1 & 0 & \cdots & 0 \\ 0 & Y_2 & \cdots & 0 \\ \vdots & & \ddots & \vdots \\ 0 & 0 & \cdots & Y_{N_f} \end{pmatrix} \text{ s.t. } \forall 1 \le x \le N_f,\ Y_x \in \mathbb{R}^{L_x\times L_x}_{\mathrm{sym}}, \sum_{x=1}^{N_f} \mathrm{Tr}(Y_x) = 0 \right\}. \quad (9)$$

The structure of the set $\mathrm{Bd}(\mathcal{D}) \subset \mathcal{P}$ is a more subtle issue that we will investigate in Section 6.

2.2 | The impurity high-level problem

Given one of the spaces X_x and a one-body density matrix $D \in \mathcal{D}$, we set:

$$W_{x,D} := X_x + DX_x = DX_x \oplus (1-D)X_x \quad (x\text{-th impurity subspace}). \quad (10)$$

We will assume in the following that

$$\dim(DX_x) = \dim((1-D)X_x) = \dim(X_x) = L_x \quad (\text{maximal-rank assumption}), \quad (11)$$

so that $\dim(W_{x,D}) = 2L_x$. Decomposing $\mathrm{Ran}(D)$ and $\mathrm{Ker}(D)$ as

$$\mathrm{Ran}(D) = DX_x \oplus \mathcal{H}^{\mathrm{core}}_{x,D} \quad \text{and} \quad \mathrm{Ker}(D) = (1-D)X_x \oplus \mathcal{H}^{\mathrm{virt}}_{x,D},$$

we obtain the following decomposition of $\mathcal{H} = \mathbb{R}^L$:

$$\mathcal{H} = W_{x,D} \oplus \underbrace{\mathcal{H}^{\mathrm{core}}_{x,D} \oplus \mathcal{H}^{\mathrm{virt}}_{x,D}}_{=:\mathcal{H}^{\mathrm{env}}_{x,D}}.$$

Note that the space $\mathcal{H}^{\mathrm{core}}_{x,D}$ has dimension $(N - L_x)$. The matrix D can be seen as the one-body density matrix associated with the Slater determinant

$$\Psi^0_{N,D} = \Psi^{0,\mathrm{imp}}_{x,D} \wedge \Psi^{0,\mathrm{core}}_{x,D} \quad \text{with} \quad \Psi^{0,\mathrm{imp}}_{x,D} \in \bigwedge^{L_x} DX_x \quad \text{and} \quad \Psi^{0,\mathrm{core}}_{x,D} \in \bigwedge^{(N-L_x)} \mathcal{H}^{\mathrm{core}}_{x,D},$$

where $\Psi^{0,\mathrm{imp}}_{x,D}$ and $\Psi^{0,\mathrm{core}}_{x,D}$ are normalized. More precisely, $\Psi^0_{N,D}$ is the Slater determinant built from an orthonormal basis of L_x orbitals in DX_x and an orthonormal basis of $(N - L_x)$ orbitals in $\mathcal{H}^{\mathrm{core}}_{x,D}$. The so-defined wave-function $\Psi^0_{N,D}$ is unique up to an irrelevant sign.

We denote by $\widehat{N}_{X_x} \in \mathcal{L}(\mathrm{Fock}(\mathcal{H}))$ the projection of the number operator onto the fragment Fock space $\mathrm{Fock}(X_x)$. Solving the impurity problem aims at minimizing, for a given $\mu \in \mathbb{R}$ which will be specified later, the thermodynamic potential

$$\langle \Psi | (\widehat{H} - \mu \widehat{N}_{X_x}) | \Psi \rangle \quad (12)$$

over the set of normalized trial states in Fock($\mathcal{H}$) of the form

$$\Psi = \Psi^{\mathrm{imp}}_{x,D} \wedge \Psi^{0,\mathrm{core}}_{x,D} \tag{13}$$

with $\Psi^{0,\mathrm{core}}_{x,D}$ fixed, and $\Psi^{\mathrm{imp}}_{x,D}$ in

$$\mathrm{Fock}(W_{x,D}) := \bigoplus_{n=0}^{2L_x} \bigwedge^{n} W_{x,D} \quad (x\text{-th impurity Fock space}).$$

The impurity Hamiltonian is the unique operator $\widehat{H}^{\mathrm{imp}}_{x,D}$ on Fock($W_{x,D}$) such that

$$\forall \Psi^{\mathrm{imp}}_{x,D} \in \mathrm{Fock}(W_{x,D}), \quad \langle \Psi^{\mathrm{imp}}_{x,D} | \widehat{H}^{\mathrm{imp}}_{x,D} | \Psi^{\mathrm{imp}}_{x,D} \rangle = \langle \Psi^{\mathrm{imp}}_{x,D} \wedge \Psi^{0,\mathrm{core}}_{x,D} | \widehat{H} | \Psi^{\mathrm{imp}}_{x,D} \wedge \Psi^{0,\mathrm{core}}_{x,D} \rangle. \tag{14}$$

For an explicit expression of $\widehat{H}^{\mathrm{imp}}_{x,D}$, see Proposition 8.

The impurity problem defined by (12)–(13) can then be reformulated as

$$\min_{\Psi^{\mathrm{imp}}_{x,D} \in \mathrm{Fock}(W_{x,D}), \|\Psi^{\mathrm{imp}}_{x,D}\|=1} \langle \Psi^{\mathrm{imp}}_{x,D} | \widehat{H}^{\mathrm{imp}}_{x,D} - \mu \widehat{N}_{X_x} | \Psi^{\mathrm{imp}}_{x,D} \rangle \quad (\text{impurity problem}). \tag{15}$$

In practice, this full-CI problem in the Fock space Fock($W_{x,D}$) is solved by an approximate correlated wave-function method such as CASSCF, CCSD, or DMRG for example, but we assume in this analysis that it can be solved exactly.

If (15) has a non-degenerate ground state for all x, we denote the one-body ground-state density matrices by $P_{\mu,x}(D)$, seen as matrices in $\mathbb{R}^{L\times L}_{\mathrm{sym}}$, and finally set

$$\boxed{F^{\mathrm{HL}}_{\mu,x}(D) := \Pi_{X_x} P_{\mu,x}(D) \Pi_{X_x}.} \tag{16}$$

Let us remark incidentally that if the ground state of the impurity problem is degenerate, we can either consider $F^{\mathrm{HL}}_{\mu,x}(D)$ as a multivalued function or define them from finite-temperature versions of (15), which are strictly convex compact problems on the set of density operators on the Fock space, and therefore always have a unique minimizer. We will not proceed further in this direction and only consider here the case of impurity problems with non-degenerate ground states.

The combination of the N_f impurity problems introduced in (15) (see also (16)) gives rise to a high-level DMET map F^{HL}

$$\mathcal{D} \ni D \mapsto F^{\mathrm{HL}}(D) \in \mathcal{P} \tag{17}$$

formally defined by

$$\boxed{F^{\mathrm{HL}}(D) := \sum_{x=1}^{N_f} F^{\mathrm{HL}}_{\mu,x}(D) \quad (\text{high-level map})} \tag{18}$$

with $\mu \in \mathbb{R}$ chosen such that $\mathrm{Tr}(F^{\mathrm{HL}}(D)) = N$. The domain of F^{HL} and the regularity properties of this map will be studied in Section 5.

2.3 | The global low-level problem

The low-level map is defined by

$$\boxed{F^{\mathrm{LL}}(P) := \underset{D\in\mathcal{D},\ \mathrm{Bd}(D)=P}{\operatorname{argmin}} \mathcal{E}^{\mathrm{HF}}(D) \quad \text{(low-level map)},} \tag{19}$$

where $\mathcal{E}^{\mathrm{HF}}$ is the HF (mean-field) energy functional of the trial density-matrix D. The latter reads

$$\mathcal{E}^{\mathrm{HF}}(D) := \mathrm{Tr}(hD) + \frac{1}{2}\mathrm{Tr}(J(D)D) - \frac{1}{2}\mathrm{Tr}(K(D)D), \tag{20}$$

where

$$[J(D)]_{\kappa\lambda} := \sum_{\nu,\xi=1}^{L} V_{\lambda\xi\kappa\nu}D_{\nu\xi} \quad \text{and} \quad [K(D)]_{\kappa\lambda} := \sum_{\nu,\xi=1}^{L} V_{\kappa\xi\nu\lambda}D_{\nu\xi}. \tag{21}$$

The existence and uniqueness of a minimizer to (19) will be discussed in Section 6.

2.4 | The DMET problem

Finally, the full DMET map is formally defined as the self-consistent solution to the system

$$\boxed{\begin{aligned} D &= F^{\mathrm{LL}}(P) \in \mathcal{D},\\ P &= F^{\mathrm{HL}}(D) \in \mathcal{P}. \end{aligned}}$$

In particular, $D = F^{\mathrm{LL}}(P)$ implies that $P = \mathrm{Bd}(D)$. Equivalently, we can formulate the problem as

$$\boxed{P = F^{\mathrm{DMET}}(P) := F^{\mathrm{HL}}(F^{\mathrm{LL}}(P)).}$$

Assuming that the solution to this fixed-point problem exists and is unique, P is expected to provide a good approximation of the diagonal blocks (in the decomposition (6) of $\mathcal{H}$) of the ground-state one-body density matrix of the interacting system. The mathematical properties of this self-consistent loop will be studied in the next section, first for the non-interacting case, and second, for the interacting case in a perturbative regime.

3 | MAIN RESULTS

We now embed the Hamiltonian H into the family of Hamiltonians

$$\widehat{H}_\alpha := \sum_{\kappa,\lambda=1}^{L} h_{\kappa\lambda}\widehat{a}_\kappa^\dagger\widehat{a}_\lambda + \frac{\alpha}{2}\sum_{\kappa,\lambda,\nu\xi=1}^{L} V_{\kappa\lambda\nu\xi}\widehat{a}_\kappa^\dagger\widehat{a}_\lambda^\dagger\widehat{a}_\xi\widehat{a}_\nu, \quad \alpha\in\mathbb{R}, \tag{22}$$

acting on Fock($\mathcal{H}$). For $\alpha = 0$, we obtain the one-body Hamiltonian

$$\widehat{H}_0 := \sum_{\kappa,\lambda=1}^{L} h_{\kappa\lambda} \widehat{a}_\kappa^\dagger \widehat{a}_\lambda \tag{23}$$

describing non-interacting particles, and we recover the original Hamiltonian $\widehat{H}$ for $\alpha = 1$. We denote by F_α^{HL}, F_α^{LL}, and F_α^{DMET} the high-level, low-level, and DMET maps constructed from $\widehat{H}_\alpha$.

We first assume that the non-interacting problem is non-degenerate. Denoting by ε_n the n-th lowest eigenvalue of h (counting multiplicities), this condition reads

(A1) $\varepsilon_N < 0 < \varepsilon_{N+1}$,

where without loss of generality we have chosen the Fermi level to be 0. Assumption (A1) indeed implies that the ground-state of $\widehat{H}_0$ in the N-particle sector of the Fock space is non-degenerate, and that the ground-state one-body density is the rank-N orthogonal projector given by

$$D_0 = \mathbb{1}_{(-\infty,0]}(h). \tag{24}$$

By perturbation theory, the ground state of $\widehat{H}_\alpha$ in the N-particle sector is non-degenerate for all $\alpha \in (-\alpha_+, \alpha_+)$ for some $0 < \alpha_+ \leq +\infty$. We denote by $D_\alpha^{\mathrm{exact}}$ the corresponding ground-state one-body density matrix. As a consequence of analytic perturbation theory for hermitian matrices, the map $(-\alpha_+, \alpha_+) \ni \alpha \mapsto D_\alpha^{\mathrm{exact}} \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$ is real-analytic.

Second, we make the maximal-rank assumption:

(A2) For all $1 \leq x \leq N_f$, $\dim(D_0 X_x) = \dim((1 - D_0)X_x) = \dim(X_x) = L_x$.

Assumption (A2) implies that the impurity problem (15) for $\widehat{H} = \widehat{H}_0$ and $D = D_0$ is well-defined for each x and each μ. We emphasize however that this does not prejudge that the so-obtained N_f impurity problems are well-posed (i.e., have a unique ground-state) for a given value of μ, nor *a fortiori* that D_0 is in the domain of the high-level map F_0^{HL}. We will elaborate more on the meaning of Assumptions (A2) in Section 5.

DMET is then consistent in the non-interacting case:

Proposition 1 ($P_0 := \mathrm{Bd}(D_0)$ is a fixed point of the DMET map for $\alpha = 0$). *Under Assumptions (A1)–(A2), $P_0 := \mathrm{Bd}(D_0)$ is a fixed point of the non-interacting DMET iterative scheme, that is P_0 is in the domain of F_0^{LL}, D_0 is in the domain of F_0^{HL}, and $F_0^{\mathrm{DMET}}(P_0) = P_0$.*

Remark 2. We formally define the *high-level HF* map

$$F_{\mathrm{MF}}^{\mathrm{HL}} : \mathcal{D} \to \mathcal{P},$$

as the high-level map constructed from the HF N-body Hamiltonian

$$\widehat{H}_D^{\mathrm{HF}} := \sum_{\kappa,\lambda=1}^{L} [h^{\mathrm{HF}}(D)]_{\kappa\lambda} \widehat{a}_\kappa^\dagger \widehat{a}_\lambda,$$

where

$$h^{\mathrm{HF}}(D) = h + J(D) - K(D) \tag{25}$$

is the one-particle mean-field (Fock) Hamiltonian. Using exactly the same arguments as in the proof of Proposition 1, we obtain that the low-level map F^{LL} satisfies the mean-field consistency property

$$F^{\mathrm{LL}}(F^{\mathrm{HL}}_{\mathrm{HF}}(D_*)) = D_*,$$

for any HF ground state D_*. We will make use of this important observation in the proof of Theorem 4.

We now study the DMET equations in the perturbative regime of α small. In order to use perturbative techniques, we need to determine the space in which we seek P. Generically, at $\alpha \neq 0$, we expect P to be equal to the block diagonal of the one-body density matrix, which is not a projector. Therefore it is natural to seek P in $\mathcal{P} = \mathrm{Bd}(\mathrm{CH}(\mathcal{D}))$. However, in the DMET method, D is constrained to be a projector, and therefore P will necessarily belong to $\mathrm{Bd}(\mathcal{D})$. We will study in Section 6 the relationship between the two sets $\mathcal{P}$ and $\mathrm{Bd}(\mathcal{D})$ (the N-representability problem), and in particular show that, in the regime of interest to DMET (many relatively small fragments, so that $L \gg \max_x L_x$), the two sets are (generically) locally the same. Therefore, it is natural to assume the local N-representability condition:

(A3) The linear map Bd is surjective from $\mathcal{T}_{D_0}\mathcal{D}$ to $\mathcal{Y}$,

where $\mathcal{Y}$ is the vector subspace defined in (9). Indeed, $\mathcal{P}$ is a (non-empty, compact, convex) subset of the affine space $P_0 + \mathcal{Y}$ and Assumption (A2) implies that $P_0 \in \mathcal{P}^\circ$, where $\mathcal{P}^\circ$ is the interior of $\mathcal{P}$ in $P_0 + \mathcal{Y}$. Thus $\mathcal{Y}$ can be identified with the tangent space at P_0 to the manifold $\mathcal{P}^\circ$. By the local submersion theorem, this implies that any P in the neighborhood of P_0 can be expressed as the block diagonal of a density matrix in the neighborhood of D_0 in $\mathcal{D}$.

Our last assumption is concerned with the response properties of the impurity problems at the non-interacting level. Consider a self-adjoint perturbation $Y \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$ of the one-particle Hamiltonian h, non-local but block-diagonal in the fragment decomposition, that is such that $Y \in \mathbb{R}I_L + \mathcal{Y}$, and denote by $\widetilde{F^{\mathrm{HL}}}_{h+Y}(D)$ the non-interacting high-level map obtained by replacing h with $h + Y$ (so that $\widetilde{F^{\mathrm{HL}}}_{h}(D) = F^{\mathrm{HL}}_0$). Formally, we have

$$\widetilde{F^{\mathrm{HL}}}_{h+Y}(D_0) = P_0 + R[Y] + o(\|Y\|), \tag{26}$$

with $R : \mathbb{R}I_L + \mathcal{Y} \to \mathcal{Y}$ linear (the fact that $R[Y] \in \mathcal{Y}$ is due to particle-number conservation). The map R can be interpreted as a non-interacting static 4-point density-density linear response function for frozen impurity spaces. It follows from Assumption (A1) that constant perturbations do not modify the density matrix: $R(I_L) = 0$. Our fourth assumption reads:

(A4) the 4-point linear response function $R : \mathcal{Y} \to \mathcal{Y}$ is invertible.

This condition is somewhat reminiscent of the Hohenberg–Kohn theorem from Density Functional Theory. Together with the local inversion theorem, it implies that, locally around h, in the

non-interacting case and for frozen impurity spaces W_{x,D_0}, the high-level map defines a one-to-one correspondence between non-local fragment potentials (up to a constant shift) and fragment density matrices.

Remark 3. We will show in Section 7.6.4 that in the case when $N_f = L$ (one site per fragment), it holds: under Assumptions (A1)–(A2),

$$\text{(A3) is satisfied} \Rightarrow D_0 \text{ is an irreducible matrix} \iff \text{(A4) is satisfied.}$$

On the other hand, numerical simulations indicate that in the general case, Assumption (A4) is not a consequence of Assumptions (A1)–(A3).

We are now in position to state our main results.

Theorem 4 (DMET is well-posed in the perturbative regime). *Under assumptions (A1)–(A4), there exist* $0 < \widetilde{\alpha}_+ \leq \alpha_+$, *and a neighborhood* Ω *of* D_0 *in* $\mathcal{D}$ *such that for all* $\alpha \in (-\widetilde{\alpha}_+, \widetilde{\alpha}_+)$, *the fixed-point DMET problem*

$$P_\alpha^{\mathrm{DMET}} = F_\alpha^{\mathrm{HL}}(D_\alpha^{\mathrm{DMET}}), \quad D_\alpha^{\mathrm{DMET}} = F_\alpha^{\mathrm{LL}}(P_\alpha^{\mathrm{DMET}})$$

has a unique solution $(D_\alpha^{\mathrm{DMET}}, P_\alpha^{\mathrm{DMET}})$ *with* $D_\alpha^{\mathrm{DMET}} \in \Omega$ *(otherwise stated, the DMET map for* H_α *has a unique fixed point* P_α^{DMET} *in the neighborhood of* P_0*). In addition, the maps* $(-\widetilde{\alpha}_+, \widetilde{\alpha}_+) \ni \alpha \mapsto D_\alpha^{\mathrm{DMET}} \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$ *and* $(-\widetilde{\alpha}_+, \widetilde{\alpha}_+) \ni \alpha \mapsto P_\alpha^{\mathrm{DMET}} \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$ *are real-analytic and such that*

$$D_0^{\mathrm{DMET}} = D_0 = \mathbb{1}_{(-\infty,0]}(h), \quad P_0^{\mathrm{DMET}} = P_0 = \mathrm{Bd}(D_0).$$

As is standard, the first-order perturbation of the exact density matrix is given by the HF method. DMET is able to reproduce this, and is therefore exact up to first order:

Theorem 5 (DMET is exact to first order). *Under Assumptions (A1)–(A4) and with the notation of Theorem 4, it holds*

$$D_\alpha^{\mathrm{DMET}} = D_\alpha^{\mathrm{exact}} + O(\alpha^2) = D_\alpha^{\mathrm{HF}} + O(\alpha^2),$$

where D_α^{HF} *is the HF ground-state density matrix for* $\widehat{H}_\alpha$, *which is unique for* α *small enough.*

The numerical simulations reported in the next section show that such exactness property is not expected to hold at second order.

In the weakly interacting regime, the solution D_α^{DMET} to the DMET fixed-point problem is the only physical one. Indeed, it is the only one laying in the vicinity of D_0, where the exact ground-state density matrix must be located for small α, by analytic perturbation theory.

Remark 6. A variant of DMET, which could be termed *density embedding theory* (DET), would consist in choosing fragments of arbitrary sizes for the high-level step, but in using only the diagonal of the density matrix (i.e., the density) in the low-level step. Denoting by Δ the space of $L \times L$ real diagonal matrices with entries between 0 and 1 and trace N, and by $\mathrm{Dg} : \mathcal{P} \ni P \mapsto \mathrm{Diag}(P) \in \Delta$,

the DET fixed-point problem would consist in solving $F^{\mathrm{DET}}(\rho) = \rho$, where $F^{\mathrm{DET}} : \Delta \to \Delta$ is defined as

$$F^{\mathrm{DET}} = \mathrm{Dg} \circ F^{\mathrm{HL}} \circ F^{\mathrm{LL}}_{\mathrm{DET}} \quad \text{with} \quad F^{\mathrm{LL}}_{\mathrm{DET}}(\rho) := \operatorname*{argmin}_{D \in \mathcal{D},\, \mathrm{Dg}(D)=\rho} \mathcal{E}^{\mathrm{HF}}(D).$$

Similar arguments as the ones used in the proofs of Proposition 1, Theorem 4, and Theorem 5, allow one to show that

1. under Assumptions (A1)–(A2), $\rho_0 = \mathrm{Dg}(D_0)$ is a fixed point of the non-interacting DET scheme;
2. under Assumptions (A1), (A2),
   (A3') all diagonal elements of D_0 are in the open interval (0,1),
   (A4') the non-interacting high-level map satisfies some local Hohenberg–Kohn theorem, in the sense that, locally around the one-body Hamiltonian h, there is a one-to-one correspondence between diagonal perturbations of h (up to uniform shift) and perturbations of the density,
   the DET fixed-point problem has a unique physical solution in the weakly-interacting regime ($0 \leq \alpha \ll 1$), which is exact up to first-order in α.

A rigorous analysis of the respective merits of the different variants of quantum embedding methods is outside the scope of this article, and is the subject of ongoing work by some of us.

4 | NUMERICAL SIMULATIONS

In this section, we perform numerical investigations of DMET for two distinct test systems: The first system is H_{10} in a circular geometry which serves as a benchmark where DMET has been previously recognized for its exceptional performance [24]. By studying this system, we aim to reaffirm the efficacy of DMET and numerically showcase that DMET is exact to first order in the non-interacting limit. However, to gain a comprehensive understanding of DMET's limitations, we also explore a second system which is an H_6 variant. This particular system allows us to numerically scrutinize the assumptions made in the analysis presented above. Through these numerical investigations, we aim to provide valuable insights into the mathematical structure of DMET, paving the way for further advancements and improvements in this promising computational approach. Throughout this section, we denote by $\|\cdot\|_{\mathrm{F}}$ the Frobenius norm on matrix spaces.

4.1 | H_{10} ring

We consider a circular arrangement of ten hydrogen atoms, with a nearest-neighbor distance of 1.5 a_0 between each pair of atoms (where $a_0 \simeq 0.529$ Å is the Bohr radius). The system is treated using the STO-6G basis set and is half-filled, that is, containing ten electrons. We partition the system into five fragments, each consisting of two atoms, as shown in Figure 1.

