7 Exact Diagonalization and Lanczos Method

Erik Koch
Jülich Supercomputer Centre
Forschungszentrum Jülich

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1 Many-electron states

One of the great surprises of quantum mechanics is the existence of indistinguishable objects. Classically this is not possible: objects can always be distinguished at least by their position in space, meaning that indistinguishable objects must be identical. This is Leibniz' Principle of the Identity of Indiscernibles [1]. For quantum objects, however, the uncertainty principle makes the distinction of particles by their position impossible. This allows for the existence of elementary particles. They form the basic units of all matter. So, quite remarkably, all the different objects we know are made of indistinguishable building blocks.

In the formalism of quantum mechanics, indistinguishability means that no observable lets us distinguish one of these particles from the other. This means that every observable for, e.g., electrons, must treat each electron in the same way. Thus, in principle, observables must act on all electrons in the universe. In practice we can, of course, distinguish electrons localized on the moon from those in our lab to an excellent approximation. Thus, for all practical purposes, we can restrict our description to the electrons in the system under consideration, assuming that the differential overlap with all other electrons vanishes. Any observable $M(x_1,\dots,x_N)$ for the N electrons in our system must then be symmetric under permutations of the variables x_i.

The consequences are straightforward: An observable $M(x)$ acting on a single-particle degree of freedom x must act on all indistinguishable particles in the same way, i.e., $\sum_i M(x_i)$. Likewise, a two-body observable $M(x,x')$ must act on all pairs in the same way, $\sum_{i,j} M(x_i,x_j)$ with $M(x,x') = M(x',x)$. We can thus write any observable in the form

$$M(\mathbf{x}) = M^{(0)} + \sum_i M^{(1)}(x_i) + \frac{1}{2!}\sum_{i\neq j} M^{(2)}(x_i,x_j) + \frac{1}{3!}\sum_{i\neq j\neq k} M^{(3)}(x_i,x_j,x_k) + \cdots \tag{1}$$

$$= M^{(0)} + \sum_i M^{(1)}(x_i) + \sum_{i<j} M^{(2)}(x_i,x_j) + \sum_{i<j<k} M^{(3)}(x_i,x_j,x_k) + \cdots, \tag{2}$$

where the summations can be restricted since the operators must be symmetric in their arguments, while for two or more identical coordinates the operator is really one of lower order: $M^{(2)}(x_i,x_i)$, e.g., only acts on a single coordinate and should be included in $M^{(1)}$.

For the many-body wave functions $\Psi(x_1,x_2,\cdots)$ the situation is slightly more complex. Since the probability density $|\Psi(x_1,x_2,\cdots)|^2$ is an observable, the wave function should transform as one-dimensional (irreducible) representations of the permutation group. Which irreducible representation applies to a given type of elementary particle is determined by the spin-statistics theorem [2, 3]: The wave functions of particles with integer spin are symmetric, those of particles with half-integer spin change sign when two arguments are exchanged. From an arbitrary N-particle wave function we thus obtain a many-electron wavefunction by antisymmetrizing

$$\mathcal{A}\Psi(x_1,\dots,x_N) := \frac{1}{\sqrt{N!}}\sum_P (-1)^P \Psi\left(x_{p(1)},\dots,x_{p(N)}\right), \tag{3}$$

where $(-1)^P$ is the parity of the permutation P that maps $n \to p(n)$. Since there are $N!$ different permutations, this can easily become an extremely costly operation. Remarkably, a product of

N single-electron states φ_α can be antisymmetrized much more efficiently (in $\mathcal{O}(N^3)$ steps) by writing it in the form of a determinant

$$\Phi_{\alpha_1,\ldots,\alpha_N}(x_1,\ldots,x_N) := \mathcal{A}\,\varphi_{\alpha_1}(x_1)\cdots\varphi_{\alpha_N}(x_N) = \frac{1}{\sqrt{N!}}\begin{vmatrix} \varphi_{\alpha_1}(x_1) & \varphi_{\alpha_2}(x_1) & \cdots & \varphi_{\alpha_N}(x_1) \\ \varphi_{\alpha_1}(x_2) & \varphi_{\alpha_2}(x_2) & \cdots & \varphi_{\alpha_N}(x_2) \\ \vdots & \vdots & \ddots & \vdots \\ \varphi_{\alpha_1}(x_N) & \varphi_{\alpha_2}(x_N) & \cdots & \varphi_{\alpha_N}(x_N) \end{vmatrix}. \tag{4}$$

For $N{=}1$ the Slater determinant is simply the one-electron orbital $\Phi_\alpha(x) = \varphi_\alpha(x)$ while for $N{=}2$ we get the familiar expression $\Phi_{\alpha,\alpha'}(x,x') = \big(\varphi_\alpha(x)\varphi_{\alpha'}(x') - \varphi_{\alpha'}(x)\varphi_\alpha(x')\big)/\sqrt{2}$ for the two-electron Slater determinant.