In order to numerically confirm that DMET is exact to first order for this "well-behaved" system, we determine P_α for $\alpha \in [0, 1]$ and compute $\|dP_\alpha/d\alpha\|_F$. Figure 2 compares the DMET result with

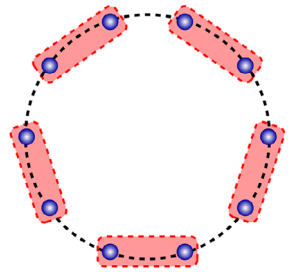

FIGURE 1 Depiction of the H_{10} system in circular geometry. The red-shaded areas show the chosen fragmentation.

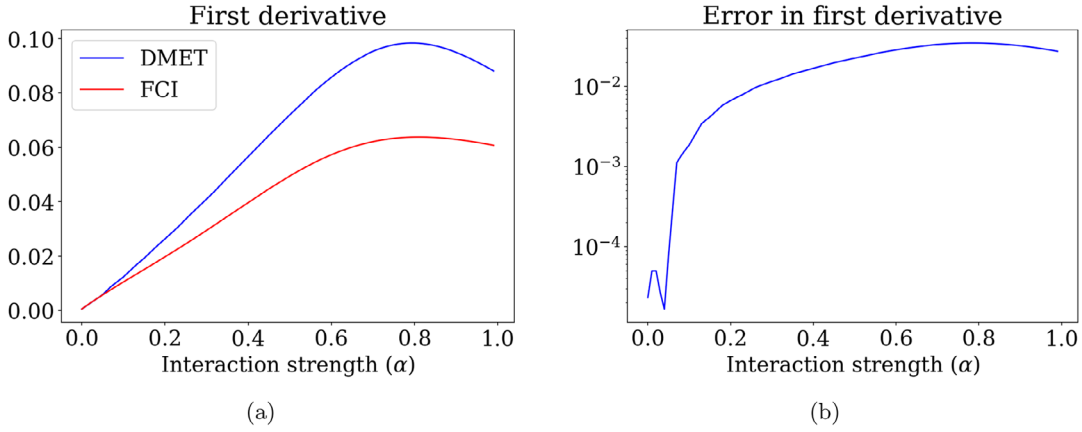

FIGURE 2 (a) Shows $\|dP_\alpha/d\alpha\|_F$ for DMET and FCI, respectively (b) Shows the error on $dP_\alpha/d\alpha$ between DMET and FCI, measured in Frobenius norm. DMET, density matrix embedding theory; FCI, full configuration interaction.

the exact diagonalization result (abbreviated FCI). We clearly see that DMET is indeed exact to first order for the considered system.

4.2 | H_6 model

In this section, we will numerically investigate the assumptions required for the analysis presented in this article. To that end, we consider a non-interacting H_6^{4-} system, undergoing the following transition on a circular geometry. We begin by placing three hydrogen molecules in equilibrium geometry, that is, bond length of $1.4\,a_0$, equidistantly on a circle of radius $3\,a_0$. We then dissociate each hydrogen molecule while maintaining a circular geometry. Specifically, we break each hydrogen molecule in such a way that the hydrogen atoms from neighboring molecules can form new molecules. We stop this transition at $\Theta = \Theta_{\max}$, when the hydrogen atoms from neighboring molecules form new hydrogen molecules in equilibrium geometry. We steer this transition with the angle Θ that measures the displacement of the individual hydrogen atoms relative to their initial positions. The dissociation is done in a manner that maintains the circular arrangement of the hydrogen atoms throughout the process, see Figure 3 for a schematic depiction of

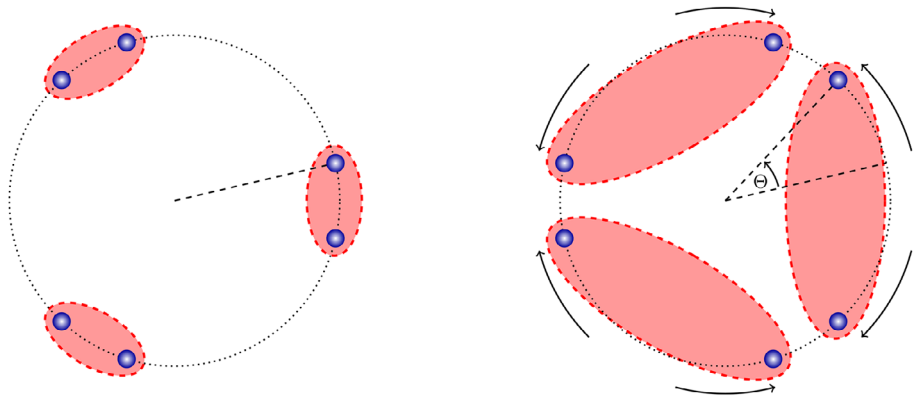

FIGURE 3 Schematic depiction of the considered H_6 transition. The left panel shows the initial configuration for $\Theta = 0$; the right panel shows the final configuration $\Theta = \Theta_{\max}$. The red-shaded areas depict the imposed fragmentation. The arrows indicate the transition of the hydrogen atoms for $\Theta \in [0, \Theta_{\max}]$.

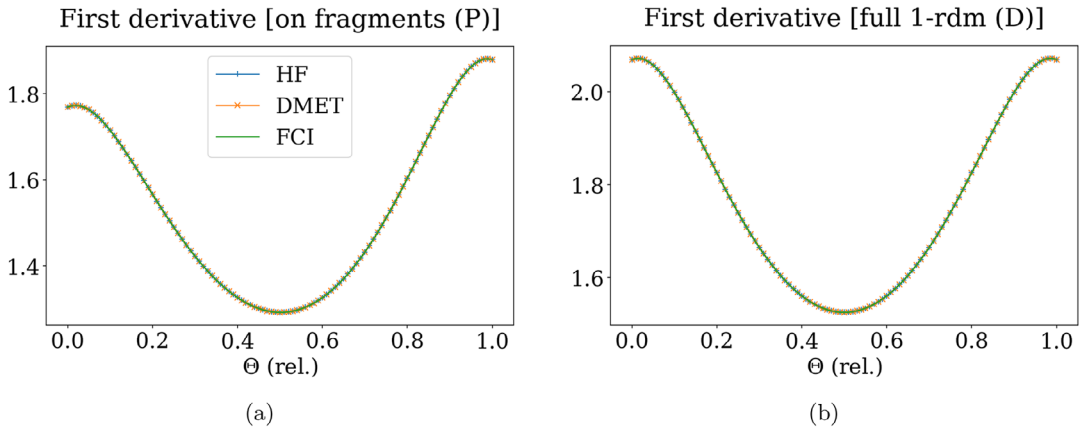

FIGURE 4 (a) Shows $\|\partial_\alpha P_\alpha|_{\alpha=0}\|_F$ for HF, DMET, and FCI (b) Shows $\|\partial_\alpha D_\alpha|_{\alpha=0}\|_F$ for HF, DMET, and FCI. DMET, density matrix embedding theory; FCI, full configuration interaction; HF, Hartree–Fock.

this process and a depiction of Θ. The system is partitioned into 3 fragments that correspond to the initial molecules. Note that the fragments remain unchanged during the transition process. In order to fulfill the N-representability condition (33) below (which is necessary for Assumption (A3) to be fulfilled), N must be chosen between 6 and 10 (in this example $N_f = 3$ and $L_x = 2$ hence $\dim(\mathcal{Y}) = 8$). As shown in [12], DMET encounters challenges when being doped; we therefore opt for $N = 10$. The system is discretized using the 6-31G basis set.

In order to numerically depict Theorem 5, we compute P_α and D_α using a mean-field theory approach (HF), DMET, and the exact diagonalization (FCI), and compare these quantities for $\alpha = 0$ as well as their first derivatives with respect to α. Note that in the non-interacting limit, the mean-field theory is exact, which is reflected in our simulations. We indeed observe that $\sup_\Theta \|P_0^{\mathrm{HF}}(\Theta) - P_0^{\mathrm{FCI}}(\Theta)\|_F$ and $\sup_\Theta \|D_0^{\mathrm{HF}}(\Theta) - D_0^{\mathrm{FCI}}(\Theta)\|_F$ are equal to zero up to numerical accuracy, while $\sup_\Theta \|P_0^{\mathrm{DMET}}(\Theta) - P_0^{\mathrm{FCI}}\|_F(\Theta)$, $\sup_\Theta \|D_0^{\mathrm{DMET}}(\Theta) - D_0^{\mathrm{FCI}}(\Theta)\|_F$ are respectively of the order of 10^{-13} and 10^{-7} with the chosen convergence thresholds. Figure 4 shows the first-order exactness of DMET in the non-interacting limit for the H_6^{4-} model.

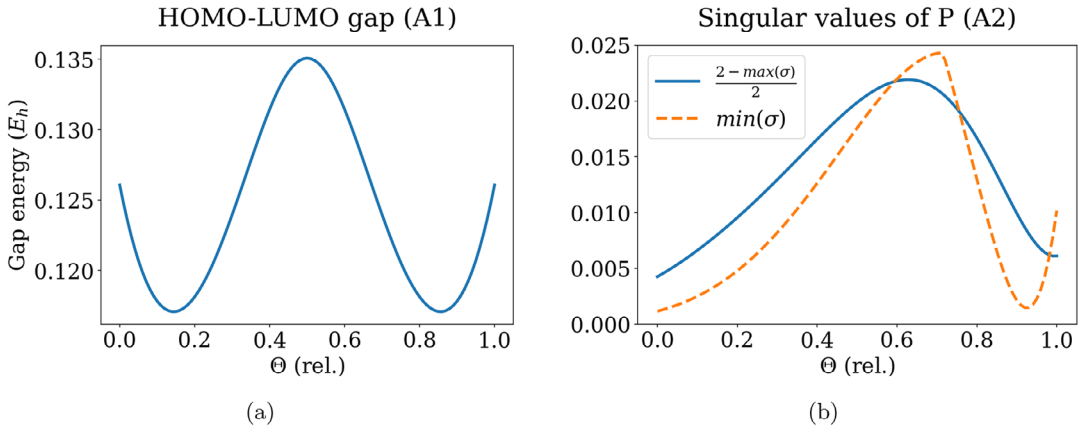

FIGURE 5 (a) Shows the HOMO-LUMO gap for the H_6 model as a function of Θ for $\alpha = 0$. (b) Shows the largest and smallest singular values of P for the H_6 model as a function of Θ for $\alpha = 0$.

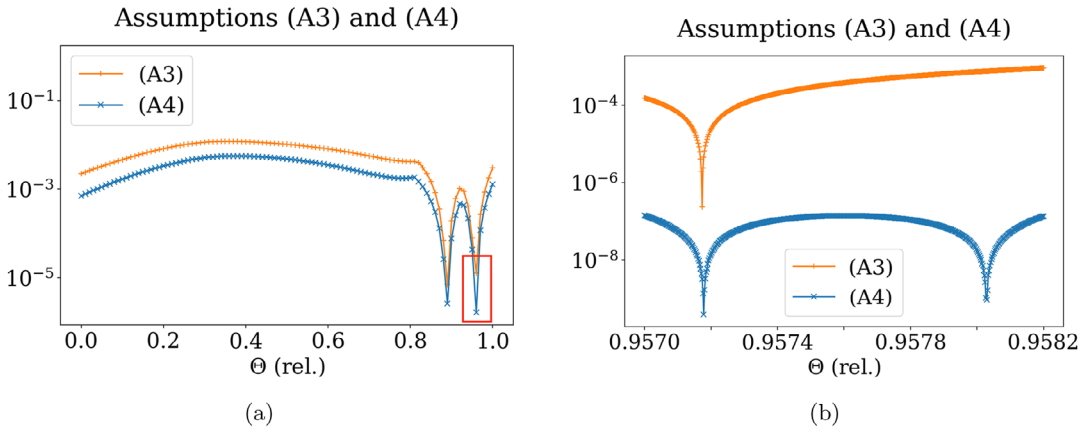

FIGURE 6 (a) The orange line shows the lowest eigenvalue of $S := (\mathrm{Bd}|_{T_{D_0}\mathcal{D}\to\mathcal{Y}})^* \mathrm{Bd}|_{T_{D_0}\mathcal{D}\to\mathcal{Y}}$ for the H_6 model as a function of Θ for $\alpha = 0$ (which corresponds to (A3)), and the blue line shows the smallest singular value $\sigma_{\min}$ of $R|_{\mathcal{Y}\to\mathcal{Y}}$ (which corresponds to (A4)). (b) Shows a zoomed version of (a) around the second (local) minimum.

Our numerical investigations include an analysis of Assumptions (A1)–(A4). We present a check of Assumptions (A1) and (A2) in Figure 5. Assumption (A1) can be directly tested by calculating the HOMO-LUMO gap of the non-interacting Hamiltonian under consideration for each value of Θ. Furthermore, Assumption (A2) can be tested by monitoring the behavior of the smallest and largest singular values of the matrix P_0 as a function of the variable Θ (see Lemma 7).

The validity of assumptions (A3) and (A4) is tested in Figure 6 by monitoring the lowest eigenvalue of the operator $S := (\mathrm{Bd}|_{T_{D_0}\mathcal{D}\to\mathcal{Y}})^* \mathrm{Bd}|_{T_{D_0}\mathcal{D}\to\mathcal{Y}}$ (which corresponds to (A3)), and the smallest singular value of the operator $R|_{\mathcal{Y}\to\mathcal{Y}}$ (which corresponds to (A4)).

We see that Assumptions (A1) and (A2) are uniformly fulfilled over the whole range $[0, \Theta_{\max}]$. Assumption (A3) seems to be satisfied for all Θ except two values $\Theta_1 \simeq 0.885$ and $\Theta_2 \simeq 0.957$. Careful testing around Θ_2 shows that Assumption (A4) is additionally not satisfied at $\Theta_3 \simeq 0.958$,

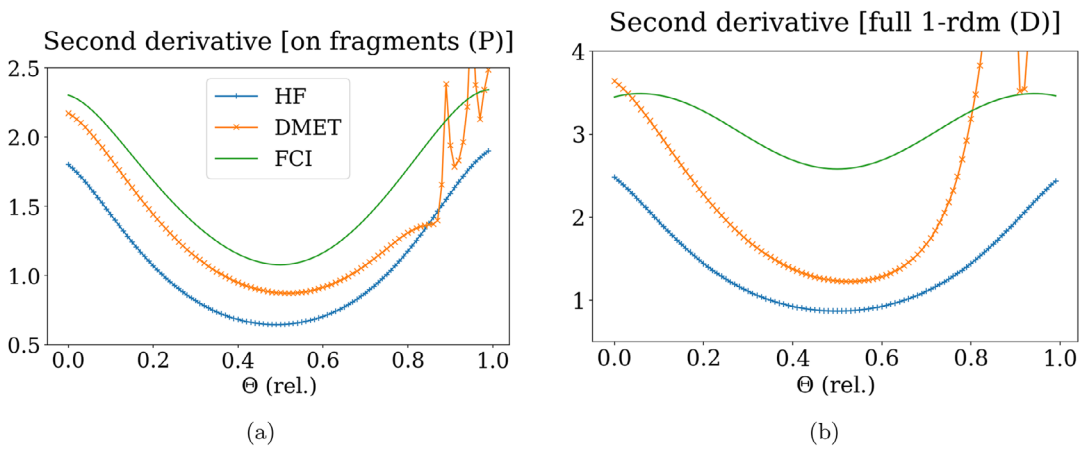

FIGURE 7 (a) shows $\|\partial_\alpha^2 P_\alpha|_{\alpha=0}\|_F$ for HF, DMET, and FCI (b) shows $\|\partial_\alpha^2 D_\alpha|_{\alpha=0}\|_F$ for HF, DMET, and FCI. DMET, density matrix embedding theory; FCI, full configuration interaction; HF, Hartree–Fock.

where all other assumptions are satisfied. This illustrates the fact that in the general case $N_f < L$, Assumption (A4) is independent of Assumptions (A1)–(A3) (see Remark 3).

Figure 7a shows the Frobenius norms of the second derivative of P_α and D_α at $\alpha = 0$ for HF, DMET, and FCI. We see that the three methods give different results, and that the result in Theorem 5 is therefore optimal. We also observe that for DMET, the second derivatives become noisy in the range of Θ's where Assumptions (A3) and (A4) are poorly or not satisfied. This is probably due to conditioning issues or to the use of convergence thresholds not directly connected to the computed quantity of interest. The numerical analysis of DMET is left for future work.

We now investigate more closely the violation of the hypotheses at Θ_3, where R is not invertible, but (A3) is still satisfied. To that end, we compute the differential of $F_0^{\mathrm{DMET}}(P_0)$ at P_0, as a function of Θ, and see that for Θ close to Θ_3, $F_0^{\mathrm{DMET}}(P_0(\Theta))$ possesses a simple real eigenvalue which transitions from being positive (for $\Theta < \Theta_3$) to being negative (for $\Theta > \Theta_3$), with all other eigenvalues having negative real parts. As is standard, this type of eigenvalue crossing generically gives rise to a transcritical bifurcation [47]. This suggests the existence of another branch of solutions $P_1(\Theta)$ of $P = F_0^{\mathrm{DMET}}(P)$, which collides with $P_0(\Theta)$ at $\Theta = \Theta_3$, and such that the largest eigenvalue of the differential of F_0^{DMET} at P_0 has the opposite sign to that at P_1.

To find this branch of solutions, we employ a Newton algorithm on F_0^{DMET}. Since we are looking at small differences, this requires accurate computations of F_0^{HL} and F_0^{LL} as well as their differentials (without resorting to finite differences). The differential of F_0^{HL} is computed analytically by perturbation theory (taking into account the self-consistent Fermi level). For F_0^{LL}, we implemented a manifold Newton algorithm to compute an accurate solution of the problem defining the low-level solver. This is done by, starting from the point D_n, parametrizing D_{n+1} as $D(X)$ with an unconstrained matrix X as in the proof of Lemma 11, and then performing a Newton step on the Lagrangian $L(X, \Lambda)$ that corresponds to minimizing $\mathcal{E}^{\mathrm{HF}}(D(X))$ subject to $\mathrm{Bd}(D(X)) = P$. From the Hessian of the Lagrangian one can also compute the differential of F_0^{LL}, and then ultimately of F_0^{DMET}.

To initialize the Newton algorithm on F_0^{DMET} at a given Θ close to Θ_3, we start from P_0, and compute the eigenvector Y of dF_0^{DMET} associated with the eigenvalue that crosses zero. Then, we run a Newton algorithm started from $P_0 + \alpha(\Theta - \Theta_3)Y$, where α is an empirically chosen parameter (its precise determination involves higher derivatives [47], which are cumbersome to compute). We

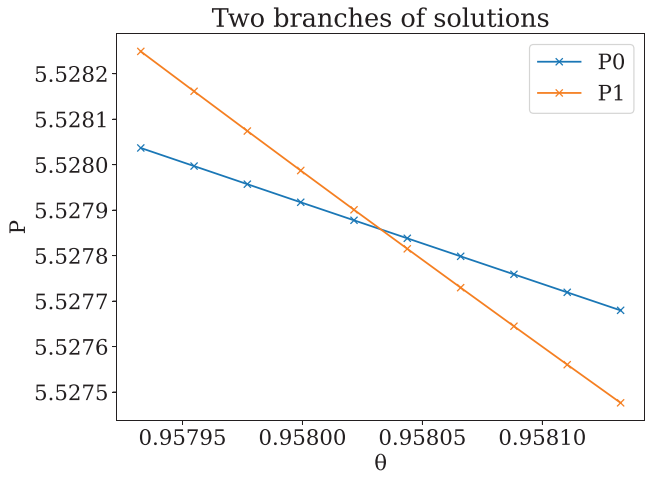

FIGURE 8 The two branches P_0 and P_1 (displayed are the scalars $\sum_{ij} P_{ij}$) as functions of Θ near $\Theta = \Theta_3$.

observe the two branches P_0 and P_1 shown in Figure 8, confirming the transcritical bifurcation. Let us emphasize that this bifurcation is not due to symmetry breaking, as can be shown from a detailed analysis of the solutions P_0 and P_1 (see Appendix B).

5 | IMPURITY PROBLEMS AND HIGH-LEVEL MAP

5.1 | Impurity Hamiltonians

It follows from the considerations in Section 2.2 that if

$$\forall x \in [\![1, N_f]\!], \quad \dim(DX_x) = \dim((1-D)X_x) = L_x.$$

the impurity problem is well-defined for each fragment since the maximal rank assumption (11) is satisfied for each X_x. The next lemma gives useful equivalent characterizations of these conditions.

Let us introduce the matrix

$$E_x := \mathrm{mat}_{\mathcal{B}_{\mathrm{at}}}(e_\kappa, \kappa \in \mathcal{I}_x) = \begin{pmatrix} 0_{L'_x \times L_x} \\ I_{L_x} \\ 0_{L''_x \times L_x} \end{pmatrix} \in \mathbb{R}^{L \times L_x} \quad \text{with} \begin{cases} L'_x := \sum_{1 \le x' < x} L_{x'} \\ L''_x := \sum_{x < x' \le N_f} L_{x'} \end{cases} \tag{27}$$

representing the orbitals of fragment $x \in [\![1, N_f]\!]$, whose range is X_x. We recall that $\mathcal{P}^\circ$ denotes the interior of the set $\mathcal{P} = \mathrm{Bd}(\mathrm{CH}(\mathcal{D}))$ in the affine space $P_0 + \mathcal{Y}$.

Lemma 7 (Compatibility conditions). *Let $D \in \mathcal{D}$. The following assertions are equivalent:*

1. $\mathrm{Bd}(D) \in \mathcal{P}^\circ$;
2. $\forall x \in [\![1, N_f]\!], \quad \dim(DX_x) = \dim((1-D)X_x) = L_x$;

3. $\forall x \in [\![1, N_f]\!]$, $0 < E_x^T D E_x < 1$ *(all the eigenvalues of* $E_x^T D E_x$ *are in (0,1));*
4. $\forall x \in [\![1, N_f]\!]$, $E_x^T D E_x \in \mathrm{GL}_{\mathbb{R}}(L_x)$ *and* $E_x^T (1-D) E_x \in \mathrm{GL}_{\mathbb{R}}(L_x)$.

If D satisfies these conditions, we say that it is compatible with the fragment decomposition.

It is easily seen that if D is compatible with the fragment decomposition, then the column vectors defined by the matrix

$$C^x(D) := \left(D E_x \left(E_x^T D E_x\right)^{-1/2} \middle| (1-D) E_x \left(E_x^T (1-D) E_x\right)^{-1/2} \right) \in \mathbb{R}^{L \times 2L_x} \tag{28}$$

form an orthonormal basis of the impurity one-particle state space $W_{x,D}$ defined in (10). More precisely, the first L_x columns of $C^x(D)$ form an orthonormal basis of DX_x and its last L_x columns form an orthonormal basis of $(1-D)X_x$. Likewise, the column vectors of the matrix

$$\widetilde{C}^x(D) := \left(E_x \middle| (1-\Pi_x) D E_x \left(E_x^T D (1-\Pi_x) D E_x\right)^{-1/2} \right) \in \mathbb{R}^{L \times 2L_x} \tag{29}$$

form an orthonormal basis of $X_x \oplus (1-\Pi_x) D X_x$.

We denote by $\widehat{a}_j^x(D)$ and $\widehat{a}_j^x(D)^\dagger$, $1 \le j \le 2L_x$ the annihilation and creation operators in the basis of the columns of $C^x(D)$:

$$\widehat{a}_j^x(D) = \sum_{\kappa=1}^{L} (C^x(D))_{\kappa j} \widehat{a}_\kappa, \quad \widehat{a}_j^x(D)^\dagger = \sum_{\kappa=1}^{L} (C^x(D))_{\kappa j} \widehat{a}_\kappa^\dagger.$$

These operators allow for an explicit form of the impurity Hamiltonian $\widehat{H}_{x,D}^{\mathrm{imp}}$ as follows.

Proposition 8 (Impurity Hamiltonian). *Let* $D \in \mathcal{D}$ *be compatible with the fragment decomposition. The x-th impurity Hamiltonian* $\widehat{H}_{x,D}^{\mathrm{imp}}$ *is the operator on* $\mathrm{Fock}(W_{x,D})$ *given by*

$$\widehat{H}_{x,D}^{\mathrm{imp}} = E_x^{\mathrm{env}}(D) + \sum_{i,j=1}^{2L_x} \left[C^x(D)^T (h + J(\mathfrak{D}^x(D)) - K(\mathfrak{D}^x(D))) C^x(D) \right]_{ij} \widehat{a}_i(D)^\dagger \widehat{a}_j(D)$$
$$+ \frac{1}{2} \sum_{i,j,k,\ell=1}^{2L_x} [V^x(D)]_{ijkl} \widehat{a}_i(D)^\dagger \widehat{a}_j(D)^\dagger \widehat{a}_\ell(D) \widehat{a}_k(D), \tag{30}$$

where

- *the Coulomb and exchange matrices* $J(\mathfrak{D}^x(D)) \in \mathbb{R}^{L \times L}$ *and* $K(\mathfrak{D}^x(D)) \in \mathbb{R}^{L \times L}$ *for the x-th impurity are constructed from the density matrix*

$$\mathfrak{D}^x(D) := D - D E_x (E_x^T D E_x)^{-1} E_x^T D \in \mathrm{Gr}(N - L_x, L); \tag{31}$$

- *the rank-4 tensor* $V^x(D)$ *is given by*

$$[V^x(D)]_{ijkl} := \sum_{\kappa,\lambda,\nu,\xi=1}^{L} V_{\kappa\lambda\nu\xi} [C^x(D)]_{\kappa i} [C^x(D)]_{\lambda j} [C^x(D)]_{\nu k} [C^x(D)]_{\xi \ell}; \tag{32}$$

- *the value of the (irrelevant) constant* $E_x^{\mathrm{env}}(D)$ *is given in (39).*

Note that the matrix $\mathfrak{D}^x(D)$ is in fact the one-body density matrix associated with the Slater determinant $\Psi^{0,\mathrm{core}}_{x,D}$ (see Section 2.2).

5.2 | Domain of the high-level map

A matrix $D \in \mathcal{D}$ is in the domain of the high-level map F^{HL} formally defined in Section 2.3 if and only if

1. D is compatible with the fragment decomposition (see Lemma 7), in such a way that the impurity problem (15) is well defined for each x;
2. the set

$$M_D := \left\{ \mu \in \mathbb{R} \;\middle|\; \forall x, \text{ the impurity problem (15) has a unique ground-state 1-RDM } P_{x,D,\mu}, \right.$$
$$\left. \text{and } \sum_{x=1}^{N_f} \mathrm{Tr}\big(\Pi_x P_{x,D,\mu} \Pi_x\big) = N \right\}$$

   is non-empty;
3. the function

$$\mathcal{F}_D : M_D \ni \mu \mapsto \sum_{x=1}^{N_f} \Pi_x P_{x,D,\mu} \Pi_x \in \mathcal{P}$$

   is a constant over M_D, which we denote by $F^{\mathrm{HL}}(D)$.

In the proof of Theorem 4, we will study F^{HL}_α in the non-interacting ($\alpha = 0$) and weakly interacting ($|\alpha|$ small) cases. We will see that in these regimes the domain of F^{HL}_α contains a neighborhood of D_0 in $\mathcal{D}$.

6 | *N*-REPRESENTABILITY AND LOW-LEVEL MAP

In this section, we focus our study on the low level map defined in (19). Clearly, (19) has minimizers if and only if $P \in \mathrm{Bd}(\mathcal{D})$ (otherwise, the feasible set of the minimization problem is empty).

The next Lemma covers the extreme cases of minimal ($N_f = 2$) and maximal ($N_f = L$) numbers of fragments.

Lemma 9 (Global N-representability).

1. *If $N_f = L$ (one site per fragment), then* $\mathrm{Bd}(\mathcal{D}) = \mathrm{Bd}(\mathrm{CH}(\mathcal{D})) = \mathcal{P}$.
2. *If $N_f = 2$ and $L \geq 3$, then* $\mathrm{Bd}(\mathcal{D}) \subsetneq \mathrm{Bd}(\mathrm{CH}(\mathcal{D})) = \mathcal{P}$. *More precisely,*

$$\mathrm{Bd}(\mathcal{D}) = \big\{ P \in \mathcal{P} \;\big|\; \forall 0 < n < 1,\ \dim(\mathrm{Ker}(\Pi_1 P \Pi_1 - n)) = \dim(\mathrm{Ker}(\Pi_2 P \Pi_2 - (1-n)) \big\}.$$

Our analysis of the DMET method in the non-interacting and weakly perturbative settings relies on the following weaker N-representability result.

Definition 10 (Local N-representability). Let $D \in \mathcal{D}$ be compatible with the fragment decomposition. We say that the local N-representability condition is satisfied at D if the linear map Bd is surjective from $T_D\mathcal{D}$ to $\mathcal{Y}$.

Note that Assumption (A3) can be rephrased as: the local N-representability condition is satisfied at D_0.

A necessary condition for the local N-representability condition to be satisfied at some $D \in \mathrm{Bd}^{-1}\mathring{\mathcal{P}}$ is that

$$N(L-N) = \dim(\mathcal{D}) = \dim(\mathbb{R}^{N_v \times N}) \geq \dim(\mathcal{Y}) = \sum_{x=1}^{N_f} \frac{L_x(L_x+1)}{2} - 1. \tag{33}$$

If $N_f = L$ (one site per fragment), the above condition reads $N(L-N) \geq L-1$, and is therefore satisfied for any $1 \leq N \leq L-1$, that is for any non-trivial case. On the other hand, if $N_f = 2$ and $L = 2L_1 = 2L_2$ (two fragments of identical sizes), the necessary condition reads $N(L-N) \geq \frac{L(L+2)}{4} - 1$ and is never satisfied as soon as $L \geq 3$. This result is in agreement with the global N-representability results in Lemma 9. In usual DMET calculations, condition (33) is always satisfied, so that, generically, $\mathcal{P}$ and $\mathrm{Bd}(\mathcal{D})$ coincide in the neighborhood of P_0.

The next lemma provides a sufficient local N-representability criterion.

Lemma 11 (A local N-representability criterion). *Let $D \in \mathcal{D}$ be compatible with the fragment decomposition (i.e., $D \in \mathrm{Bd}^{-1}\mathring{\mathcal{P}}$). The following assertions are equivalent:*

1. *the local N-representability condition is satisfied at D;*
2. *the only matrices $M \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$ commuting with both D and the matrices Π_x for all $1 \leq x \leq N_f$ are of the form $M = \lambda I_L$ for some $\lambda \in \mathbb{R}$;*
3. *if $\Phi \in \mathbb{R}^{L\times L}$ is an orthogonal matrix such that*

$$D = \Phi \begin{pmatrix} I_N & 0 \\ 0 & 0 \end{pmatrix} \Phi^T, \tag{34}$$

then the linear map

$$\mathbb{R}^{(L-N)\times N} \ni X \mapsto \sum_{x=1}^{N_f} \Pi_x \Phi \begin{pmatrix} 0 & X^T \\ X & 0 \end{pmatrix} \Phi^T \Pi_x \in \mathcal{Y} \tag{35}$$

is surjective.

The third assertion of Lemma 11 gives a practical way to check the local N-representability criterion: it suffices to (i) diagonalize D in order to write it as in (34) (the columns of $\Phi \in O(L)$ form an orthonormal basis of eigenvectors of D), (ii) assemble the matrix of the linear map (35) in the canonical bases of $\mathbb{R}^{(L-N)\times N}$ and $\mathcal{Y}$, and (iii) check whether the number of positive singular values of this matrix is equal to $\dim(\mathcal{Y}) = \sum_{x=1}^{N_f} \frac{L_x(L_x+1)}{2} - 1$.

7 | PROOFS

7.1 | Proof of Lemma 7

Let $D \in \mathcal{D}$.

2) $\iff$ 3). Assume that

$$\forall 1 \leq x \leq N_f, \quad \dim(DX_x) = \dim((1-D)X_x) = L_x.$$

Since $D^2 = D$, we have for all $y \in \mathbb{R}^{L_x}$,

$$y^T(E_x^T D E_x)y = y^T(E_x^T D^2 E_x)y = (D(E_x y))^T(D(E_x y)) = |D(E_x y)|^2, \tag{36}$$

and therefore,

$$0 \leq y^T(E_x^T D E_x)y = |D(E_x y)|^2 \leq |E_x y|^2 = |y|^2.$$

Thus $0 \leq E_x^T D E_x \leq 1$ in the sense of hermitian matrices. Assume now that $y^T(E_x^T D E_x)y = 0$. Then, $E_x y \in \mathrm{Ker}(D)$. But we also have $E_x y \in X_x$. Since $\dim(DX_x) = L_x$, this implies that $y = 0$. Thus $0 < E_x^T D E_x$ in the sense of hermitian matrices. Likewise, we have $E_x^T D E_x < 1$. This proves that 2) $\Rightarrow$ 3). Conversely, if for all $1 \leq x \leq N_f$, $0 < E_x^T D E_x$, we infer from (36) that $D(E_x y) = 0$ implies $y = 0$, hence that $\dim(DX_x) = L_x$. Likewise, $E_x^T D E_x < 1$ implies $\dim((1-D)X_x) = L_x$. Therefore, 3) $\Rightarrow$ 2).

3) $\iff$ 4). Since $0 < E_x^T D E_x$ is equivalent to $E_x^T D E_x \in \mathrm{GL}_{\mathbb{R}}(L_x)$ and $E_x^T D E_x < 1$ is equivalent to $E_x^T(1-D)E_x \in \mathrm{GL}_{\mathbb{R}}(L_x)$, we conclude that 3) $\iff$ 4).

Lastly, it follows from the definition of $\mathcal{P}$ that

$$P = \begin{pmatrix} P_1 & 0 & \cdots & 0 \\ 0 & P_2 & \cdots & 0 \\ \vdots & & \ddots & \vdots \\ 0 & 0 & \cdots & P_{N_f} \end{pmatrix} \in \overset{\circ}{\mathcal{P}} \quad \iff \quad (\forall 1 \leq x \leq N_f,\ 0 < P_x = E_x^T P E_x < 1). \tag{37}$$

This shows that 1) $\iff$ 3), which concludes the proof.

7.2 | Proof of Proposition 8

Let $D \in \mathcal{D}$ and $1 \leq x \leq N_f$. Let us first concatenate the matrix $C^x(D) \in \mathbb{R}^{L \times 2L_x}$ introduced in (28) with a matrix $C^x_{\mathrm{env}}(D) \in \mathbb{R}^{L \times (L-2L_x)}$ in order to form an orthogonal matrix

$$\mathfrak{C}^x(D) = (C^x(D)|C^x_{\mathrm{env}}(D)) \in O(L).$$

The column vectors of $\mathfrak{C}^x(D)$ define an orthonormal basis of $\mathcal{H} = \mathbb{R}^L$ adapted to the decomposition $\mathcal{H} = W_{x,D} \oplus \mathcal{H}^{\mathrm{env}}_{x,D}$. The generators of the real CAR algebra associated with this basis are

given by

$$\widehat{a}_i^x(D) = \sum_{\kappa=1}^{L} \mathfrak{C}^x(D)_{\kappa i} \widehat{a}_\kappa, \qquad \widehat{a}_i^x(D)^\dagger = \sum_{\kappa=1}^{L} \mathfrak{C}^x(D)_{\kappa i} \widehat{a}_\kappa^\dagger,$$

so that the Hamiltonian

$$\widehat{H} = \sum_{\kappa,\lambda=1}^{L} h_{\kappa\lambda} \widehat{a}_\kappa^\dagger \widehat{a}_\lambda + \frac{1}{2} \sum_{\kappa,\lambda,\nu,\xi=1}^{L} V_{\kappa\lambda\nu\xi} \widehat{a}_\kappa^\dagger \widehat{a}_\lambda^\dagger \widehat{a}_\xi \widehat{a}_\nu$$

can be rewritten as

$$\widehat{H} = \sum_{i,j=1}^{L} [h^x(D)]_{ij} \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) + \frac{1}{2} \sum_{i,j,k,l=1}^{L} [V^x(D)]_{ijkl}\, \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D)^\dagger \widehat{a}_l^x(D) \widehat{a}_k^x(D)$$

with

$$[h^x(D)]_{ij} := \sum_{\kappa,\lambda=1}^{L} h_{\kappa\lambda} \mathfrak{C}^x(D)_{\kappa i} \mathfrak{C}^x(D)_{\lambda j} \quad \text{that is} \quad h^x(D) = \mathfrak{C}^x(D)^T h \mathfrak{C}^x(D)$$

and

$$[V^x(D)]_{ijkl} := \sum_{\kappa,\lambda,\nu,\xi=1}^{L} V_{\kappa\lambda\nu\xi} \mathfrak{C}^x(D)_{\kappa i} \mathfrak{C}^x(D)_{\lambda j} \mathfrak{C}^x(D)_{\nu k} \mathfrak{C}^x(D)_{\xi l}.$$

Note that if $1 \le i, j, k, l \le 2L_x$,

$$[h^x(D)]_{ij} = [C^x(D)^T h C^x(D)]_{ij} \quad \text{and} \quad [V^x(D)]_{ijkl} := \sum_{\kappa,\lambda,\nu,\xi=1}^{L} V_{\kappa\lambda\nu\xi} C^x(D)_{\kappa i} C^x(D)_{\lambda j} C^x(D)_{\nu k} C^x(D)_{\xi l},$$

in agreement with (32). Let $\Psi \in \mathrm{Fock}(\mathcal{H})$ be of the form

$$\Psi = \Psi_{x,D}^{\mathrm{imp}} \wedge \Psi_{x,D}^{0,\mathrm{core}} \quad \text{with} \quad \Psi_{x,D}^{\mathrm{imp}} \in \mathrm{Fock}(W_{x,D}) \quad \text{and} \quad \Psi_{x,D}^{0,\mathrm{core}} \in \bigwedge^{(N-L_x)} \mathcal{H}_{x,D}^{\mathrm{core}}.$$

We have

$$\langle \Psi | \widehat{H} | \Psi \rangle = \Big\langle \Psi_{x,D}^{\mathrm{imp}} \wedge \Psi_{x,D}^{0,\mathrm{core}} \Big| \sum_{i,j=1}^{L} [h^x(D)]_{ij} \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) + \frac{1}{2} \sum_{i,j,k,l=1}^{L} [V^x(D)]_{ijkl} \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D)^\dagger \widehat{a}_l^x(D) \widehat{a}_k^x(D) \Big| \Psi_{x,D}^{\mathrm{imp}} \wedge \Psi_{x,D}^{0,\mathrm{core}} \Big\rangle.$$

The terms in the Hamiltonian which change the number of particles in the impurity space or the environment do not contribute. The terms which act only on the environment subspace yield a term proportional to $\|\Psi_{x,D}^{\text{imp}}\|^2$. Expanding the above expression, we thus obtain

$$\langle\Psi|\widehat{H}|\Psi\rangle = a_1 + a_2 + a_3 + a_4 + a_5 + a_6 + a_7$$

with

$$\begin{aligned}
a_1 :&= \sum_{i,j=1}^{2L_x} [h^x(D)]_{ij} \langle \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} | \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) | \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} \rangle \\
&= \sum_{i,j=1}^{2L_x} [h^x(D)]_{ij} \langle \Psi_{x,D}^{\text{imp}} | \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) | \Psi_{x,D}^{\text{imp}} \rangle \\
&= \sum_{i,j=1}^{2L_x} [C^x(D)^T h C^x(D)]_{ij} \langle \Psi_{x,D}^{\text{imp}} | \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) | \Psi_{x,D}^{\text{imp}} \rangle,
\end{aligned}$$

$$\begin{aligned}
a_2 :&= \sum_{i,j=2L_x+1}^{L} [h^x(D)]_{ij} \langle \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} | \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) | \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} \rangle \\
&= \left(\sum_{i,j=2L_x+1}^{L} [h^x(D)]_{ij} \langle \Psi_{x,D}^{0,\text{core}} | \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) | \Psi_{x,D}^{0,\text{core}} \rangle \right) \|\Psi_{x,D}^{\text{imp}}\|^2,
\end{aligned}$$

$$\begin{aligned}
a_3 :&= \sum_{i=2L_x+1}^{L} \sum_{j=1}^{2L_x} [h^x(D)]_{ij} \langle \underbrace{\Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}}}_{\substack{L_x \text{ part. in imp.} \\ (N-L_x) \text{ part. in env.}}} | \underbrace{\widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) | \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} \rangle}_{\substack{(L_x+1) \text{ part. in imp.} \\ (N-L_x-1) \text{ part. in env.}}} \\
&= 0,
\end{aligned}$$

$$\begin{aligned}
a_4 :&= \sum_{i=1}^{2L_x} \sum_{j=2L_x+1}^{L} [h^x(D)]_{ij} \langle \underbrace{\Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}}}_{\substack{L_x \text{ part. in imp.} \\ (N-L_x) \text{ part. in env.}}} | \underbrace{\widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D) | \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} \rangle}_{\substack{(L_x-1) \text{ part. in imp.} \\ (N-L_x+1) \text{ part. in env.}}} \\
&= 0,
\end{aligned}$$

$$\begin{aligned}
a_5 :&= \frac{1}{2} \sum_{i,j,k,l=1}^{2L_x} [V^x(D)]_{ijkl} \langle \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} | \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D)^\dagger \widehat{a}_l^x(D) \widehat{a}_k^x(D) | \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} \rangle \\
&= \frac{1}{2} \sum_{i,j,k,l=1}^{2L_x} [V^x(D)]_{ijkl} \langle \Psi_{x,D}^{\text{imp}} | \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D)^\dagger \widehat{a}_l^x(D) \widehat{a}_k^x(D) | \Psi_{x,D}^{\text{imp}} \rangle,
\end{aligned}$$

$$a_6 := \frac{1}{2} \sum_{i,j,k,l=2L_x+1}^{L} [V^x(D)]_{ijkl} \langle \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} | \widehat{a}_i^x(D)^\dagger \widehat{a}_j^x(D)^\dagger \widehat{a}_l^x(D) \widehat{a}_k^x(D) | \Psi_{x,D}^{\text{imp}} \wedge \Psi_{x,D}^{0,\text{core}} \rangle$$

$$= \left(\frac{1}{2} \sum_{i,j,k,l=2L_x+1}^{L} [V^x(D)]_{ijkl} \langle \Psi^{0,\mathrm{core}}_{x,D} | \widehat{a}^x_i(D)^\dagger \widehat{a}^x_j(D)^\dagger \widehat{a}^x_l(D) \widehat{a}^x_k(D) | \Psi^{0,\mathrm{core}}_{x,D} \rangle \right) \| \Psi^{\mathrm{imp}}_{x,D} \|^2,$$

$$a_7 := \frac{1}{2} \sum_{i,k=1}^{2L_x} \sum_{j,l=2L_x+1}^{L} ([V^x(D)]_{ijkl} - [V^x(D)]_{ijlk} - [V^x(D)]_{jikl} + [V^x(D)]_{jilk}) \times \underbrace{\langle \Psi^{\mathrm{imp}}_{x,D} \wedge \Psi^{0,\mathrm{core}}_{x,D} | \widehat{a}^x_i(D)^\dagger \widehat{a}^x_j(D)^\dagger \widehat{a}^x_l(D) \widehat{a}^x_k(D) | \Psi^{\mathrm{imp}}_{x,D} \wedge \Psi^{0,\mathrm{core}}_{x,D} \rangle}_{\langle \Psi^{\mathrm{imp}}_{x,D} | \widehat{a}^x_i(D)^\dagger \widehat{a}^x_k(D) | \Psi^{\mathrm{imp}}_{x,D} \rangle \langle \Psi^{0,\mathrm{core}}_{x,D} | \widehat{a}^x_j(D)^\dagger \widehat{a}^x_l(D) | \Psi^{0,\mathrm{core}}_{x,D} \rangle}.$$

Noticing that

$$\forall 2L_x + 1 \le j, l \le L, \quad \langle \Psi^{0,\mathrm{core}}_{x,D} | \widehat{a}^x_j(D)^\dagger \widehat{a}^x_l(D) | \Psi^{0,\mathrm{core}}_{x,D} \rangle = (\mathfrak{C}^x(D)^T D \mathfrak{C}^x(D))_{jl} \tag{38}$$

we get

$$a_7 = \frac{1}{2} \sum_{i,k=1}^{2L_x} \sum_{j,l=2L_x+1}^{L} ([V^x(D)]_{ijkl} - [V^x(D)]_{ijlk} - [V^x(D)]_{jikl} + [V^x(D)]_{jilk}) \times (\mathfrak{C}^x(D)^T D \mathfrak{C}^x(D))_{jl} \langle \Psi^{\mathrm{imp}}_{x,D} | \widehat{a}^x_i(D)^\dagger \widehat{a}^x_k(D) | \Psi^{\mathrm{imp}}_{x,D} \rangle$$

$$= \sum_{i,j=1}^{2L_x} \left(\sum_{k,l=2L_x+1}^{L} ([V^x(D)]_{ikjl} - [V^x(D)]_{iklj} - [V^x(D)]_{kijl} + [V^x(D)]_{kilj}) (\mathfrak{C}^x(D)^T D \mathfrak{C}^x(D))_{kl} \right) \times \langle \Psi^{\mathrm{imp}}_{x,D} | \widehat{a}^x_i(D)^\dagger \widehat{a}^x_j(D) | \Psi^{\mathrm{imp}}_{x,D} \rangle.$$