Slater determinants are important because they can be used to build a basis of the many-electron Hilbert space. To see how, we consider a complete set of orthonormal single-electron states

$$\sum_n \overline{\varphi_n(x)}\,\varphi_n(x') = \delta(x-x') \text{ (complete)} \qquad \int dx\,\overline{\varphi_n(x)}\,\varphi_m(x) = \delta_{n,m} \text{ (orthonormal)}\,. \tag{5}$$

To expand an arbitrary N-particle function $a(x_1,\ldots,x_N)$, we start by considering it as a function of x_1 with $x_2,\ldots,x_N$ kept fixed. We can then expand it in the complete set $\{\varphi_n\}$ as

$$a(x_1,\ldots,x_N) = \sum_{n_1} a_{n_1}(x_2,\ldots,x_N)\,\varphi_{n_1}(x_1)$$

with expansion coefficients that depend on the remaining coordinates

$$a_{n_1}(x_2,\ldots,x_N) = \int dx_1\;\overline{\varphi_{n_1}(x_1)}\,a(x_1,x_2,\ldots,x_N)\,.$$

These, in turn, can be expanded as a functions of x_2

$$a_{n_1}(x_2,\ldots,x_N) = \sum_{n_2} a_{n_1,n_2}(x_3,\ldots,x_N)\,\varphi_{n_2}(x_2)\,.$$

Repeating this, we obtain the expansion of a in product states

$$a(x_1,\ldots,x_N) = \sum_{n_1,\ldots,n_N} a_{n_1,\ldots,n_N}\,\varphi_{n_1}(x_1)\cdots\varphi_{n_N}(x_N)\,.$$

When the N-particle function is antisymmetric, applying the antisymmetrizer (3) will leave it unchanged, i.e., the expansion coefficients will be antisymmetric under permutation of the indices: $a_{n_{p(1)},\ldots,n_{p(N)}} = (-1)^P a_{n_1,\ldots,n_N}$. Fixing some particular order of the indices, e.g., $n_1 < n_2 < \ldots < n_N$, we thus get an expansion in Slater determinants

$$\Psi(x_1,\ldots,x_N) = \sum_{n_1<\ldots<n_N} a_{n_1,\ldots,n_N}\sqrt{N!}\,\Phi_{n_1,\ldots,n_N}(x_1,\ldots,x_N)\,.$$

Since we can write any antisymmetric function as such a configuration-interaction expansion, the set of Slater determinants

$$\Big\{\Phi_{n_1,\ldots,n_N}(x_1,\ldots,x_N)\;\Big|\;n_1<n_2<\cdots<n_N\Big\} \tag{6}$$

forms a basis of the N-electron Hilbert space. Since the overlap of two Slater determinants

$$\int d\boldsymbol{x}\, \overline{\Phi_{\alpha_1,\dots,\alpha_N}(\boldsymbol{x})}\, \Phi_{\beta_1,\dots,\beta_N}(\boldsymbol{x}) = \frac{1}{N!}\sum_{P,P'}(-1)^{P+P'}\prod_n \int dx_n\, \overline{\varphi_{\alpha_{p(n)}}(x_n)}\, \varphi_{\alpha_{p'(n)}}(x_n)$$
$$= \begin{vmatrix} \langle\varphi_{\alpha_1}|\varphi_{\beta_1}\rangle & \cdots & \langle\varphi_{\alpha_1}|\varphi_{\beta_N}\rangle \\ \vdots & \ddots & \vdots \\ \langle\varphi_{\alpha_N}|\varphi_{\beta_1}\rangle & \cdots & \langle\varphi_{\alpha_N}|\varphi_{\beta_N}\rangle \end{vmatrix} \tag{7}$$

is the determinant of the overlap of the constituent orbitals, the Slater determinants (6) form a complete orthonormal basis of the N-electron Hilbert space when the orbitals $\varphi_n(x)$ are a complete orthonormal basis of the one-electron Hilbert space.