It holds for all $1 \le i, j \le 2L_x$,

$$\sum_{k,l=2L_x+1}^{L} [V^x(D)]_{ikjl} (\mathfrak{C}^x(D)^T D \mathfrak{C}^x(D))_{kl}$$

$$= \sum_{k,l=2L_x+1}^{L} \sum_{\kappa,\lambda,\nu,\xi,\sigma,\tau=1}^{L} V_{\kappa\lambda\nu\xi} [\mathfrak{C}^x(D)]_{\kappa i} [\mathfrak{C}^x(D)]_{\lambda k} [\mathfrak{C}^x(D)]_{\nu j} [\mathfrak{C}^x(D))]_{\xi l} [\mathfrak{C}^x(D)]_{\sigma k} D_{\sigma\tau} [\mathfrak{C}^x(D)]_{\tau l}$$

$$= \sum_{k,l=1}^{L-2L_x} \sum_{\kappa,\lambda,\nu,\xi,\sigma,\tau=1}^{L} V_{\kappa\lambda\nu\xi} [C^x(D)]_{\kappa i} [C^x_{\mathrm{env}}(D)]_{\lambda k} [C^x(D)]_{\nu j} [C^x_{\mathrm{env}}(D))]_{\xi l} [C^x_{\mathrm{env}}(D)]_{\sigma k} D_{\sigma\tau} [C^x_{\mathrm{env}}(D)]_{\tau l}$$

$$= \sum_{\kappa,\nu=1}^{L} [C^x(D)]_{\kappa i} \left(\sum_{\lambda,\xi,\sigma,\tau=1}^{L} V_{\kappa\lambda\nu\xi} \left(\sum_{k=1}^{L-2L_x} [C^x_{\mathrm{env}}(D)]_{\lambda k} [C^x_{\mathrm{env}}(D)]_{\sigma k} \right) \times D_{\sigma\tau} \left(\sum_{l=1}^{L-2L_x} [C^x_{\mathrm{env}}(D)]_{\tau l} [C^x_{\mathrm{env}}(D)]_{\xi l} \right) \right) [C^x(D)]_{\nu j}$$

$$= \sum_{\kappa,\nu=1}^{L} [C^x(D)]_{\kappa i} \left(\sum_{\lambda,\xi,\sigma,\tau=1}^{L} V_{\kappa\lambda\nu\xi} \left(C^x_{\mathrm{env}}(D) C^x_{\mathrm{env}}(D)^T D C^x_{\mathrm{env}}(D) C^x_{\mathrm{env}}(D)^T \right)_{\lambda\xi} \right) [C^x(D)]_{\nu j}$$

$$= \left[C^x(D)^T J(\widetilde{\mathfrak{D}}^x(D)) C^x(D) \right]_{ij},$$

with, recalling that $\mathfrak{C}^x(D) = (C^x(D)|C^x_{\mathrm{env}}(D))$ is an orthogonal matrix,

$$\begin{aligned}\widetilde{\mathfrak{D}}^x(D) :&= C^x_{\mathrm{env}}(D)C^x_{\mathrm{env}}(D)^T D C^x_{\mathrm{env}}(D)C^x_{\mathrm{env}}(D)^T \\ &= (1 - C^x(D)C^x(D)^T)D(1 - C^x(D)C^x(D)^T) \\ &= D - DE_x(E_x^T DE_x)^{-1}E_x^T D \\ &= \mathfrak{D}^x(D) \qquad \text{(see (31))}.\end{aligned}$$

Using similar arguments, we get

$$a_7 = \sum_{i,k=1}^{2L_x} \left[C^x(D)^T(J(\mathfrak{D}^x(D)) - K(\mathfrak{D}^x(D)))C^x(D)\right]_{ij} \langle \Psi^{\mathrm{imp}}_{x,D}|\hat{a}^x_i(D)^\dagger \hat{a}^x_j(D)|\Psi^{\mathrm{imp}}_{x,D}\rangle.$$

We finally obtain

$$\langle \Psi|\widehat{H}|\Psi\rangle = \langle \Psi^{\mathrm{imp}}_{x,D}|\widehat{H}^{\mathrm{imp}}_{x,D}|\Psi^{\mathrm{imp}}_{x,D}\rangle,$$

where $\widehat{H}^{\mathrm{imp}}_{x,D}$ is given by (30) with

$$\begin{aligned}E^{\mathrm{env}}(D) = &\sum_{i,j=2L_x+1}^{L} [h^x(D)]_{ij} \langle \Psi^{0,\mathrm{core}}_{x,D}|\hat{a}^x_i(D)^\dagger \hat{a}^x_j(D)|\Psi^{0,\mathrm{core}}_{x,D}\rangle \\ &+ \frac{1}{2}\sum_{i,j,k,l=2L_x+1}^{L} [V^x(D)]_{ijkl} \langle \Psi^{0,\mathrm{core}}_{x,D}|\hat{a}^x_i(D)^\dagger \hat{a}^x_j(D)^\dagger \hat{a}^x_l(D)\hat{a}^x_k(D)|\Psi^{0,\mathrm{core}}_{x,D}\rangle. \qquad (39)\end{aligned}$$

7.3 | Proof of Lemma 9

The first assertion is a direct consequence of [21, Theorem 6].

We now prove the second assertion. Let

$$\mathcal{K} := \left\{P = (P_1, P_2) \in \mathbb{R}^{L_1\times L_1}_{\mathrm{sym}} \times \mathbb{R}^{L_2\times L_2}_{\mathrm{sym}} \;\middle|\; \forall 0 < n < 1,\ \dim(\mathrm{Ker}(P_1 - n)) = \dim(\mathrm{Ker}(P_2 - (1-n))\right\}.$$

Let $P = (P_1, P_2) \in \mathrm{Bd}(\mathcal{D})$ and $D \in \mathcal{D}$ be such that $\mathrm{Bd}(D) = P$. Let U_1 and U_2 be two orthogonal matrices of sizes $(L_1 \times L_1)$ and $(L_2 \times L_2)$ respectively, and $D_1 = \mathrm{diag}(m_1, \dots, m_{L_1})$ and $D_2 = \mathrm{diag}(m'_1, \dots, m'_{L_2})$ two diagonal matrices with entries in the range [0,1] ranked such that $m_1 \geq \cdots \geq m_{L_1}$ and $m'_1 \leq \cdots \leq m'_{L_2}$, such that $P_1 = U_1 D_1 U_1^T$ and $P_2 = U_2 D_2 U_2^T$. It holds

$$D = \begin{pmatrix} U_1 & 0 \\ 0 & U_2 \end{pmatrix} \begin{pmatrix} D_1 & C \\ C^T & D_2 \end{pmatrix} \begin{pmatrix} U_1^T & 0 \\ 0 & U_2^T \end{pmatrix} \quad \text{for some } C \in \mathbb{R}^{L_1\times L_2}.$$

The condition $D^2 = D$ reads

$$CC^T = D_1 - D_1^2, \quad C^T C = D_2 - D_2^2, \quad C - D_1 C - C D_2 = 0,$$

that is

$$\forall 1 \le i \le L_1, \quad \forall 1 \le j \le L_2, \quad \sum_{k=1}^{L_2} C_{ik}^2 = m_i - m_i^2, \quad \sum_{k=1}^{L_1} C_{kj}^2 = m'_j - {m'_j}^2, \quad (1 - m_i - m'_j) C_{ij} = 0.$$

This implies that $C_{ij} = 0$ unless $m'_j = 1 - m_j$ and that $C_{ij} = 0$ whenever $m_i = 0$ or 1, or $m'_j = 0$ or 1. It follows that

$$\begin{pmatrix} U_1^T & 0 \\ 0 & U_2^T \end{pmatrix} D \begin{pmatrix} U_1 & 0 \\ 0 & U_2 \end{pmatrix}$$

$$= \left(\begin{array}{ccc|ccc} I_{r_1} & & & & & \\ & n_1 I_{d_1} & & & C_1 & \\ & & \ddots & & & \ddots \\ & & & n_\ell I_{d_\ell} & & & C_\ell \\ & & & & 0_{s_1} & & \\ \hline & & & & 0_{s_2} & & \\ & C_1^T & & & (1-n_1) I_{d'_1} & & \\ & & \ddots & & & \ddots & \\ & & & C_\ell^T & & & (1-n_\ell) I_{d'_\ell} \\ & & & & & & & I_{r_2} \end{array}\right), \tag{40}$$

with $0 < n_\ell < \cdots < n_1 < 1$. Using again the idempotency of D, we obtain the relations $C_j C_j^T = n_j(1-n_j) I_{d_j}$ and $C_j^T C_j = n_j(1-n_j) I_{d'_j}$. Taking the trace leads to $d_j = d'_j$. Therefore, $P \in \mathcal{K}$ so that $\mathrm{Bd}(\mathcal{D}) \subset \mathcal{K}$.

Conversely, let $P \in \mathcal{K}$ and U_1, U_2, D_1, D_2 as before. Then $U_1^T P_1 U_1$ and $U_2^T P_2 U_2$ read as the diagonal blocks of the right-hand side of (40) with $d_j = d'_j$ for all j. Setting $C_j = \sqrt{n_j(1-n_j)} I_{d_j}$, the matrix D defined by (40) is in $\mathcal{M}_S$ and satisfies $\mathrm{Bd}(D) = P$. Hence, $P \in \mathrm{Bd}(\mathcal{D})$ and therefore $\mathcal{K} \subset \mathrm{Bd}(D)$.

7.4 | Proof of Lemma 11

Let $N_v := L - N$. For $X \in \mathbb{R}^{N_v \times N}$ such that $\|X\| < 1/2$, we set

$$f_\Phi(X) := \Phi \begin{pmatrix} \frac{1}{2}\left(I_N + (I_N - 4X^T X)^{1/2}\right) & X^T \\ X & \frac{1}{2}\left(I_{N_v} - (I_{N_v} - 4XX^T)^{1/2}\right) \end{pmatrix} \Phi^T,$$

$$g_\Phi(X) := \mathrm{Bd}(f_\Phi(X)).$$

The map f_Φ provides a local system of coordinates of $\mathcal{D}$ in the vicinity of D. Therefore, the local N-representability condition is satisfied at D if and only if the map

$$d_0 g_\Phi : \mathbb{R}^{N_\mathrm{v} \times N} \ni X \mapsto d_0 g_\Phi = \sum_{x=1}^{N_f} \Pi_x \Phi \begin{pmatrix} 0 & X^T \\ X & 0 \end{pmatrix} \Phi^T \Pi_x \in \mathcal{Y}$$

is surjective. This proves the equivalence between the first and third assertions of the lemma.

Writing Φ as $\Phi = (\Phi^{\mathrm{occ}} | \Phi^{\mathrm{virt}})$ with $\Phi^{\mathrm{occ}} \in \mathbb{R}^{L \times N}$ and $\Phi^{\mathrm{virt}} \in \mathbb{R}^{L \times N_\mathrm{v}}$, the adjoint of $d_0 g_\Phi$ is given by

$$d_0 g_\Phi^* : \mathcal{Y} \ni Y \mapsto d_0 g_\Phi^*(Y) = 2 {\Phi^{\mathrm{virt}}}^T Y \Phi^{\mathrm{occ}} \in \mathbb{R}^{N_\mathrm{v} \times N}.$$

We therefore have for all $Y \in \mathcal{Y}$,

$$(d_0 g_\Phi d_0 g_\Phi^*) Y = 2 \sum_{x=1}^{N_f} \Pi_x ((1-D)YD + DY(1-D)) \Pi_x, \tag{41}$$

and therefore

$$\begin{aligned} \| d_0 g_\Phi^*(Y) \|^2 &= \mathrm{Tr}\big(Y (d_0 g_\Phi d_0 g_\Phi^*)(Y) \big) \big) = 2\mathrm{Tr}\left(Y \sum_{x=1}^{N_f} \Pi_x ((1-D)YD + DY(1-D)) \Pi_x \right) \\ &= 2\mathrm{Tr}\left(\sum_{x=1}^{N_f} \Pi_x Y \Pi_x ((1-D)YD + DY(1-D)) \right) \\ &= 2\mathrm{Tr}(Y((1-D)YD + DY(1-D))) = 4 \| (1-D)YD \|^2. \end{aligned}$$

Thus

$$\forall Y \in \mathcal{Y}, \quad \| d_0 g_\Phi^*(Y) \| = 2 \| (1-D)YD \|.$$

The map $d_0 g_\Phi$ is surjective if and only if its adjoint is injective. Thus the criterion is satisfied if and only if

$$\forall Y \in \mathcal{Y}, \quad (1-D)YD = 0 \quad \Rightarrow \quad Y = 0.$$

As D is an orthogonal projector, $(1-D)YD = 0$ if and only if Y commutes with D. In addition, a matrix $Y \in \mathbb{R}^{L \times L}_{\mathrm{sym}}$ is in $\mathcal{Y}$ if and only if (i) it commutes with all the Π_x's, and (ii) its trace is equal to 0. Thus, the criterion is satisfied if and only if any zero trace matrix $Y \in \mathbb{R}^{L \times L}_{\mathrm{sym}}$ commuting with D and the Π_x's is the null matrix. Lastly, this condition is equivalent to: any matrix $Y \in \mathbb{R}^{L \times L}_{\mathrm{sym}}$ commuting with D and the Π_x's is of the form λI_L for some $\lambda \in \mathbb{R}$. This completes the proof of the second statement.

7.5 | Proof of Proposition 1

For $\alpha = 0$, the low-level map is formally given by

$$F_0^{\mathrm{LL}}(P) = \operatorname*{argmin}_{D\in\mathcal{D},\ \mathrm{Bd}(D)=P} \mathrm{Tr}(hD) \qquad \text{(formal)}. \tag{42}$$

Under Assumption (A1) (i.e., $\varepsilon_N < 0 < \varepsilon_{N+1}$), D_0 is the unique minimizer of

$$\operatorname*{argmin}_{D\in\mathcal{D}} \mathrm{Tr}(hD).$$

Since $\mathrm{Bd}(D_0) = P_0$ (by definition of P_0), D_0 is the unique minimizer of (42) for $P = P_0$. Thus, P_0 is in the domain of F_0^{LL} and $F_0^{\mathrm{LL}}(P_0) = D_0$.

For $\alpha = 0$, the high-level map takes the simple formal expression

$$F_0^{\mathrm{HL}}(D) = \sum_{x=1}^{N_f} \Pi_x C^x(D) \mathbb{1}_{(-\infty,0]}\big(C^x(D)^T(h-\mu\Pi_x)C^x(D)\big)C^x(D)^T\Pi_x \qquad \text{(formal)},$$

where $C^x(D)$ is defined in (28) and $\mu \in \mathbb{R}$ is such that

$$\sum_{x=1}^{N_f} \mathrm{Tr}\big(\Pi_x C^x(D) \mathbb{1}_{(-\infty,0]}\big(C^x(D)^T(h-\mu\Pi_x)C^x(D)\big)C^x(D)^T\Pi_x\big) = N.$$

Therefore, a matrix $D \in \mathcal{D}$ is in the domain of F_0^{HL} if and only if

1. the set

$$M_D := \left\{ \mu \in \mathbb{R} \;\middle|\; \sum_{x=1}^{N_f} \mathrm{Tr}\big(\Pi_x C^x(D) \mathbb{1}_{(-\infty,0]}\big(C^x(D)^T(h-\mu\Pi_x)C^x(D)\big)C^x(D)^T\Pi_x\big) = N \right\}$$

   is non-empty;
2. the function

$$\mathcal{F}_D : M_D \ni \mu \mapsto \sum_{x=1}^{N_f} \Pi_x C^x(D) \mathbb{1}_{(-\infty,0]}\big(C^x(D)^T(h-\mu\Pi_x)C^x(D)\big)C^x(D)^T\Pi_x \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$$

   is constant over M_D. Its value is an element of $\mathcal{P}$, which we denote by $F_0^{\mathrm{HL}}(D)$.

Let us prove that under Assumptions (A1) and (A2), D_0 belongs to the domain of F_0^{HL} and $F_0^{\mathrm{HL}}(D_0) = P_0$.

First, we observe that for each $1 \le x \le N_f$, the space $W_{x,0} := X_x + D_0 X_x$ is D_0-invariant since D_0 is a projector. The linear operator D_0 on $\mathbb{R}^L$ therefore has a a block-diagonal operator representation in the decomposition $W_{x,0} \oplus W_{x,0}^{\perp}$ of $\mathcal{H} = \mathbb{R}^L$:

$$D_0 \equiv \begin{pmatrix} D_0^x & 0 \\ 0 & \widetilde{D_0^x} \end{pmatrix} \qquad \text{(in the decomposition } \mathcal{H} = W_{x,0} \oplus W_{x,0}^{\perp}),$$

where D_0^x and $\widetilde{D_0^x}$ are both orthogonal projectors. The corresponding representation of h is not necessarily block-diagonal:

$$h \equiv \begin{pmatrix} h^x & h^x_{\mathrm{OD}} \\ {h^x_{\mathrm{OD}}}^T & \widetilde{h^x} \end{pmatrix} \qquad \text{(in the decomposition } \mathcal{H} = W_{x,0} \oplus W_{x,0}^{\perp}\text{)}.$$

Let us now focus on the operator h^x. To lighten the notation, we set

$$D_{0,x} := E_x^T D_0 E_x.$$

We infer from Assumption (A2) and Lemma 7 that $\dim(D_0 X_x) = \dim((1-D_0)X_x) = L_x$ and

$$C_0^x := C^x(D_0) = \left(D_0 E_x D_{0,x}^{-1/2} | (1-D_0) E_x (1-D_{0,x})^{-1/2} \right)$$

forms an orthonormal basis of $W_{x,0}$. In this basis, the operator h^x is represented by the matrix

$$\mathfrak{h}^x := {C_0^x}^T h C_0^x = \begin{pmatrix} \mathfrak{h}^x_- & 0 \\ 0 & \mathfrak{h}^x_+ \end{pmatrix}, \tag{43}$$

with

$$\mathfrak{h}^x_- := D_{0,x}^{-1/2} E_x^T D_0 h D_0 E_x D_{0,x}^{-1/2}, \tag{44}$$

$$\mathfrak{h}^x_+ := (1-D_{0,x})^{-1/2} E_x^T (1-D_0) h (1-D_0) E_x (1-D_{0,x})^{-1/2}. \tag{45}$$

The zeros in the off-diagonal blocks of $\mathfrak{h}^x$ come from the fact that $D_0 h (1-D_0) = (1-D_0) h D_0 = 0$ since h and D_0 commute. In addition, we have

$$\varepsilon_1 D_0 \le D_0 h D_0 = \sum_{i=1}^{N} \varepsilon_i \phi_i \phi_i^T \le \varepsilon_N D_0, \tag{46}$$

$$\varepsilon_{N+1}(1-D_0) \le (1-D_0) h (1-D_0) = \sum_{a=N+1}^{L} \varepsilon_a \phi_a \phi_a^T \le \varepsilon_L (1-D_0). \tag{47}$$

Combining (44) and (46) on the one hand, and (45) and (47) on the other hand, we obtain

$$\varepsilon_1 I_{L_x} \le \mathfrak{h}^x_- \le \varepsilon_N I_{L_x} \qquad \text{and} \qquad \varepsilon_{N+1} I_{L_x} \le \mathfrak{h}^x_+ \le \varepsilon_L I_{L_x}. \tag{48}$$

We therefore have

$$\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x) = \mathbb{1}_{(-\infty,0)}(\mathfrak{h}^x) = \begin{pmatrix} I_{L_x} & 0 \\ 0 & 0 \end{pmatrix}, \qquad \mathbb{1}_{[0,\infty)}(\mathfrak{h}^x) = \mathbb{1}_{(0,\infty)}(\mathfrak{h}^x) = \begin{pmatrix} 0 & 0 \\ 0 & I_{L_x} \end{pmatrix}, \tag{49}$$

and thus

$$\sum_{r=1}^{N_f} \Pi_x C_0^x \mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x) {C_0^x}^T \Pi_x = \sum_{r=1}^{N_f} \Pi_x C_0^x \begin{pmatrix} I_{L_x} & 0 \\ 0 & 0 \end{pmatrix} {C_0^x}^T \Pi_x$$

$$= \sum_{r=1}^{N_f} (E_x E_x^T) D_0 E_x D_{0,x}^{-1} E_x^T D_0 (E_x E_x^T)$$

$$= \sum_{r=1}^{N_f} \Pi_x D_0 \Pi_x = \mathrm{Bd}(D_0) = P_0.$$

As $\mathrm{Tr}(P_0) = N$, we have $0 \in M_{D_0}$ and $\mathcal{F}_{D_0}(0) = P_0$. Let us now show that $M_{D_0} = \{0\}$. It holds

$$\Pi_x \equiv \begin{pmatrix} \Pi_x^x & 0 \\ 0 & 0 \end{pmatrix} \qquad \text{(in the decomposition } \mathcal{H} = W_{x,0} \oplus W_{x,0}^\perp\text{)},$$

and in the basis defined of $W_{x,0}$ defined by C_0^x, the orthogonal projector Π_x^x is represented by the matrix

$$\mathfrak{p}^x := {C_0^x}^T \Pi_x C_0^x = \begin{pmatrix} D_{0,x} & D_{0,x}^{1/2}(1 - D_{0,x})^{1/2} \\ (1 - D_{0,x})^{1/2} D_{0,x}^{1/2} & (1 - D_{0,x}) \end{pmatrix}. \tag{50}$$

We therefore have in particular $\mathfrak{p}^{x\,2} = \mathfrak{p}^x = \mathfrak{p}^{x\,T}$. Consider the function

$$\mathbb{R} \ni \mu \mapsto \zeta(\mu) := \sum_{x=1}^{N_f} \mathrm{Tr}\Big(\Pi_x C_x^0 \mathbb{1}_{(-\infty,0]}\Big({C_x^0}^T (h - \mu \Pi_x) C_x^0 \Big) {C_x^0}^T \Pi_x \Big)$$

$$= \sum_{x=1}^{N_f} \mathrm{Tr}\big(\mathfrak{p}^x \mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x - \mu \mathfrak{p}^x) \big)$$

$$= \sum_{x=1}^{N_f} \mathrm{Tr}\big(\mathfrak{p}^x \mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x - \mu \mathfrak{p}^x) \mathfrak{p}^x \big) \geq 0.$$

We already know that $\zeta(0) = N$. We see from (48) that 0 is not in the spectrum of $\mathfrak{h}$ for all x. By a simple continuity argument, we obtain that for $|\mu|$ small enough, 0 is not in the spectrum of $\mathfrak{h}^x - \mu \mathfrak{p}^x$ for all x. We therefore have

$$\zeta(\mu) = \sum_{x=1}^{N_f} \frac{1}{2\pi i} \oint_C \mathrm{Tr}\Big(\mathfrak{p}^x (z - (\mathfrak{h}^x - \mu \mathfrak{p}^x))^{-1} \Big)\, dz \qquad \text{(for } |\mu| \text{ small enough)}, \tag{51}$$

where C is for example, a circle in the complex plane, centered on the negative real axis, containing 0 and of large enough radius. It follows that ζ is analytic in the vicinity of 0 and that

$$\zeta'(0) = -\sum_{x=1}^{N_f} \frac{1}{2\pi i} \oint_C \mathrm{Tr}\Big(\mathfrak{p}^x (z - \mathfrak{h}^x)^{-1} \mathfrak{p}^x (z - \mathfrak{h}^x)^{-1} \Big)\, dz = \sum_{x=1}^{N_f} \langle \mathfrak{p}^x, \mathfrak{L}_x^+ \mathfrak{p}^x \rangle, \tag{52}$$

where $\mathfrak{L}_x^+$ is the linear operator on $\mathbb{R}_{\mathrm{sym}}^{2L_x \times 2L_x}$ defined by

$$\forall M \in \mathbb{R}_{\mathrm{sym}}^{2L_x \times 2L_x}, \quad \mathfrak{L}_x^+ M = -\frac{1}{2\pi i} \oint_C (z - \mathfrak{h}^x)^{-1} M (z - \mathfrak{h}^x)^{-1}\, dz, \tag{53}$$

which can alternatively be defined by the linear response formula

$$\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x + M) = \mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x) - \mathfrak{L}_x^+ M + o(\|M\|). \tag{54}$$

Let us diagonalize the real symmetric matrix $\mathfrak{h}^x$ as

$$\mathfrak{h}^x = \sum_{n=1}^{2L_x} \widetilde{\varepsilon}_{x,n} \widetilde{\phi}_{x,n} \widetilde{\phi}_{x,n}^T \quad \text{with} \quad \widetilde{\varepsilon}_{x,1} \le \cdots \le \widetilde{\varepsilon}_{x,2L_x}, \quad \widetilde{\phi}_{x,m}^T \widetilde{\phi}_{x,n} = \delta_{mn},$$

with (using (48))

$$\forall 1 \le i \le L_x, \quad \forall L_x \le a \le 2L_x, \quad \widetilde{\varepsilon}_{x,i} \le \varepsilon_N < 0 < \varepsilon_{N+1} \le \widetilde{\varepsilon}_{x,a}.$$

Using Cauchy residue formula, we get

$$\forall M = \begin{pmatrix} M^{--} & M^{+-T} \\ M^{+-} & M_{++} \end{pmatrix} \in \mathbb{R}^{2L_x \times 2L_x}_{\mathrm{sym}}, \quad \mathfrak{L}_x^+ M = \begin{pmatrix} 0 & N(M^{+-})^T \\ N(M^{+-}) & 0 \end{pmatrix} \tag{55}$$

with

$$\forall 1 \le m, n \le L_x, \quad [N(M^{+-})]_{mn} = \frac{[M^{+-}]_{mn}}{\widetilde{\varepsilon}_{x,m+L_x} - \widetilde{\varepsilon}_{x,n}}. \tag{56}$$

The operator $\mathfrak{L}_x^+$ is self-adjoint and positive. Denoting by $\rho := \varepsilon_L - \varepsilon_1 > 0$ the spectral diameter of h, we have

$$\forall M = \begin{pmatrix} M^{--} & M^{-+} \\ M^{+-} & M_{++} \end{pmatrix} \in \mathbb{R}^{2L_x \times 2L_x}_{\mathrm{sym}}, \quad \langle M, \mathfrak{L}_x^+ M \rangle \ge 2\rho^{-1} \|M^{-+}\|^2. \tag{57}$$

Indeed, we have

$$\begin{aligned} \langle M, \mathfrak{L}_x^+ M \rangle = 2 \sum_{x=1}^{N_f} \sum_{i=1}^{L_x} \sum_{a=L_x+1}^{2L_x} \frac{|\widetilde{\phi}_{x,i}^T M \widetilde{\phi}_{x,a}|^2}{\widetilde{\varepsilon}_{x,a} - \widetilde{\varepsilon}_{x,i}} &\ge 2\rho^{-1} \sum_{x=1}^{N_f} \sum_{i=1}^{L_x} \sum_{a=L_x+1}^{2L_x} |\widetilde{\phi}_{x,i}^T M \widetilde{\phi}_{x,a}|^2 \\ &= 2\rho^{-1} \| \mathbb{1}_{(-\infty,0)}(\mathfrak{h}^x) M \mathbb{1}_{(0,+\infty)}(\mathfrak{h}^x) \|^2 = 2\rho^{-1} \|M^{-+}\|^2. \end{aligned}$$

Let

$$\mathcal{J}_0 := \left\{ \mu \in \mathbb{R} \;\middle|\; \prod_{x=1}^{N_f} \det(\mathfrak{h}^x - \mu \mathfrak{p}^x) = 0 \right\}.$$

Since $\mu \mapsto \det(\mathfrak{h}^x - \mu \mathfrak{p}^x)$ is a polynomial of degree L_x, the set $\mathcal{J}_0$ contains at most L elements. By similar arguments as above, the function ζ is real-analytic and non-decreasing on each connected components of $\mathbb{R} \setminus \mathcal{J}_0$. At each $\mu_0 \in \mathcal{J}_0$, the jump of ζ is given by

$$\zeta(\mu_0 + 0) - \zeta(\mu_0 - 0) = \sum_{x=1}^{N_f} \mathrm{Tr}\big(\mathfrak{p}^x \mathbb{1}_{\{0\}}(\mathfrak{h}^x - \mu_0 \mathfrak{p}^x) \mathfrak{p}^x\big) \ge 0.$$

The function ζ is therefore nondecreasing on $\mathbb{R}$. As a consequence, the set M_{D_0} is an interval I_{D_0} containing 0. Using (50), (52), and (57), we get

$$\zeta'(0) \geq 2\rho^{-1} \sum_{x=1}^{N_f} \|D_{0,x}^{1/2}(1-D_{0,x})^{1/2}\|^2 = 2\rho^{-1} \sum_{x=1}^{N_f} \mathrm{Tr}(D_{0,x}(1-D_{0,x})) > 0,$$

since, in view of Lemma 7, all the eigenvalues of the symmetric matrix $D_{0,x}(1-D_{0,x})$ are positive. Thus $M_{D_0} = \{0\}$. This proves that D_0 is in the domain of F_0^{HL} and that $F^{\mathrm{HL}}(D_0) = P_0$.

Combining this result with the previously established relation $F_0^{\mathrm{LL}}(P_0) = D_0$, we obtain that P_0 is a fixed point of the DMET map for $\alpha = 0$.

7.6 | Proof of Theorem 4

We endow $\mathcal{D}$ with the Riemannian metric induced by the Frobenius inner product on $\mathbb{R}^{L\times L}_{\mathrm{sym}}$. For $\eta > 0$, we set

$$\omega_\eta := \{P \in \mathcal{P} \mid \|P - P_0\| < \eta\} \quad \text{and} \quad \Omega_\eta := \{D \in \mathcal{D} \mid \|D - D_0\| < \eta\}.$$

7.6.1 | Low-level map in the perturbative regime

Let us introduce the maps

$$\begin{aligned}
&g : \mathcal{D} \to \mathcal{Y} && \text{s.t.} \quad \forall D \in \mathcal{D}, \quad g(D) := \mathrm{Bd}(D) - P_0,\\
&a : \mathcal{D} \to \mathbb{R} && \text{s.t.} \quad \forall D \in \mathcal{D}, \quad a(D) := \mathrm{Tr}(hD),\\
&b : \mathcal{D} \to \mathbb{R} && \text{s.t.} \quad \forall D \in \mathcal{D}, \quad b(D) := \frac{1}{2}\mathrm{Tr}((J(D) - K(D))D),\\
&E : \mathbb{R} \times \mathcal{D} \to \mathbb{R} && \text{s.t.} \quad \forall (\alpha, D) \in \mathbb{R} \times \mathcal{D}, \quad E(\alpha, D) := \mathcal{E}_\alpha^{\mathrm{HF}}(D) = a(D) + \alpha b(D).
\end{aligned}$$

Since the maps $\mathrm{Bd}, J, K : \mathbb{R}^{L\times L}_{\mathrm{sym}} \to \mathbb{R}^{L\times L}_{\mathrm{sym}}$ are linear, the maps g, a, b, and E are real-analytic. With this notation, we have

$$(\text{Assumption (A3)}) \iff \big(B := d_{D_0} g = \mathrm{Bd} : T_{D_0}\mathcal{D} \to \mathcal{Y} \text{ surjective}\big).$$

Lemma 12 (Low-level map in the perturbative regime). *Under Assumptions (A1)–(A3), there exists* $\alpha_{\mathrm{LL}} > 0$ *and* $0 < \eta_{\mathrm{LL}} < \frac{1}{2}$ *such that*

1. $\omega_{\eta_{\mathrm{LL}}} \subset \mathrm{Dom}(F_\alpha^{\mathrm{LL}})$ *for all* $\alpha \in (-\alpha_{\mathrm{LL}}, \alpha_{\mathrm{LL}})$*;*
2. *the function* $(\alpha, P) \mapsto F_\alpha^{\mathrm{LL}}(P)$ *is real-analytic on* $(-\alpha_{\mathrm{LL}}, \alpha_{\mathrm{LL}}) \times \omega^{\mathcal{Y}}_{\eta_{\mathrm{LL}}}$.

Proof. The first assertion means that for all $(\alpha, P) \in (-\alpha_{\mathrm{LL}}, \alpha_{\mathrm{LL}}) \times \omega_{\eta_{\mathrm{LL}}}$, the problem

$$\min_{D\in\mathcal{D} \mid \mathrm{Bd}(D)=P} \mathcal{E}_\alpha^{\mathrm{HF}}(D) = \min_{D\in\mathcal{D} \mid g(D)=P-P_0} E(\alpha, D) \tag{58}$$

has a unique minimizer, which we denote by $F_\alpha^{\mathrm{LL}}(P)$.

Using Lemma 11 and the submersion theorem, we deduce from Assumptions (A2)–(A3) that there exists $\eta > 0$ and $C \in \mathbb{R}_+$ such that for all $P \in \omega_\eta$, the set $\mathrm{Bd}^{-1}(P)$ is nonempty and there exists $D_P \in \mathrm{Bd}^{-1}(P)$ such that $\|D_P - D_0\| \leq C\|P - P_0\|$. Let $D_{\alpha,P}$ be a minimizer of $\mathcal{E}_\alpha^{\mathrm{HF}}$ on $\mathrm{Bd}^{-1}(P)$. Such a minimizer exists since $\mathcal{E}_\alpha^{\mathrm{HF}}$ is continuous on $\mathcal{D}$ and $\mathrm{Bd}^{-1}(P)$ is a nonempty compact subset of $\mathcal{D}$, and satisfies the optimality conditions

$$\nabla_D E(\alpha, D_{\alpha,P}) + d_{D_{\alpha,P}} g^* \Lambda_{\alpha,P} = 0, \quad g(D_{\alpha,P}) = P - P_0, \tag{59}$$

where $\nabla_D E(\alpha, D_{\alpha,P}) \in T_{D_{\alpha,P}}\mathcal{D}$ is the gradient at $D_{\alpha,P}$ of the function $\mathcal{D} \ni D \to E(\alpha, D) \in \mathbb{R}$ for the Riemannian metric induced with the Frobenius inner product, and $\Lambda_{\alpha,P} \in \mathcal{Y}$ the Lagrange multiplier of the constraint $g(D_{\alpha,P}) = P - P_0$.

Denoting by

$$C_{\mathrm{nl}} := \frac{1}{2} \max_{D \in \mathcal{D}} |\mathrm{Tr}((J(D) - K(D))D)|,$$

we have

$$\mathcal{E}_\alpha^{\mathrm{HF}}(D_{\alpha,P}) \leq \mathcal{E}_\alpha^{\mathrm{HF}}(D_P) \leq \mathcal{E}_0^{\mathrm{HF}}(D_P) + \alpha C_{\mathrm{nl}} \leq \mathcal{E}_0^{\mathrm{HF}}(D_0) + \|h\|\|P - P_0\| + \alpha C_{\mathrm{nl}}. \tag{60}$$

To obtain a lower bound of $\mathcal{E}_\alpha^{\mathrm{HF}}(D_{\alpha,P})$, we use that

$$\forall D \in \mathcal{D}, \quad \mathcal{E}_0^{\mathrm{HF}}(D) = \mathrm{Tr}(hD) \geq \mathcal{E}_0^{\mathrm{HF}}(D_0) + \frac{\gamma}{2}\|D - D_0\|^2.$$

This inequality is classical, but we recall its proof for the sake of completeness. For $M \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$ we set

$$M^{--} := D_0 M D_0, \quad M^{-+} := D_0 M (1 - D_0), \quad M^{+-} := (1 - D_0) M D_0, \quad M^{++} := (1 - D_0) M (1 - D_0).$$

Let $D \in \mathcal{D}$ and $Q := D - D_0$. Since $D_0 = \mathbb{1}_{(-\infty,0]}(h)$, we have

$$h^{-+} = h^{+-} = 0, \quad h^{--} \leq \varepsilon_N, \quad h^{++} \geq \varepsilon_{N+1}, \quad Q^{++} \geq 0, \quad Q^{--} \leq 0,$$

and we deduce from the fact that both D and D_0 are rank-N orthogonal projectors that

$$Q^2 = Q^{++} - Q^{--} \quad \text{and} \quad \mathrm{Tr}(Q^{++}) + \mathrm{Tr}(Q^{--}) = 0.$$

Combining all the above properties, we obtain

$$\begin{aligned}
\forall D \in \mathcal{D}, \quad a(D) &= \mathrm{Tr}(hD) \\
&= \mathrm{Tr}(hD_0) + \mathrm{Tr}(h(D - D_0)) \\
&= a(D_0) + \mathrm{Tr}\left(h^{++}Q^{++}\right) + \mathrm{Tr}(h^{--}Q^{--}) \\
&\geq a(D_0) + \varepsilon_{N+1}\mathrm{Tr}\left(Q^{++}\right) + \varepsilon_N \mathrm{Tr}(Q^{--}) \\
&= a(D_0) + \frac{\gamma}{2}\mathrm{Tr}\left(Q^{++} - Q^{--}\right) \\
&= a(D_0) + \frac{\gamma}{2}\|D - D_0\|^2.
\end{aligned} \tag{61}$$

As $\mathcal{E}_0^{\mathrm{HF}}(D) = a(D)$, this implies that

$$\mathcal{E}_\alpha^{\mathrm{HF}}(D_{\alpha,P}) \geq \mathcal{E}_0^{\mathrm{HF}}(D_{\alpha,P}) - \alpha C_{\mathrm{nl}} \geq \mathcal{E}_0^{\mathrm{HF}}(D_0) + \frac{\gamma}{2}\|D_{\alpha,P} - D_0\|^2 - \alpha C_{\mathrm{nl}}.$$

Combining this result with (60), we obtain

$$\|D_{\alpha,P} - D_0\|^2 \leq 2\gamma^{-1}(2\alpha C_{\mathrm{nl}} + \|h\|P - P_0\|).$$

This implies in particular that for $|\alpha|$ and $\|P - P_0\|$ small enough, any minimizer $D_{\alpha,P}$ of (58) is close to D_0. To conclude, it suffices to prove that for $|\alpha|$ and $\|P - P_0\|$ small enough, (59) has a unique critical point close to D_0. This leads us to introduce the function

$$\Theta : (\mathbb{R} \times \mathcal{P}) \times (\mathcal{D} \times \mathcal{Y}) \ni ((\alpha, P), (D, \Lambda)) \mapsto \Theta((\alpha, P), (D, \Lambda)) \in T_D\mathcal{D} \times \mathcal{Y}$$

defined by

$$\Theta((\alpha, P), (D, \Lambda)) := (\nabla_D E(\alpha, D) + (d_D g)^* \Lambda, g(D) - (P - P_0)).$$

As D_0 is the unique minimizer of $D \mapsto E(0, D)$ on $\mathcal{D}$ and $P_0 = \mathrm{Bd}(D_0)$, we have $\nabla_D E(0, D_0) = 0$ and $g(D_0) = 0$, so that

$$\Theta((0, P_0), (D_0, 0)) = (0, 0).$$

In addition, denoting by

$$A := D_D^2 a(D_0) : T_{D_0}\mathcal{D} \to T_{D_0}\mathcal{D} \tag{62}$$

the Hessian at D_0 of the function a for the Riemannian metric induced by the Frobenius inner product, we have

$$\forall (Q, \Lambda) \in T_{D_0}\mathcal{D} \times \mathcal{Y}, \quad \left[d_{D,\Lambda}\Theta((0, P_0), (D_0, 0))\right] \begin{pmatrix} Q \\ \Lambda \end{pmatrix} = \begin{pmatrix} A & B^* \\ B & 0 \end{pmatrix} \begin{pmatrix} Q \\ \Lambda \end{pmatrix},$$

where we recall that $B := d_{D_0} g$. In view of (61), we have

$$\forall Q \in T_{D_0}\mathcal{D}, \quad \langle Q, AQ \rangle \geq \gamma \|Q\|^2. \tag{63}$$

Since A is coercive and $B : T_{D_0}\mathcal{D} \to \mathcal{Y}$ is surjective, it follows from the Schur complement formula that the map

$$d_{D,\Lambda}\Theta((0, P_0), (D_0, 0)) : T_{D_0}\mathcal{D} \times \mathcal{Y} \to T_{D_0}\mathcal{D} \times \mathcal{Y}$$

is invertible. It follows from the real-analytic implicit function theorem on manifolds that there exist $\alpha_{\mathrm{LL}} > 0$ and $\eta > 0$, such that for all $(\alpha, P) \in (-\alpha_{\mathrm{LL}}, \alpha_{\mathrm{LL}}) \times \omega_\eta$, (59) has a unique solution $(D_{\alpha,P}, \Lambda_{\alpha,P})$ with $D_{\alpha,P} \in \omega_\eta$ and the map $(\alpha, P) \mapsto D_{\alpha,P}$ is real-analytic on $(-\alpha_{\mathrm{LL}}, \alpha_{\mathrm{LL}}) \times \omega_\eta$. □

7.6.2 | High-level map in the perturbative regime

The following result states that the high-level map $(\alpha, D) \mapsto F_\alpha^{\mathrm{HL}}(D)$ is well-defined and real-analytic on a neighborhood of $(0, D_0)$.

Lemma 13 (High-level map in the perturbative regime). *Under Assumptions (A1)-(A2), there exists* $\alpha_{\mathrm{HL}} > 0$ *and* $0 < \eta_{\mathrm{HL}} < \frac{1}{2}$ *such that*

1. $\Omega_{\eta_{\mathrm{HL}}} \subset \mathrm{Dom}(F^{\mathrm{HL}}_{\alpha})$ *for all* $\alpha \in (-\alpha_{\mathrm{HL}}, \alpha_{\mathrm{HL}})$*;*
2. *the function* $(\alpha, D) \mapsto F^{\mathrm{HL}}_{\alpha}(D)$ *is real-analytic on* $(-\alpha_{\mathrm{HL}}, \alpha_{\mathrm{HL}}) \times \Omega_{\eta_{\mathrm{HL}}}$.

Proof. For $D \in \mathcal{D}$ compatible with the fragment decomposition, we set

$$[\widetilde{h}_x(D)]_{\kappa\lambda} := \left[\widetilde{C}^x(D)^T h \widetilde{C}^x(D)\right]_{\kappa\lambda} = \sum_{\kappa'\lambda'=1}^{L} [\widetilde{C}^x(D)]_{\kappa,\kappa'} [\widetilde{C}^x(D)]_{\lambda,\lambda'} h_{\kappa'\lambda'}, \tag{64}$$

$$[\widetilde{V}_x(D)]_{\kappa\lambda\nu\xi} := \sum_{\kappa'\lambda'\nu'\xi'=1}^{L} [\widetilde{C}^x(D)]_{\kappa,\kappa'} [\widetilde{C}^x(D)]_{\lambda,\lambda'} [\widetilde{C}^x(D)]_{\nu,\nu'} [\widetilde{C}^x(D)]_{\xi,\xi'} V_{\kappa'\lambda'\nu'\xi'}, \tag{65}$$

where $\widetilde{C}^x(D)$ is defined in Lemma 7. Denoting by c_κ, $c_\kappa^\dagger$, $1 \le \kappa \le 2L_x$ the generators of the CAR algebra on $\mathrm{Fock}(\mathbb{R}^{2L_x})$ associated with the canonical basis of $\mathbb{R}^{2L_x}$, the high-level map can be formally written as

$$F^{\mathrm{HL}}_{\alpha}(D) = \sum_{x=1}^{N_f} \sum_{\kappa,\lambda=1}^{L_x} e_{L'_x+\kappa} \mathrm{Tr}_{\mathrm{Fock}(\mathbb{R}^{2N_x})} \left(\Gamma_{\alpha,x,D,\mu} c_\kappa^\dagger c_\lambda \right) e^T_{L'_x+\lambda} \quad \text{(formal)}, \tag{66}$$

where $\Gamma_{\alpha,x,D,\mu} \in \mathcal{L}(\mathrm{Fock}(\mathbb{R}^{2L_x}))$ is the ground-state (many-body) density matrix associated with the grand-canonical impurity Hamiltonian

$$\widetilde{H}^{\mathrm{imp}}_{\alpha,x,D,\mu} := \sum_{\kappa,\lambda=1}^{2L_x} [\widetilde{h}_x(D)]_{\kappa\lambda} c_\kappa^\dagger c_\lambda + \alpha \sum_{\kappa,\lambda,\nu,\xi=1}^{2L_x} [\widetilde{V}_x(D)]_{\kappa\lambda\nu\xi} c_\kappa^\dagger c_\lambda^\dagger c_\xi c_\nu - \mu \sum_{\kappa=1}^{L_x} c_\kappa^\dagger c_\kappa,$$

the parameter $\mu \in \mathbb{R}$ being chosen such that

$$\sum_{x=1}^{N_f} \sum_{\kappa,\lambda=1}^{L_x} \mathrm{Tr}_{\mathrm{Fock}(\mathbb{R}^{2N_x})} \left(\Gamma_{\alpha,x,D,\mu} c_\kappa^\dagger c_\lambda \right) = N.$$

The results established in the proof of Proposition 1 can be rephrased as follows: under Assumptions (A1)–(A2),

1. the impurity Hamiltonian $\widetilde{H}^{\mathrm{imp}}_{0,x,D_0,0}$ has a non-degenerate ground-state for each x and that it holds

$$\sum_{x=1}^{N_f} \sum_{\kappa,\lambda=1}^{L_x} \mathrm{Tr}_{\mathrm{Fock}(\mathbb{R}^{2N_x})} \left(\Gamma_{0,x,D_0,0} c_\kappa^\dagger c_\lambda \right) = N;$$

2. the function

$$\mathbb{R} \ni \mu \mapsto \sum_{x=1}^{N_f} \sum_{\kappa,\lambda=1}^{L_x} \mathrm{Tr}_{\mathrm{Fock}(\mathbb{R}^{2N_x})} \left(\Gamma_{0,x,D_0,\mu} c_\kappa^\dagger c_\lambda \right) \in \mathbb{R}$$

is non-decreasing, real-analytic in the neighborhood of $\mu = 0$, and its derivative at $\mu = 0$ is positive.

Since the maps

$$\mathcal{D} \ni D \mapsto [\widetilde{h}_x(D)]_{\kappa\lambda} \in \mathbb{R} \quad \text{and} \quad \mathcal{D} \ni D \mapsto [\widetilde{V}_x(D)]_{\kappa\lambda\nu\xi} \in \mathbb{R}$$

are real-analytic in the neighborhood of D_0, we deduce from Kato's analytic perturbation theory and the implicit function theorem that there exists $\alpha_{\rm HL} > 0$, $\eta_{\rm HL} > 0$, and $\mu_{\rm HL} > 0$ such that

1. for each $(\alpha, D, \mu) \in (-\alpha_{\rm HL}, \alpha_{\rm HL}) \times \Omega_{\eta_{HL}} \times (-\mu_{\rm HL}, \mu_{\rm HL})$, the impurity Hamiltonian $H^{\rm imp}_{\alpha,x,D,\mu}$ has a non-degenerate ground-state for each x; we denote by $\Gamma_{\alpha,x,D,\mu(\alpha,X)}$ the corresponding ground-state many-body density matrix;
2. for each $(\alpha, D) \in (-\alpha_{\rm HL}, \alpha_{\rm HL}) \times \Omega_{\eta_{HL}}$, there exists a unique $\mu(\alpha, D) \in (-\mu_{\rm HL}, \mu_{\rm HL})$ such that

$$\sum_{x=1}^{N_f} \sum_{\kappa,\lambda=1}^{L_x} \mathrm{Tr}_{\mathrm{Fock}(\mathbb{R}^{2N_x})}\left(\Gamma_{\alpha,x,D,\mu(\alpha,D)} c^\dagger_\kappa c_\lambda\right) = N;$$

3. the maps $(\alpha, D) \mapsto \mu(\alpha, D)$, $(\alpha, D) \mapsto \Gamma_{\alpha,x,D,\mu(\alpha,D)}$, and

$$(\alpha, D) \mapsto F^{\rm HL}_\alpha(D) := \left(\sum_{x=1}^{N_f} \sum_{\kappa,\lambda=1}^{L_x} e_{L'_x+\kappa} \mathrm{Tr}_{\mathrm{Fock}(\mathbb{R}^{2N_x})}\left(\Gamma_{\alpha,x,D,\mu(\alpha,D)} c^\dagger_\kappa c_\lambda\right) e^T_{L'_x+\lambda}\right)$$

are real-analytic on $(-\alpha_{\rm HL}, \alpha_{\rm HL}) \times \Omega_{\eta_{\rm HL}}$.

This proves the two assertions of Lemma 13. □

7.6.3 | Existence, uniqueness, and analyticity

We infer from Lemma 12 and Lemma 13 that there exist $\alpha_{\rm DMET} > 0$ and $\eta_{\rm DMET} > 0$ such that the function

$$(-\alpha_{\rm DMET}, \alpha_{\rm DMET}) \times \omega_{\eta_{\rm DMET}} \ni (\alpha, P) \mapsto \Phi(\alpha, P) := F^{\rm DMET}_\alpha(P) - P := F^{\rm HL}_\alpha(F^{\rm LL}_\alpha(P)) - P \in \mathcal{Y}$$

is well-defined and real-analytic, and we know from Proposition 1 that

$$\Phi(0, P_0) = 0.$$

To complete the proof of Theorem 4, we have to check that the function Φ satisfies all the hypotheses of the implicit function theorem, namely that

$$d_P\Phi(0, P_0) = (d_{D_0}F^{\rm HL}_0)(d_{P_0}F^{\rm LL}_0) - I_{\mathcal{Y}} : \mathcal{Y} \to \mathcal{Y} \tag{67}$$

is invertible.

Let us first compute $d_{P_0}F^{\rm LL}_0 : \mathcal{Y} \to T_{D_0}\mathcal{D}$. Differentiating the equality

$$\forall P \in \omega_\eta, \quad \Theta((0, P), (F^{\rm LL}_0(P), \Lambda_{0,P})) = (0, 0),$$

we obtain that the derivatives at P_0 of the functions $\omega_\eta \ni P \mapsto F_0^{\mathrm{LL}}(P) \in \mathcal{D}$ and $\omega_\eta \ni P \mapsto \lambda(P) := \Lambda_{0,P} \in \mathcal{Y}$ are characterized by the relation

$$\forall Y \in \mathcal{Y}, \quad \underbrace{[d_P\Theta((0,P_0),(D_0,0))]Y}_{=(0,-Y)} + \underbrace{[d_{(D,\Lambda)}\Theta((0,P_0),(D_0,0))]((d_{P_0}F_0^{\mathrm{LL}})Y,(d_{P_0}\lambda)Y)}_{=(A[(d_{P_0}F_0^{\mathrm{LL}})Y]+B^*(d_{P_0}\lambda)Y),B[(d_{P_0}F_0^{\mathrm{LL}})Y]} = 0,$$

from which we infer that

$$d_{P_0}F_0^{\mathrm{LL}} = A^{-1}B^*(BA^{-1}B^*)^{-1}. \tag{68}$$

Let us now compute $d_{D_0}F_0^{\mathrm{HL}} : T_{D_0}\mathcal{D} \to \mathcal{Y}$. We have

$$\forall D \in \Omega_{\eta_{\mathrm{HL}}}, \quad F_0^{\mathrm{HL}}(D) = \sum_{x=1}^{N_f} \Pi_x C^x(D) \mathbb{1}_{(-\infty,0]}\big(C^x(D)^T(h-\mu(0,D)\Pi_x)C^x(D)\big)C^x(D)^T\Pi_x,$$

where the function

$$\mathcal{D} \ni D \mapsto C^x(D) = (\underbrace{DE_x(E_x^TDE_x)^{-1/2}}_{C_-^x(D)} \,|\, \underbrace{(1-D)E_x(E_x^T(1-D)E_x)^{-1/2}}_{C_+^x(D)}) \in \mathbb{R}^{L\times(2L_x)}$$

has been introduced in (28). Setting as previously $C_0^x := C^x(D_0)$, and denoting by $M(Q) := [d_{D_0}C^x](Q)$ and $\ell(Q) := [d_D\mu(0,D_0)](Q)$, we get

$$\begin{aligned} d_{D_0}F^{\mathrm{HL}}(Q) = &\sum_{x=1}^{N_f} \Pi_x\Big(M(Q)\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x){C_0^x}^T + C_0^x\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x)M(Q)^T\Big)\Pi_x \\ &- \sum_{x=1}^{N_f} \Pi_x C_0^x\mathfrak{L}_x^+\Big(M(Q)^ThC_0^x + {C_0^x}^ThM(Q) - \ell(Q)\mathfrak{p}^x\Big){C_0^x}^T\Pi_x. \end{aligned}$$

Using (49), we obtain

$$\begin{aligned} M(Q)\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x){C_0^x}^T + C_0^x\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x)M(Q)^T &= [d_{D_0}C_-^x(Q)][C_-^x(D_0)]^T + C_-^x(D_0)[d_{D_0}C_-^x(Q)]^T \\ &= d_{D_0}[C_-^x{C_-^x}^T](Q). \end{aligned}$$

This implies that

$$\Pi_x\Big(M(Q)\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x){C_0^x}^T + C_0^x\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x)M(Q)^T\Big)\Pi_x = d_{D_0}[\Pi_xC_-^x{C_-^x}^T\Pi_x](Q).$$

Since

$$\Pi_xC_-^x(D){C_-^x(D)}^T\Pi_x = (E_xE_x^T)(DE_x(E_x^TDE_x)^{-1/2})((E_x^TDE_x)^{-1/2}E_x^TD)(E_xE_x^T) = \Pi_xD\Pi_x,$$

we get $d_{D_0}[\Pi_xC_-^x{C_-^x}^T\Pi_x](Q) = \Pi_xQ\Pi_x$ and therefore

$$\sum_{x=1}^{N_f} \Pi_x\Big(M(Q)\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x){C_0^x}^T + C_0^x\mathbb{1}_{(-\infty,0]}(\mathfrak{h}^x)M(Q)^T\Big)\Pi_x = \mathrm{Bd}(Q) = BQ.$$

Next, observing that for all $Q \in T_{D_0}\mathcal{D}$,

$$d_{D_0}C^x_-(Q) = D_0E_xS_-(Q) + QE_x(E_x^TD_0E_x)^{-1/2},$$

$$d_{D_0}C^x_+(Q) = (1-D_0)E_xS_+(Q) - QE_x(E_x^T(1-D_0)E_x)^{-1/2},$$

with $Q \mapsto S_\pm(Q) \in \mathbb{R}^{L_x\times L_x}$ linear and

$$Q = D_0Q(1-D_0) + (1-D_0)QD_0, \tag{69}$$

we obtain that

$$M(Q)^T hC_0^x + C_0^{xT} hM(Q) = \begin{pmatrix} * & N(Q)^T \\ N(Q) & * \end{pmatrix}$$

with

$$\begin{aligned} N(Q) &:= (E_x^T(1-D_0)E_x)^{-1/2}E_x^T((1-D_0)hQ - QhD_0)E_x(E_x^TD_0E_x)^{-1/2} \\ &= (E_x^T(1-D_0)E_x)^{-1/2}E_x^T(1-D_0)[h,Q]D_0E_x(E_x^TD_0E_x)^{-1/2}. \end{aligned}$$

We thus have

$$M(Q)^T hC_0^x + C_0^{xT} hM(Q) = \begin{pmatrix} * & 0 \\ 0 & * \end{pmatrix} - C_0^{xT}[D_0,[h,Q]]C_0^x,$$

which implies, using (55),

$$\mathfrak{L}_x^+\Big(M(Q)^T hC_0^x + C_0^{xT} hM(Q) - \ell(Q)\mathfrak{p}^x\Big) = \mathfrak{L}_x^+\Big(-C_0^{xT}[D_0,[h,Q]]C_0^x - \ell(Q)\mathfrak{p}^x\Big).$$

We therefore obtain

$$d_{D_0}F_0^{\mathrm{HL}} = B + L,$$

with $L : T_{D_0}\mathcal{D} \to \mathcal{Y}$ given by

$$\forall Q \in T_{D_0}\mathcal{D}, \quad LQ := \sum_{x=1}^{N_f} \Pi_x C_0^x \mathfrak{L}_x^+\Big(C_0^{xT}[D_0,[h,Q]]C_0^x + \ell(Q)\mathfrak{p}^x\Big)C_0^{xT}\Pi_x. \tag{70}$$

Combining with (68), and setting

$$R := LA^{-1}B^* : \mathcal{Y} \to \mathcal{Y}, \tag{71}$$

we obtain

$$d_P\Phi(0,P_0) = (B+L)(A^{-1}B^*(BA^{-1}B^*)^{-1}) - I_{\mathcal{Y}} = R(BA^{-1}B^*)^{-1}.$$

To conclude, we just have to show that the map R rigorously defined by (71) actually coincides with the 4-point response function formally defined by (26) (the latter is bijective by Assumption (A4)). We have for all $Q \in T_{D_0}\mathcal{D}$ and $Y \in \mathcal{Y}$,

$$\begin{aligned} \langle Q, B^*Y\rangle_{T_{D_0}\mathcal{D}} &= \langle BQ, Y\rangle_{\mathcal{Y}} = \mathrm{Tr}((BQ)Y) \\ &= \mathrm{Tr}\left(\left(\sum_{x=1}^{N_f}\Pi_xQ\Pi_x\right)Y\right) = \sum_{x=1}^{N_f}\mathrm{Tr}(\Pi_xQ\Pi_xY) = \sum_{x=1}^{N_f}\mathrm{Tr}(Q\Pi_xY\Pi_x) \end{aligned}$$

$$= \mathrm{Tr}\left(Q\left(\sum_{x=1}^{N_f}\Pi_x Y \Pi_x\right)\right) = \mathrm{Tr}(QY) = \mathrm{Tr}(Q(\underbrace{D_0Y(1-D_0)+(1-D_0)YD_0)}_{\in T_{D_0}\mathcal{D}})$$

$$= \langle Q, D_0Y(1-D_0)+(1-D_0)YD_0\rangle_{T_{D_0}\mathcal{D}}.$$

Therefore

$$Y \in \mathcal{Y}, \quad B^*Y = D_0Y(1-D_0)+(1-D_0)YD_0. \tag{72}$$

By a classical calculation (see e.g. [4, Section 2.2]), we have

$$\forall Q \in \mathcal{Y}, \quad AQ = -[D_0,[h,Q]]. \tag{73}$$

It is also easily checked that

$$C_0^{x\,T}(B^*Y)C_0^x = C_0^{x\,T}(D_0Y(1-D_0)+(1-D_0)YD_0)C_0^x = \begin{pmatrix} * & 0 \\ 0 & * \end{pmatrix} + C_0^{x\,T}YC_0^x. \tag{74}$$

Putting together (55) and (70)–(74) yields

$$R[Y] = \sum_{x=1}^{N_f}\Pi_x C_0^x \mathfrak{L}_x^+\left(C_0^{x\,T}\left(Y-\widetilde{\ell}(Y)\Pi_x\right)C_0^x\right)C_0^{x\,T}\Pi_x, \tag{75}$$

where

$$\widetilde{\ell}(Y) := \ell(A^{-1}B^*Y) = \mathrm{Tr}(GY) \quad \text{with} \quad G := \sum_{x=1}^{N_f} C_0^x \mathfrak{L}_x^+(\mathfrak{p}^x) C_0^{x\,T} \in \mathbb{R}^{L\times L}_{\mathrm{sym}}. \tag{76}$$

Using the notation introduced in (26), we have

$$\widetilde{F^{\mathrm{HL}}}_{h+Y}(D_0) = \sum_{x=1}^{N_f}\Pi_x C_0^x \mathbb{1}_{(-\infty,0]}\left(C_0^{x\,T}(h+Y-\mu_Y\Pi_x)C_0^x\right)C_0^{x\,T}\Pi_x,$$

where $\mu_Y \in \mathbb{R}$ is chosen such that $\mathrm{Tr}(\widetilde{F^{\mathrm{HL}}}_{h+Y}(D_0)) = N$. Using similar perturbation argument as in Section 7.6.2, one can check that $\widetilde{F^{\mathrm{HL}}}_{h+Y}(D_0)$ is well-defined for $Y \in \mathcal{Y}$ small enough, and that

$$\widetilde{F^{\mathrm{HL}}}_{h+Y}(D_0) = \sum_{x=1}^{N_f}\Pi_x C_0^x \mathbb{1}_{(-\infty,0]}\left(\mathfrak{h}^x + (C_0^{x\,T}(Y-\mu_Y\Pi_x)C_0^x\right)C_0^{x\,T}\Pi_x$$

$$= \widetilde{F^{\mathrm{HL}}}_h(D_0) + \sum_{x=1}^{N_f}\Pi_x C_0^x \mathfrak{L}_x^+\left(C_0^{x\,T}(Y+\mu_Y\Pi_x)C_0^x\right)C_0^{x\,T}\Pi_x + o(\|Y\|),$$

with $\mu_Y = \widetilde{\ell}(Y)$ by particle conservation. This shows that the map R defined by (75)-(76) actually coincides with the 4-point response function in Assumption (A4).

7.6.4 | About Assumptions (A3) and (A4) in the one-site-per-fragment setting

Let us show that when $N_f = L$, we have under Assumptions (A1)–(A2),

$$\text{(A3) are satisfied} \Rightarrow D_0 \text{ is an irreducible matrix} \iff \text{(A4) is satisfied.}$$

Throughout this section, we assume that (A1)–(A2) are fulfilled.

Let us first show that (A3) implies that D_0 is irreducible. We deduce from the second assertion of Lemma 11 that (A3) is satisfied if and only if the only matrices in $\mathbb{R}^{L\times L}_{\rm sym}$ which commute with D_0 and all the Π_x's are the multiples of the identity matrix. When $N_f = L$, the matrices in $\mathbb{R}^{L\times L}_{\rm sym}$ which commute with all the Π_x are the diagonal matrices. The diagonal matrices $\Lambda = \mathrm{diag}(\lambda_1, \dots, \lambda_L)$ which commute with D_0 are the ones for which

$$\forall 1 \le i, j \le L, \quad \lambda_i [D_0]_{ij} = [D_0]_{ij} \lambda_j.$$

If D_0 was reducible, then one could find a permutation matrix $P \in O(L)$ such that PD_0P^{-1} is a 2×2 block-diagonal matrix. The matrix $P\mathrm{diag}(1, \dots, 1, 2, \dots, 2)P^{-1}$, where the numbers of entries 1 and 2 match the sizes of the blocks of PD_0P^{-1}, would then be a diagonal matrix which commutes with D_0 and is not proportional to the identity matrix. We reach a contradiction. Thus, (A3) implies that D_0 is irreducible.

Let us now show the equivalence

$$D_0 \text{ is an irreducible matrix} \iff \text{(A4) is satisfied.}$$

We have for all $Y \in \mathcal{Y}$,

$$\begin{aligned}
\|R[Y]\|^2 &= \mathrm{Tr}((R[Y])(R[Y])) \\
&= \sum_{x,x'=1}^{N_f} \mathrm{Tr}\left(\Pi_x C_0^x \mathfrak{L}_x^+ \left(C_0^{xT} (Y - \widetilde{\ell}(Y)\Pi_x) C_0^x \right) C_0^{xT} \Pi_x \right. \\
&\quad \left. \times \Pi_{x'} C_0^{x'} \mathfrak{L}_{x'}^+ \left(C_0^{x'T} (Y - \widetilde{\ell}(Y)\Pi_{x'}) C_0^{x'} \right) C_0^{x'T} \Pi_{x'} \right) \\
&= \sum_{x=1}^{N_f} \mathrm{Tr}\left(\Pi_x C_0^x \mathfrak{L}_x^+ \left(C_0^{xT} (Y - \widetilde{\ell}(Y)\Pi_x) C_0^x \right) C_0^{xT} \Pi_x C_0^x \mathfrak{L}_x^+ \left(C_0^{xT} (Y - \widetilde{\ell}(Y)\Pi_x) C_0^x \right) C_0^{xT} \Pi_x \right) \\
&= \sum_{x=1}^{N_f} \mathrm{Tr}\left(\mathfrak{p}_x \mathfrak{L}_x^+ \left(C_0^{xT} (Y - \widetilde{\ell}(Y)\Pi_x) C_0^x \right) \mathfrak{p}_x \mathfrak{L}_x^+ \left(C_0^{xT} (Y - \widetilde{\ell}(Y)\Pi_x) C_0^x \right) \right) \\
&= \sum_{x=1}^{N_f} \| \mathfrak{p}^x \mathfrak{L}_x^+ \left(C_0^{xT} (Y - \widetilde{\ell}(Y)\Pi_x) C_0^x \right) \mathfrak{p}^x \|^2.
\end{aligned}$$

Using (50) and (55)–(56), we obtain after straightforward algebraic manipulations that

$$\begin{aligned}
(R[Y] = 0) &\iff \left(\forall 1 \le x \le N_f,\ \mathfrak{p}^x \mathfrak{L}_x^+ \left(C_0^{xT} (Y - \widetilde{\ell}(Y)\Pi_x) C_0^x \right) \mathfrak{p}^x = 0 \right) \\
&\iff \left(\forall 1 \le x \le N_f,\ (1 - D_{0,x})^{1/2} \widetilde{N}_x(Y) D_{0,x}^{1/2} + D_{0,x}^{1/2} \widetilde{N}_x(Y)^T (1 - D_{0,x})^{1/2} = 0 \right),
\end{aligned}$$

with

$$\widetilde{N}_x(Y) := N\Big((1-D_{0,x})^{-1/2}E_x^T(1-D_0)YD_0E_xD_{0,x}^{-1/2} - \widetilde{\ell}(Y)D_{0,x}^{1/2}(1-D_{0,x})^{1/2}\Big).$$

In the case when $N_f = L$, we have $L_x = 1$ for all x, and thus, $D_{0,x}$ and $\widetilde{N}(Y)$ are scalar quantities. We then have in this special case by assumption (A2),

$$(R[Y]=0) \iff \big(\forall 1 \le x \le N_f,\ N_x(Y)=0\big) \iff \big(My = \widetilde{\ell}(Y)z\big),$$

where $y = (Y_{11}, \dots, Y_{LL})^T \in \mathbb{R}^L$, $z = (D_{0,1}(1-D_{0,1}), \dots, D_{0,L}(1-D_{0,L}))^T \in \mathbb{R}^L$, and $M \in \mathbb{R}^{L\times L}_{\mathrm{sym}}$ is the matrix with entries

$$M_{xx} = [D_0]_{xx} - [D_0]_{xx}^2, \quad M_{xx'} = -[D_0]_{xx'}^2, \text{ if } x \neq x'.$$

Still by Assumption (A2), $\sum_{x=1}^{N_f} z_x > 0$, and therefore using the fact that D_0 is an orthogonal projector (hence that $\sum_{x=1}^{N_f}[D_0]_{x,x'}^2 = [D_0^2]_{xx} = [D_0]_{xx}$), we get

$$(My = \widetilde{\ell}(Y)z) \Rightarrow \left(\widetilde{\ell}(Y) = \frac{\sum_{x,x'=1}^{N_f} M_{x,x'}y_{x'}}{\sum_{x=1}^{N_f} z_x} = \frac{\sum_{x=1}^{N_f}[D_0]_{x,x}y_x - \sum_{x,x'=1}^{N_f}[D_0]_{x,x'}^2 y_{x'}}{\sum_{x=1}^{N_f} z_x} = 0\right).$$

Therefore,

$$(R[Y]=0) \iff (My = 0).$$

The matrix M is hermitian, diagonal dominant with positive diagonal elements and non-positive off-diagonal elements, and such that

$$\forall 1 \le x \le N_f, \quad M_{xx} = -\sum_{x'\neq x} M_{xx'}.$$

Therefore the kernel of M is reduced to $\mathbb{R}(1, \dots, 1)^T$ if and only if M is irreducible. Besides, we see from the expressions of the coefficients of M and Assumption (A2) that M is irreducible if and only if D_0 is irreducible. We conclude that R is injective, hence bijective, if and only if D_0 is irreducible.

7.7 | Proof of Theorem 5

7.7.1 | Perturbation expansion in the Fock space

This calculation is classical in the physics and chemistry literature, but we report it here for the sake of completeness. Consider a family of Hamiltonians $(\widehat{H}_\alpha)_{\alpha\in\mathbb{R}}$ of the form

$$\widehat{H}_\alpha := \widehat{H}_0 + \alpha(\widehat{W}_1 + \widehat{W}_2)$$

on the real Fock space $\mathrm{Fock}(\mathbb{R}^{N_b})$ where

$$\widehat{H}_0 := \sum_{m,n=1}^{N_b} [h_0]_{mn}c_m^\dagger c_n \quad \text{and} \quad \widehat{W}_1 := \sum_{m,n=1}^{N_b} [W_1]_{mn}c_m^\dagger c_n$$

are one-body Hamiltonians and

$$\widehat{W}_2 := \frac{1}{2} \sum_{m,n,p,q,=1}^{N_b} [W_2]_{mnpq} c_m^\dagger c_n^\dagger c_q c_p$$

is a two-body Hamiltonian.

Let us provisionally assume that h_0 is diagonal, and more precisely that

$$h_0 = \mathrm{diag}(\varepsilon_1^0, \dots, \varepsilon_{N_b}^0) \quad \text{with} \quad \varepsilon_1^0 \le \cdots \le \varepsilon_{\mathcal{N}}^0 < 0 < \varepsilon_{\mathcal{N}+1}^0 \le \cdots \varepsilon_{N_b}^0.$$

This amounts to working in a molecular orbital basis set of the unperturbed one-body Hamiltonian h_0 and assuming that the Fermi level ϵ_{F} for having $\mathcal{N}$ particles in the ground state can be chosen equal to zero. The ground state Ψ_0 of $\widetilde{H}_0$ in the $\mathcal{N}$-particle sector then is unique and so is the one of $\widetilde{H}_\alpha$ for α small by perturbation theory. We have

$$\Psi_0 = \frac{1}{\sqrt{\mathcal{N}!}} c_{\mathcal{N}}^\dagger \cdots c_1^\dagger |0\rangle, \quad E_0 := \langle \Psi_0 | \widehat{H}_0 | \Psi_0 \rangle = \sum_{i=1}^{\mathcal{N}} \varepsilon_i^0.$$

Denoting by $d(\alpha)$ the ground-state one-body reduced density matrix of $\widehat{H}_\alpha$, the map $\alpha \mapsto d(\alpha)$ is real-analytic in the neighborhood of 0 and

$$d(\alpha) = d_0 + \alpha d_1 + O(\alpha^2) \quad \text{with} \quad d_0 := \begin{pmatrix} I_{\mathcal{N}} & 0 \\ 0 & 0 \end{pmatrix}.$$

In addition, we have

$$[d_1]_{mn} = \langle \Psi_1 | c_m^\dagger c_n | \Psi_0 \rangle + \langle \Psi_0 | c_m^\dagger c_n | \Psi_1 \rangle,$$

where Ψ_1 is the first-order perturbation of the ground-state wave-function Ψ_0, solution to

$$(\widehat{H}_0 - E_0)\Psi_1 = -\Pi_{\Psi_0^\perp}\big((\widehat{W}_1 + \widehat{W}_2)\Psi_0\big), \qquad \Psi_1 \in \Psi_0^\perp.$$

For $1 \le i_1 < \cdots < i_r \le \mathcal{N}$ (occupied orbitals) and $m+1 \le a_1 < \cdots < a_r \le N_b$ (virtual orbitals), we set

$$\Phi_0^0 := \Psi_0 \quad \text{and} \quad \Phi_{i_1 \cdots i_r}^{a_1 \cdots a_r} = c_{a_r}^\dagger \cdots c_{a_1}^\dagger c_{i_1} \cdots c_{i_r} \Phi_0^0.$$

The $\Phi_{i_1 \cdots i_r}^{a_1 \cdots a_r}$'s $(0 \le r \le \min(\mathcal{N}, N_b - \mathcal{N}),\ 1 \le i_1 < \cdots < i_r \le \mathcal{N},\ a_1 < \cdots < a_r \le N_b$, form an orthonormal basis of eigenfunctions of the restriction of $\widehat{H}_0$ to the $\mathcal{N}$-particle sector and it holds

$$\widehat{H}_0 \Phi_{i_1 \cdots i_r}^{a_1 \cdots a_r} = E_{i_1 \cdots i_r}^{a_1 \cdots a_r} \Phi_{i_1 \cdots i_r}^{a_1 \cdots a_r} \quad \text{with} \quad E_{i_1 \cdots i_r}^{a_1 \cdots a_r} = E_0 + \sum_{s=1}^{r} \varepsilon_{a_s} - \sum_{s=1}^{r} \varepsilon_{i_s}.$$

We thus obtain the sum-over-state formula

$$\Psi_1 = - \sum_{1 \le r \le \min(\mathcal{N}, N_b - \mathcal{N})} \sum_{1 \le i_1 < \cdots < i_r \le \mathcal{N}} \sum_{\mathcal{N}+1 \le a_1 < \cdots < a_r \le N_b} \frac{\langle \Phi_{i_1 \cdots i_r}^{a_1 \cdots a_r} | \widehat{W}_1 + \widehat{W}_2 | \Phi_0^0 \rangle}{E_{i_1 \cdots i_r}^{a_1 \cdots a_r} - E_0} \Phi_{i_1 \cdots i_r}^{a_1 \cdots a_r},$$

yielding

$$[d_1]_{mn} = -\sum_{1\le r\le \min(\mathcal{N},N_b-\mathcal{N})}\ \sum_{1\le i_1<\cdots<i_r\le\mathcal{N}}\ \sum_{\mathcal{N}+1\le a_1<\cdots<a_r\le N_b} \frac{\langle\Phi^{a_1\cdots a_r}_{i_1\cdots i_r}|\widehat{W}_1+\widehat{W}_2|\Phi^0_0\rangle}{E^{a_1\cdots a_r}_{i_1\cdots i_r}-E_0} \times\Big(\langle\Phi^{a_1\cdots a_r}_{i_1\cdots i_r}|c^\dagger_m c_n|\Phi^0_0\rangle+\langle\Phi^0_0|c^\dagger_m c_n|\Phi^{a_1\cdots a_r}_{i_1\cdots i_r}\rangle\Big).$$

Since $\langle\Phi^{a_1\cdots a_r}_{i_1\cdots i_r}|a^\dagger_m a_n|\Phi^0_0\rangle = 0$ if $r\ge 2$, and

$$\langle\Phi^a_i|c^\dagger_m c_n|\Phi^0_0\rangle = \delta_{n,i}\delta_{m,a},$$

$$\langle\Phi^a_i|c^\dagger_m c^\dagger_n c_q c_p|\Phi^0_0\rangle = -\delta_{m,q}\delta_{n,i}\delta_{p,a}\delta_{q\le\mathcal{N}}+\delta_{m,p}\delta_{n,i}\delta_{q,a}\delta_{p\le\mathcal{N}}+\delta_{m,i}\delta_{n,q}\delta_{p,a}\delta_{q\le\mathcal{N}} -\delta_{m,i}\delta_{n,p}\delta_{q,a}\delta_{p\le\mathcal{N}},$$

this expression reduces to

$$[d_1]_{mn} = -\sum_{i=1}^{N}\sum_{a=\mathcal{N}+1}^{N_b}\frac{\langle\Phi^a_i|\widehat{W}_1+\widehat{W}_2|\Phi^0_0\rangle}{\varepsilon^0_a-\varepsilon^0_i}(\delta_{n=i}\delta_{m=a}+\delta_{m=i}\delta_{n=a}).$$

We obtain that d_1 is of the form

$$d_1 = \begin{pmatrix} 0 & d_1^{+-} \\ d_1^{+-T} & 0\end{pmatrix} \quad\text{with}\quad \forall 1\le i\le\mathcal{N}<\mathcal{N}+1\le a\le N_b,\quad [d_1]_{ai} = \frac{\langle\Phi^a_i|\widehat{W}_1+\widehat{W}_2|\Phi^0_0\rangle}{\varepsilon^0_a-\varepsilon^0_i}.$$

Finally, we have

$$\begin{aligned}[d_1]_{ai} &= \sum_{m,n=1}^{N_b}[W_1]_{mn}\frac{\langle\Phi^a_i|c^\dagger_m c_n|\Phi^0_0\rangle}{\varepsilon^0_a-\varepsilon^0_i}+\sum_{m,n,p,q=1}^{N_b}[W_2]_{mnpq}\frac{\langle\Phi^a_i|c^\dagger_m c^\dagger_q c_q c_p|\Phi^0_0\rangle}{\varepsilon^0_a-\varepsilon^0_i}\\ &= \frac{[W_1+J_{W_2}(d_0)-K_{W_2}(d_0)]_{ai}}{\varepsilon^0_a-\varepsilon^0_i},\end{aligned}$$

where the direct and exchange operators are respectively given by

$$[J_{W_2}(d)]_{mn} := \sum_{p,q=1}^{N_b}[W_2]_{npmq}d_{pq} \quad\text{and}\quad [K_{W_2}(d)]_{mn} := \sum_{p,q=1}^{N_b}[W_2]_{npqm}d_{pq}.$$

Introducing the linear response operator $\mathfrak{L}^+_{h_0}$ such that

$$\mathbb{1}_{(-\infty,\varepsilon_F]}(h_0+W) = \underbrace{\mathbb{1}_{(-\infty,\varepsilon_F]}(h_0+W)}_{=d_0} - \mathfrak{L}^+_{h_0}W + O(\|W\|),$$

we finally obtain

$$d_1 = -\mathfrak{L}^+_{h_0}\big(W_1+J_{W_2}(d_0)-K_{W_2}(d_0)\big), \tag{77}$$

this formula remaining valid in the general case when h_0 is not *a priori* diagonal and ϵ_F not *a priori* equal to zero.

7.7.2 | Perturbation expansion of the DMET ground-state

Under Assumption (A1), the HF problem

$$\operatorname*{argmin}_{D\in\mathcal{D}} \mathcal{E}_\alpha^{\mathrm{HF}}(D)$$

has a unique minimizer $D^{\mathrm{HF}}(\alpha)$ for α small enough and the map $\alpha \mapsto D^{\mathrm{HF}}(\alpha)$ is real-analytic in the neighborhood of 0. This results from a straightforward application of nonlinear perturbation theory, which we do not detail here for the sake of brevity. We set $P^{\mathrm{HF}}(\alpha) := \mathrm{Bd}(D^{\mathrm{HF}}(\alpha))$, and

$$D_1^{\mathrm{exact}} := \frac{dD^{\mathrm{exact}}}{d\alpha}(0), \qquad D_1^{\mathrm{HF}} := \frac{dD^{\mathrm{HF}}}{d\alpha}(0), \qquad D_1^{\mathrm{DMET}} := \frac{dD^{\mathrm{DMET}}}{d\alpha}(0),$$

$$P_1^{\mathrm{exact}} := \frac{dP^{\mathrm{exact}}}{d\alpha}(0), \qquad P_1^{\mathrm{HF}} := \frac{dP^{\mathrm{HF}}}{d\alpha}(0), \qquad P_1^{\mathrm{DMET}} := \frac{dP^{\mathrm{DMET}}}{d\alpha}(0).$$

We are going to prove that the above first three matrices on the one hand, and the last three ones on the other hand are equal in $T_{D_0}\mathcal{D}$ and $\mathcal{Y}$ respectively.

First, we deduce from (77) applied with $N_b = L$, $\epsilon_{\mathrm{F}} = 0$, $h_0 = h$, $W_1 = 0$, $W_2 = v$, that

$$D_1^{\mathrm{exact}} = -\mathfrak{L}_h^+(J(D_0) - K(D_0)),$$

where J and K are the direct and exchange operators for the two-body interaction potential $\widehat{V}$ introduced in (21).

Next, by differentiating the self-consistent equation

$$D^{\mathrm{HF}}(\alpha) = \mathbb{1}_{(-\infty,0]}\big(h^{\mathrm{MF}}(\alpha, D^{\mathrm{HF}}(\alpha))\big),$$

where

$$h^{\mathrm{MF}}(\alpha, D) = h + \alpha(J(D) - K(D))$$

is the Fock Hamiltonian for the interaction parameter α, we get

$$D_1^{\mathrm{HF}} = -\mathfrak{L}_h^+(J(D_0) - K(D_0)).$$

Hence

$$D_1^{\mathrm{HF}} = D_1^{\mathrm{exact}} \quad \text{and} \quad P_1^{\mathrm{HF}} = \mathrm{Bd}(D_1^{\mathrm{HF}}) = \mathrm{Bd}(D_1^{\mathrm{exact}}) = P_1^{\mathrm{exact}}.$$

Let us now show that $P_1^{\mathrm{DMET}} = P_1^{\mathrm{HF}}$. For convenience, we will use the following notation

$$F^{\mathrm{LL}}(\alpha, P) := F_\alpha^{\mathrm{LL}}(P), \qquad F^{\mathrm{HL}}(\alpha, D) = F_\alpha^{\mathrm{HL}}(D),$$

$$F_{\mathrm{HF}}^{\mathrm{HL}}(\alpha, D) := \sum_{x=1}^{N_f} \Pi_x C^x(D) \mathbb{1}_{(-\infty,0]}\big(C^x(D)^T\big(h^{\mathrm{MF}}(\alpha, D) - \mu^{\mathrm{HF}}(\alpha, D)\Pi_x\big)C^x(D)\big)C^x(D)^T\Pi_x,$$

where $\mu^{\mathrm{HF}}(\alpha, D) \in \mathbb{R}$ is the Lagrange parameter of the charge conservation constraint. The map $F_{\mathrm{HF}}^{\mathrm{HL}}(\alpha, D)$ is the high-level HF map for the interacting parameter α, introduced in Remark 2 for $\alpha = 1$.

We know from Theorem 4 that for all α small enough

$$F^{\mathrm{HL}}\big(\alpha, F^{\mathrm{LL}}\big(\alpha, P^{\mathrm{DMET}}(\alpha)\big)\big) = P^{\mathrm{DMET}}(\alpha).$$

Taking the derivative at $\alpha = 0$, we get

$$\partial_\alpha F^{\mathrm{HL}}(0, D_0) + \partial_D F^{\mathrm{HL}}(0, D_0)\big(\partial_\alpha F^{\mathrm{LL}}(0, P_0) + \partial_P F^{\mathrm{LL}}(0, P_0) P_1^{\mathrm{DMET}}\big) = P_1^{\mathrm{DMET}}. \tag{78}$$

The same arguments as in the proof of Proposition 1 allow one to show that for all α small enough

$$F^{\mathrm{HL}}_{\mathrm{HF}}\big(\alpha, F^{\mathrm{LL}}\big(\alpha, P^{\mathrm{HF}}(\alpha)\big)\big) = P^{\mathrm{HF}}(\alpha),$$

yielding

$$\partial_\alpha F^{\mathrm{HL}}_{\mathrm{HF}}(0, D_0) + \partial_D F^{\mathrm{HL}}_{\mathrm{HF}}(0, D_0)\big(\partial_\alpha F^{\mathrm{LL}}(0, P_0) + \partial_P F^{\mathrm{LL}}(0, P_0) P_1^{\mathrm{HF}}\big) = P_1^{\mathrm{HF}}. \tag{79}$$

Since $F^{\mathrm{HL}}_{\mathrm{HF}}(0, D) = F^{\mathrm{HL}}(0, D)$ for all D in the neighborhood of D_0, we have

$$\partial_P F^{\mathrm{HL}}_{\mathrm{HF}}(0, D_0) = \partial_P F^{\mathrm{HL}}(0, D_0).$$

Using (67) and the invertibility of $d_P\Phi(0, P_0)$ established in Section 7.6.3, we obtain

$$P_1^{\mathrm{DMET}} = -\big(d_P\Phi(0, P_0)\big)^{-1}\big(\partial_\alpha F^{\mathrm{HL}}(0, D_0) + \partial_D F^{\mathrm{HL}}(0, D_0)\partial_\alpha F^{\mathrm{LL}}(0, P_0)\big), \tag{80}$$

$$P_1^{\mathrm{HF}} = -\big(d_P\Phi(0, P_0)\big)^{-1}\big(\partial_\alpha F^{\mathrm{HL}}_{\mathrm{HF}}(0, D_0) + \partial_D F^{\mathrm{HL}}(0, D_0)\partial_\alpha F^{\mathrm{LL}}(0, P_0)\big). \tag{81}$$

Let us show that $\partial_\alpha F^{\mathrm{HL}}(0, D_0) = \partial_\alpha F^{\mathrm{HL}}_{\mathrm{HF}}(0, D_0)$. On the one hand, we have

$$F^{\mathrm{HL}}_{\mathrm{HF}}(\alpha, D_0) = \sum_{x=1}^{N_f} \Pi_x C_0^x \mathbb{1}_{(-\infty,0]}\Big(C_0^{x\,T}(h + \alpha(J(D_0) - K(D_0)) - \mu_{\mathrm{HF}}(\alpha, D_0)\Pi_x) C_0^x\Big) C_0^{x\,T}\Pi_x,$$

and therefore

$$\partial_\alpha F^{\mathrm{HL}}_{\mathrm{HF}}(0, D_0) = -\sum_{x=1}^{N_f} \Pi_x C_0^x \mathfrak{Q}_x^+\Big(C_0^{x\,T}(J(D_0) - K(D_0)) C_0^x - \partial\mu_{\mathrm{HF}}(0, D_0)\mathfrak{p}^x\Big) C_0^{x\,T}\Pi_x. \tag{82}$$

On the other hand, we have

$$F^{\mathrm{HL}}(\alpha, D_0) = \sum_{x=1}^{N_f} \Pi_x C_0^x D^{\mathrm{imp}}_{x,D_0}(\alpha) C_0^{x\,T}\Pi_x,$$

where $D^{\mathrm{imp}}_{x,D_0}(\alpha)$ is the ground-state one-body reduced density matrix in the basis of Y_{x,D_0} defined by C_0^x of the impurity Hamiltonian (see Proposition 8)

$$\widehat{H}^{\mathrm{imp}}_{x,D_0}(\alpha) = \sum_{i,j=1}^{2L_x} \Big[C_0^{x\,T}(h + \alpha(J(\mathfrak{D}^x(D_0)) - K(\mathfrak{D}^x(D_0))))C_0^x\Big]_{ij} \widehat{a}_i(D_0)^\dagger \widehat{a}_j(D_0)$$

$$+\frac{\alpha}{2}\sum_{i,j,k,\ell=1}^{2L_x}[V^x(D_0)]_{ijkl}\widehat{a}_i(D_0)^\dagger\widehat{a}_j(D_0)^\dagger\widehat{a}_\ell(D_0)\widehat{a}_k(D_0)$$

$$-\mu(\alpha)\sum_{i,j=1}^{2L_x}\left[C_0^{x\,T}\Pi_x C_0^x\right]_{ij}\widehat{a}_i(D_0)^\dagger\widehat{a}_j(D_0),$$

where $\mu(\alpha)$ is the Lagrange multiplier of the charge neutrality constraint and where we have discarded the irrelevant constant $E_x^{\rm env}(D_0)$. Using the notation introduced in (43), this Hamiltonian can be rewritten as

$$\widehat{H}^{\rm imp}_{x,D_0}(\alpha)=\sum_{i,j=1}^{2L_x}[\mathfrak{h}^x]_{ij}\widehat{a}_i(D_0)^\dagger\widehat{a}_j(D_0)$$

$$+\alpha\Bigg(\sum_{i,j=1}^{2L_x}\left[C_0^{x\,T}(J(\mathfrak{D}^x(D_0))-K(\mathfrak{D}^x(D_0)))C_0^x\right]_{ij}\widehat{a}_i(D_0)^\dagger\widehat{a}_j(D_0)$$

$$+\frac{1}{2}\sum_{i,j,k,\ell=1}^{2L_x}[V^x(D_0)]_{ijkl}\widehat{a}_i(D_0)^\dagger\widehat{a}_j(D_0)^\dagger\widehat{a}_\ell(D_0)\widehat{a}_k(D_0)\Bigg)$$

$$-\mu(\alpha)\sum_{i,j=1}^{2L_x}\left[C_0^{x\,T}\Pi_x C_0^x\right]_{ij}\widehat{a}_i(D_0)^\dagger\widehat{a}_j(D_0).$$

We have

$$D^{\rm imp}_{x,D_0}(0)=\begin{pmatrix}I_{L_x}&0\\0&0\end{pmatrix}.$$

Since $\mu(0)=0$ and $\alpha\mapsto\mu(\alpha)$ is real-analytic, we can easily adapt the analysis done in the previous section to the case when

$$N_b=2L_x,\quad h_0=\mathfrak{h}^x,\quad W_1=C_0^{x\,T}\big(J(\mathfrak{D}^x(D_0))-K(\mathfrak{D}^x(D_0))-\mu'(0)\Pi_x\big)C_0^x,\quad W_2=V^x(D_0),$$

and infer that

$$D^{\rm imp}_{x,D_0}(\alpha)=D^{\rm imp}_{x,D_0}(0)-\alpha\mathfrak{L}_x^+\Big(C_0^{x\,T}\big(J(\mathfrak{D}^x(D_0))-K(\mathfrak{D}^x(D_0))-\mu'(0)\Pi_x\big)C_0^x$$

$$+J_{V^x(D_0)}(D^{\rm imp}_{x,D_0}(0))-K_{V^x(D_0)}(D^{\rm imp}_{x,D_0}(0))\Big)+O(\alpha^2),$$

where $\mathfrak{L}_x^+$ is the linear response operator introduced in (54). Observing that

$$C_0^{x\,T}(J(\mathfrak{D}^x(D_0))-K(\mathfrak{D}^x(D_0)))C_0^x+J_{V^x(D_0)}(D^{\rm imp}_{x,D_0}(0))-K_{V^x(D_0)}(D^{\rm imp}_{x,D_0}(0))$$

$$=C_0^{x\,T}(J(D_0)-K(D_0))C_0^x,$$

we obtain that

$$\partial_\alpha F^{\rm HL}(0,D_0)=-\sum_{x=1}^{N_f}\Pi_x C_0^x\mathfrak{L}_x^+\Big(C_0^{x\,T}(J(D_0)-K(D_0))C_0^x-\mu'(0)\mathfrak{p}^x\Big)C_0^{x\,T}\Pi_x. \tag{83}$$

Since the roles of the scalars $\partial_\alpha \mu(0, D_0)$ in (82) and $\mu'(0)$ in (83) are simply to ensure charge neutrality, these two scalars are the same. It follows that $\partial_\alpha F^{\mathrm{HL}}_{\mathrm{HF}}(0, D_0) = \partial_\alpha F^{\mathrm{HL}}(0, D_0)$, which allows us to deduce from (80) to (81) that $P_1^{\mathrm{DMET}} = P_1^{\mathrm{HF}}$. Finally, we obtain that $D_1^{\mathrm{DMET}} = D_1^{\mathrm{HF}}$ by differentiating the relations

$$D^{\mathrm{DMET}}(\alpha) = F^{\mathrm{LL}}(\alpha, P^{\mathrm{DMET}}(\alpha)) \quad \text{and} \quad D^{\mathrm{HF}}(\alpha) = F^{\mathrm{LL}}(\alpha, P^{\mathrm{HF}}(\alpha)),$$

and using the fact that $P_1^{\mathrm{DMET}} = P_1^{\mathrm{HF}}$.

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REFERENCES

1. Y. Ai, Q. Sun, and H. Jiang, *Efficient multiconfigurational quantum chemistry approach to single-ion magnets based on density matrix embedding theory*, J. Phys. Chem. Lett. **13** (2022), no. 45, 10 627–10 634.
2. I. W. Bulik, W. Chen, and G. E. Scuseria, *Electron correlation in solids via density embedding theory*, J. Chem. Phys. **141** (2014), no. 5, 054113.
3. I. W. Bulik, G. E. Scuseria, and J. Dukelsky, *Density matrix embedding from broken symmetry lattice mean fields*, Phys. Rev. B **89** (2014), no. 3, 035140.
4. E. Cancès, G. Kemlin, and A. Levitt, *Convergence analysis of direct minimization and self-consistent iterations*, SIAM J. Matrix Anal. Appl. **42** (2021), no. 1, 243–274.
5. C. Cao, J. Sun, X. Yuan, H.-S. Hu, H. Q. Pham, and D. Lv, *Ab initio quantum simulation of strongly correlated materials with quantum embedding*, npj Comput. Mater. **9** (2023), 78.
6. Q. Chen, G. H. Booth, S. Sharma, G. Knizia, and G. K.-L. Chan, *Intermediate and spin-liquid phase of the half-filled honeycomb Hubbard model*, Phys. Rev. B **89** (2014), no. 16, 165134.
7. Y. Chen, Y. Khoo, and M. Lindsey, *Multiscale semidefinite programming approach to positioning problems with pairwise structure*, J. Sci. Comput. **101** (2024), 42.
8. J. Čížek, *On the correlation problem in atomic and molecular systems. Calculation of wavefunction components in Ursell-type expansion using quantum-field theoretical methods*, J. Chem. Phys. **45** (1966), 4256.
9. Z.-H. Cui, C. Sun, U. Ray, B.-X. Zheng, Q. Sun, and G. K.-L. Chan, *Ground-state phase diagram of the three-band Hubbard model from density matrix embedding theory*, Phys. Rev. Res. **2** (2020), no. 4, 043259.
10. Z.-H. Cui, T. Zhu, and G. K.-L. Chan, *Efficient implementation of ab initio quantum embedding in periodic systems: Density matrix embedding theory*, J. Chem. Theory Comput. **16** (2020), 119–129.
11. Z. Fan and Q.-l. Jie, *Cluster density matrix embedding theory for quantum spin systems*, Phys. Rev. B **91** (2015), no. 19, 195118.
12. F. M. Faulstich, R. Kim, Z.-H. Cui, Z. Wen, G. Kin-Lic Chan, and L. Lin, *Pure state v-representability of density matrix embedding theory*, J. Chem. Theory Comput . **18** (2022), no. 2, 851–864.
13. E. Fertitta and G. H. Booth, *Rigorous wave function embedding with dynamical fluctuations*, Phys. Rev. B **98** (2018), no. 23, 235132.
14. M. Ganahl, M. Aichhorn, H. G. Evertz, P. Thunström, K. Held, and F. Verstraete, *Efficient DMFT impurity solver using real-time dynamics with matrix product states*, Phys. Rev. B **92** (2015), no. 15, 155132.

15. A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, *Dynamical mean-field theory of strongly correlated fermion systems and the limit of infinite dimensions*, Rev. Modern Phys. **68** (1996), no. 1, 13.
16. A. Georges and W. Krauth, *Numerical solution of the d=∞ Hubbard model: Evidence for a Mott transition*, Phys. Rev. Lett. **69** (1992), no. 8, 1240.
17. M. S. Gordon, D. G. Fedorov, S. R. Pruitt, and L. V. Slipchenko, *Fragmentation methods: A route to accurate calculations on large systems*, Chem. Rev. **112** (2012), no. 1, 632–672.
18. K. Gunst, S. Wouters, S. De Baerdemacker, and D. Van Neck, *Block product density matrix embedding theory for strongly correlated spin systems*, Phys. Rev. B **95** (2017), no. 19, 195127.
19. M. R. Hermes and L. Gagliardi, *Multiconfigurational self-consistent field theory with density matrix embedding: The localized active space self-consistent field method*, J. Chem. Theory Comput. **15** (2019), no. 2, 972–986.
20. L. O. Jones, M. A. Mosquera, G. C. Schatz, and M. A. Ratner, *Embedding methods for quantum chemistry: Applications from materials to life sciences*, J. Am. Chem. Soc. **142** (2020), no. 7, 3281–3295.
21. R. V. Kadison, *The Pythagorean theorem: I. The finite case*, Proc. Nat. Acad. Sci. **99** (2002), no. 7, 4178–4184.
22. Y. Khoo and M. Lindsey, *Scalable semidefinite programming approach to variational embedding for quantum many-body problems*, J. Comput. Phys. **510** (2024), 113041.
23. G. Knizia and G. K.-L. Chan, *Density matrix embedding: A simple alternative to dynamical mean-field theory*, Phys. Rev. Lett. **109** (2012), no. 18, 186404.
24. G. Knizia and G. K.-L. Chan, *Density matrix embedding: A strong-coupling quantum embedding theory*, J. Chem. Theory Comput. **9** (2013), no. 3, 1428–1432.
25. P. J. Knowles and N. C. Handy, *A new determinant-based full configuration interaction method*, Chem. Phys. Lett. **111** (1984), no. 4–5, 315–321.
26. G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. Marianetti, *Electronic structure calculations with dynamical mean-field theory*, Rev. Modern Phys. **78** (2006), no. 3, 865.
27. L. Lin and M. Lindsey, *Variational embedding for quantum many-body problems*, Comm. Pure Appl. Math. **75** (2022), no. 9, 2033–2068.
28. Y. Liu, O. R. Meitei, Z. E. Chin, A. Dutt, M. Tao, T. Van Voorhis, and I. L. Chuang, *Bootstrap embedding on a quantum computer*, J. Chem. Theory. Comput. **19** (2023), no. 8, 2230–2247.
29. Y. Lu, X. Cao, P. Hansmann, and M. Haverkort, *Natural-orbital impurity solver and projection approach for Green's functions*, Phys. Rev. B **100** (2019), no. 11, 115134.
30. T. Maier, M. Jarrell, T. Pruschke, and M. H. Hettler, *Quantum cluster theories*, Rev. Modern Phys. **77** (2005), no. 3, 1027.
31. O. R. Meitei and T. Van Voorhis, *Periodic bootstrap embedding*, J. Chem. Theory Comput. **19** (2023), no. 11, 3123–3130.
32. W. Metzner and D. Vollhardt, *Correlated lattice fermions in* d = ∞ *dimensions*, Phys. Rev. Lett. **62** (1989), no. 3, 324.
33. A. Mitra, M. R. Hermes, M. Cho, V. Agarawal, and L. Gagliardi, *Periodic density matrix embedding for co adsorption on the MgO (001) surface*, J. Phys. Chem. Lett . **13** (2022), no. 32, 7483–7489.
34. A. Mitra, H. Q. Pham, R. Pandharkar, M. R. Hermes, and L. Gagliardi, *Excited states of crystalline point defects with multireference density matrix embedding theory*, J. Phys. Chem. Lett. **12** (2021), no. 48, 11 688–11 694.
35. M. Nusspickel and G. H. Booth, *Efficient compression of the environment of an open quantum system*, Phys. Rev. B **102** (2020), no. 16, 165107.
36. M. Nusspickel and G. H. Booth, *Frequency-dependent and algebraic bath states for a dynamical mean-field theory with compact support*, Phys. Rev. B **101** (2020), no. 4, 045126.
37. M. Nusspickel and G. H. Booth, *Systematic improvability in quantum embedding for real materials*, Phys. Rev. X **12** (2022), no. 1, 011046.
38. M. Nusspickel, B. Ibrahim, and G. H. Booth, *On the effective reconstruction of expectation values from ab initio quantum embedding*, J. Chem. Theory Comput. **19** (2023), no. 10, 2769–2791.
39. J. Olsen, P. Jørgensen, and J. Simons, *Passing the one-billion limit in full configuration-interaction (FCI) calculations*, Chem. Phys. Lett. **169** (1990), no. 6, 463–472.
40. H. Q. Pham, V. Bernales, and L. Gagliardi, *Can density matrix embedding theory with the complete activate space self-consistent field solver describe single and double bond breaking in molecular systems?* J. Chem. Theory Comput. **14** (2018), no. 4, 1960–1968.
41. M. Potthoff, *Two-site dynamical mean-field theory*, Phys. Rev. B **64** (2001), no. 16, 165114.

42. N. Ricke, M. Welborn, H.-Z. Ye, and T. Van Voorhis, *Performance of bootstrap embedding for long-range interactions and 2d systems*, Mol. Phys. **115** (2017), no. 17–18, 2242–2253.
43. C. J. Scott and G. H. Booth, *Extending density matrix embedding: A static two-particle theory*, Phys. Rev. B **104** (2021), no. 24, 245114.
44. S. Sekaran, M. Saubanère, and E. Fromager, *Local potential functional embedding theory: A self-consistent flavor of density functional theory for lattices without density functionals*, Computation **10** (2022), no. 3, 45.
45. B. Senjean, *Projected site-occupation embedding theory*, Phys. Rev. B **100** (2019), no. 3, 035136.
46. B. Senjean, N. Nakatani, M. Tsuchiizu, and E. Fromager, *Site-occupation embedding theory using Bethe ansatz local density approximations*, Phys. Rev. B **97** (2018), no. 23, 235105.
47. S. H. Strogatz, *Nonlinear dynamics and chaos with student solutions manual: With applications to physics, biology, chemistry, and engineering*. CRC press, 2018.
48. C. Sun, U. Ray, Z.-H. Cui, M. Stoudenmire, M. Ferrero, and G. K.-L. Chan, *Finite-temperature density matrix embedding theory*, Phys. Rev. B **101** (2020), no. 7, 075131.
49. H. K. Tran, T. Van Voorhis, and A. J. Thom, *Using SCF metadynamics to extend density matrix embedding theory to excited states*, J. Chem . Phys. **151** (2019), no. 3, 034112.
50. H. K. Tran, H.-Z. Ye, and T. Van Voorhis, *Bootstrap embedding with an unrestricted mean-field bath*, J. Chem . Phys. **153** (2020), no. 21, 214101.
51. T. Tsuchimochi, M. Welborn, and T. Van Voorhis, *Density matrix embedding in an antisymmetrized geminal power bath*, J. Chem. Phys **143** (2015), no. 2, 024107.
52. K. D. Vogiatzis, D. Ma, J. Olsen, L. Gagliardi, and W. A. de Jong, *Pushing configuration-interaction to the limit: Towards massively parallel MCSCF calculations*, J. Chem. Phys **147** (2017), no. 18, 184111.
53. C. Vorwerk, N. Sheng, M. Govoni, B. Huang, and G. Galli, *Quantum embedding theories to simulate condensed systems on quantum computers*, Nat. Computer. Sci . **2** (2022), no. 7, 424–432.
54. M. Welborn, T. Tsuchimochi, and T. Van Voorhis, *Bootstrap embedding: An internally consistent fragment-based method*, J. Chem. Phys **145** (2016), no. 7, 074102.
55. S. R. White, *Density matrix formulation for quantum renormalization groups*, Phys. Rev. Lett. **69** (1992), no. 19, 2863–2866.
56. S. Wouters, C. A. Jiménez-Hoyos, Q. Sun, and G. K.-L. Chan, *A practical guide to density matrix embedding theory in quantum chemistry*, J. Chem. Theory Comput . **12** (2016), no. 6, 2706–2719.
57. X. Wu, Z.-H. Cui, Y. Tong, M. Lindsey, G. K.-L. Chan, and L. Lin, *Projected density matrix embedding theory with applications to the two-dimensional Hubbard model*, J. Chem. Phys. **151** (2019), no. 6, 064108.
58. X. Wu, M. Lindsey, T. Zhou, Y. Tong, and L. Lin, *Enhancing robustness and efficiency of density matrix embedding theory via semidefinite programming and local correlation potential fitting*, Phys. Rev. B **102** (2020), no. 8, 085123.
59. S. Yalouz, S. Sekaran, E. Fromager, and M. Saubanère, *Quantum embedding of multi-orbital fragments using the block-householder transformation*, J. Chem. Phys. **157** (2022), no. 21, 214112.
60. H.-Z. Ye, N. D. Ricke, H. K. Tran, and T. Van Voorhis, *Bootstrap embedding for molecules*, J. Chem. Theory Comput. **15** (2019), no. 8, 4497–4506.
61. H.-Z. Ye, H. K. Tran, and T. Van Voorhis, *Bootstrap embedding for large molecular systems*, J. Chem. Theory Comput. **16** (2020), no. 8, 5035–5046.
62. H.-Z. Ye, H. K. Tran, and T. Van Voorhis, *Accurate electronic excitation energies in full-valence active space via bootstrap embedding*, J. Chem. Theory Comput. **17** (2021), no. 6, 3335–3347.
63. H.-Z. Ye and T. Van Voorhis, *Atom-based bootstrap embedding for molecules*, J. Phys. Chem. Lett . **10** (2019), no. 20, 6368–6374.
64. H.-Z. Ye, M. Welborn, N. D. Ricke, and T. Van Voorhis, *Incremental embedding: A density matrix embedding scheme for molecules*, J. Chem. Phys. **149** (2018), no. 19, 194108.
65. B.-X. Zheng and G. K.-L. Chan, *Ground-state phase diagram of the square lattice Hubbard model from density matrix embedding theory*, Phys. Rev. B **93** (2016), no. 3, 035126.
66. B.-X. Zheng, C.-M. Chung, P. Corboz, G. Ehlers, M.-P. Qin, R. M. Noack, H. Shi, S. R. White, S. Zhang, and G. K.-L. Chan, *Stripe order in the underdoped region of the two-dimensional Hubbard model*, Science **358** (2017), no. 6367, 1155–1160.

67. B.-X. Zheng, J. S. Kretchmer, H. Shi, S. Zhang, and G. K.-L. Chan, *Cluster size convergence of the density matrix embedding theory and its dynamical cluster formulation: A study with an auxiliary-field quantum Monte Carlo solver*, Phys. Rev. B **95** (2017), no. 4, 045103.

APPENDIX A: NOTATION TABLE

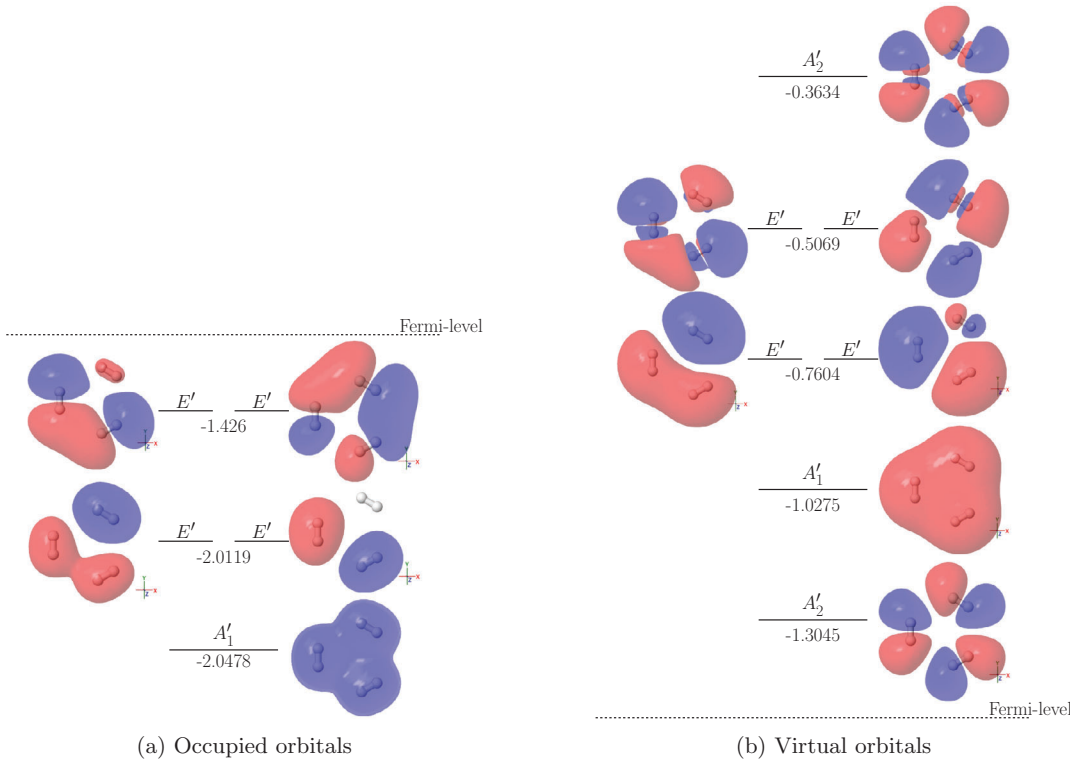
The following table collects the main notations in use in this article.

TABLE A1 Collection of the main notations used in the paper.

Symbol	Meaning	See Eq.
$\mathrm{Fock}(E)$	Fermionic Fock space associated with the one-particle state space $E \subset \mathcal{H}$	
$\mathcal{H} = \mathbb{R}^L$	One-particle state space of the whole system, L its dimension	(1)
$\mathcal{B}_{\mathrm{at}} = (e_\kappa)_{1\le\kappa\le L}$	Canonical basis of $\mathcal{H}$	(1)
$\widehat{H}$	Hamiltonian of the whole system (op. on $\mathrm{Fock}(\mathcal{H})$)	(2)
$\widehat{H}_0$	Non-interacting Hamiltonian of the whole system	(23)
$\widehat{H}_\alpha$	Hamiltonian of the whole system for coupling parameter α	(22)
N	Number of electrons in the system	
$\mathcal{D}$	Set of 1-RDMs associated with N-particles Slater states (Grassmann manifold $\mathrm{Gr}(N,L)$)	(3)
$\mathrm{CH}(\mathcal{D})$	Convex hull of $\mathcal{D}$ (set of mixed-state 1-RDMs with N particles)	(4)
D_0	N-particle round-state 1-RDM of $\widehat{H}_0$	(24)
$D_\alpha^{\mathrm{exact}}$	N-particle ground-state 1-RDM of $\widehat{H}_\alpha$	
D_α^{HF}	Hartree–Fock N-particle ground state 1-RDM of $\widehat{H}_\alpha$	
$\mathcal{E}^{\mathrm{HF}}$	Hartree–Fock energy functional	(20)
J and K	Coulomb and exchange energy functionals	(21)
$h^{\mathrm{HF}}(D)$	Mean-field (Fock) Hamiltonian (op. on $\mathcal{H}$)	(25)
N_f	Number of fragments	
L_x	Number of sites in fragment x	
X_x	x-th fragment subspace, $X_x = \mathrm{Span}(e_\kappa, \kappa \in \mathcal{I}_x) \subset \mathcal{H}$	(6)
Π_x	Orthogonal projector on X_x (op. on $\mathbb{R}^{L\times L}_{\mathrm{sym}}$)	
E_x	Matrix of the L_x orbitals of fragment x ($E_x \in \mathbb{R}^{L\times L_x}$)	(27)
Bd	Projector defined by $\mathrm{Bd}(M) = \sum_{x=1}^{N_f} \Pi_{X_x} M \Pi_{X_x}$ (op. on $\mathbb{R}^{L\times L}_{\mathrm{sym}}$)	(7)
$\mathcal{P}$	Convex set of block-diagonal matrices with eigenvalues in [0,1]	(8)
$\mathcal{Y}$	Space of traceless block-diagonal matrices $\mathcal{Y} \subset \mathbb{R}^{L\times L}_{\mathrm{sym}}$	(9)
$W_{x,D}$	x-th impurity space, subspace of $\mathcal{H}$, $W_{x,D} = X_x + DX_x \subset \mathcal{H}$	(10)
$C^x(D)$, $\widetilde{C}^x(D)$	Matrices in $\mathbb{R}^{L\times 2L_x}$ defining orthonormal bases of $W_{x,D}$	(28), (29)
$\widehat{H}^{\mathrm{imp}}_{x,D}$	x-th impurity Hamiltonian (op. on $\mathrm{Fock}(W_{x,D})$)	(14), (30)
R	4-point DMET linear response function (op. on $\mathcal{Y}$)	(26), (75)
F^{LL}, resp. F^{LL}_α	Low-level map for $\widehat{H}$, resp. $\widehat{H}_\alpha$	(19)
F^{HL}, resp. F^{HL}_α	High-level map for $\widehat{H}$, resp. $\widehat{H}_\alpha$	(18), (66)
μ	DMET global chemical potential	

APPENDIX B: ANALYSIS OF THE DMET BIFURCATION FOR H_6^{4-}

We shall finally proceed with the analysis of the DMET solutions along the two bifurcation paths for H_6^{4-} around Θ_3 (see Section 4.2). To begin with, we calculate the molecular orbitals at Θ_3. The molecular orbital energies exhibit two-fold degeneracies resulting from the fact that the E' and E'' are irreducible representations of the H_6^{4-} symmetry point group (D_{3h}) are two-dimensional. For a visual representation of the molecular orbital energies and their corresponding molecular orbitals, see Figure B1a,b.



(a) Occupied orbitals (b) Virtual orbitals

FIGURE B1 Depiction of the molecular orbitals, their irreducible representation with respect to the D_{3h} point group symmetry and molecular energies. The left panel shows the occupied molecular orbitals and the right panel shows the virtual molecular orbitals.

For the two solutions on the respective bifurcation branches, P_0 and P_1, we compute

$$P_0(\Theta) - P_1(\Theta) = (\Theta - \Theta_3)\left[\begin{array}{c|c} 0 & Q_{+-} \\ \hline Q_{-+} & 0 \end{array}\right] + o(\Theta - \Theta_3), \tag{B1}$$

where $Q_{-+} = Q_{+-}^{\top} \in \mathbb{R}^{7\times5}$. From the matrix Q_{-+} we deduce "excitation" patterns that give physical insight into the different branches. The numerical values of Q_{-+} are given by

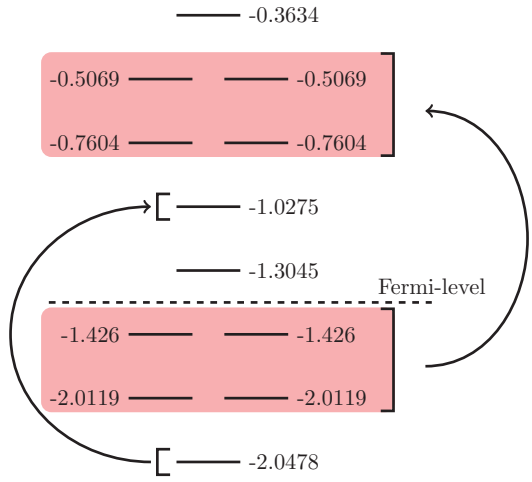


FIGURE B2 Molecular energies and "excitation" patterns concluded from Q_{-+}.

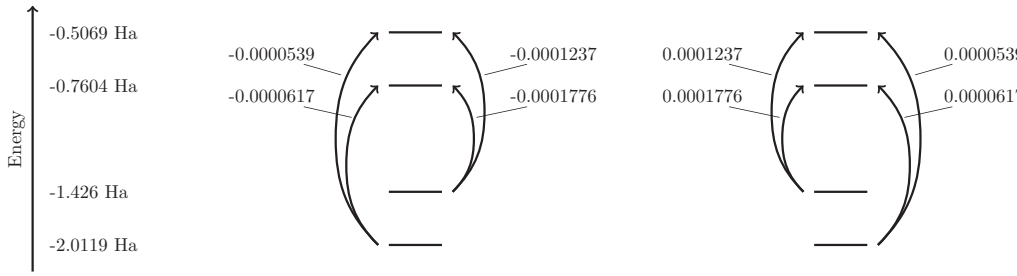


FIGURE B3 Excitation patterns concluded from Q_{-+} for symmetric and anti-symmetric molecular orbitals respectively.

$$Q_{-+} = \begin{bmatrix} 0 & 0 & 0 & 0 & 0 \\ -0.0004 & 0 & 0 & 0 & 0 \\ 0 & 0.0001 & 0 & 0.0002 & 0 \\ 0 & 0 & -0.0001 & 0 & -0.0002 \\ 0 & 0.0001 & 0 & 0.0001 & 0 \\ 0 & 0 & -0.0001 & 0 & -0.0001 \\ 0 & 0 & 0 & 0 & 0 \end{bmatrix} \tag{B2}$$

Upon inspecting Q_{-+}, we observe the following "excitation" pattern: The first molecular orbital (A_1' symmetry) is rotated in the direction of the seventh molecular orbital (A_1' symmetry), while the 4-dimensional space generated by the second to fifth molecular orbitals (E' symmetry) is tilted according to directions which are linear combinations of the eighth to eleventh molecular orbitals (E' symmetry). We summarize this "excitation" pattern in Figure B2.

We see that the pair of degenerate occupied orbitals are excited into the pair of degenerate virtual orbitals. This block of excitations is highlighted by the red shaded area in Figure B2. A more detailed depiction of the excitations between the red-shaded areas is given in Figure B3.