While we use a set of N one-electron orbitals $\varphi_n(x)$ to define an N-electron Slater determinant $\Phi_{\alpha_1,\dots,\alpha_N}(\boldsymbol{x})$, this representation is not unique: Any unitary transformation among the N occupied orbitals will not change the determinant. Thus, strictly, a Slater determinant is not determined by the set of indices we usually give, but, up to a phase, by the N-dimensional subspace spanned by the orbitals $\varphi_1, \dots, \varphi_N$ in the single-electron Hilbert space. The projector to this space is the one-body density matrix

$$\Gamma^{(1)}(x,x') = N\int dx_2\cdots dx_N\, \overline{\Phi(x,x_2,\dots,x_N)}\,\Phi(x',x_2,\dots,x_N)\,. \tag{8}$$

To see this, we expand the Slater determinant along its first row

$$\Phi_{\alpha_1\cdots\alpha_N}(x_1,\dots,x_N) = \frac{1}{\sqrt{N}}\sum_{n=1}^{N}(-1)^{1+n}\,\varphi_{\alpha_n}(x_1)\,\Phi_{\alpha_{i\neq n}}(x_2,\dots,x_N)\,, \tag{9}$$

where $\Phi_{\alpha_{i\neq n}}(x_2,\dots,x_N)$ is the determinant with the first row and the n-th column removed, which can be written as $N-1$-electron Slater determinants with orbital α_n removed. Inserting this into (8) we find

$$\Gamma^{(1)}_{\Phi}(x,x') = \sum_{n=1}^{N}\overline{\varphi_{\alpha_n}(x)}\,\varphi_{\alpha_n}(x')\,, \tag{10}$$

which is the expansion of the one-body density matrix in eigenfunctions (natural orbitals), with eigenvalues (natural occupation numbers) either one or zero. Any many-electron wave function $\Psi(\boldsymbol{x})$ with the same one-body density matrix $\Gamma^{(1)}_{\Phi}$ equals the Slater determinant $\Phi(\boldsymbol{x})$ up to a phase, i.e., $|\langle\Psi|\Phi\rangle| = 1$.

We can generalize this procedure and calculate higher order density matrices by introducing the generalized Laplace expansion

$$\Phi_{\alpha_1\cdots\alpha_N}(\mathbf{x}) = \frac{1}{\sqrt{\binom{N}{p}}}\sum_{n_1<\cdots<n_p}(-1)^{1+\sum_i n_i}\,\Phi_{\alpha_{n_1}\cdots\alpha_{n_p}}(x_1,\dots,x_p)\Phi_{\alpha_{i\notin\{n_1,\dots,n_p\}}}(x_{p+1},\dots,x_N),$$

which is obtained by writing the permutation of all N indices as a permutation of $N-p$ indices and the remaining p indices separately summing over all distinct sets of p indices. This allows us to evaluate arbitrary matrix elements and higher order density matrices [4]. But as can be seen from the above expansion, the expressions very quickly get quite cumbersome. Fortunately there is a representation that is much better suited to handling antisymmetric wave functions. It is called second quantization.

2 Second quantization

While originally introduced for quantizing the electromagnetic field, we can use the formalism of second quantization just as a convenient way of handling antisymmetric wave functions [5,6]. The idea behind this approach is remarkably simple: When writing Slater determinants in the form (4) we are working in a real-space basis. It is, however, often simpler to consider abstract states: Instead of a wave function $\varphi_\alpha(x)$, we write a Dirac state $|\alpha\rangle$. Second quantization allows us to do the same for Slater determinants.

Let us consider a Slater determinant for two electrons, one in state $\varphi_\alpha(x)$, the other in state $\varphi_\beta(x)$. It is simply the antisymmetrized product of the two states

$$\Phi_{\alpha\beta}(x_1, x_2) = \frac{1}{\sqrt{2}} \big(\varphi_\alpha(x_1)\varphi_\beta(x_2) - \varphi_\beta(x_1)\varphi_\alpha(x_2)\big) . \tag{11}$$

This expression is quite cumbersome because we explicitly specify the coordinates. We can try to get rid of the coordinates by defining a two-particle Dirac state

$$|\alpha, \beta\rangle := \frac{1}{\sqrt{2}} \big(|\alpha\rangle|\beta\rangle - |\beta\rangle|\alpha\rangle\big) .$$

While the expression is somewhat simpler, we still have to keep track of the order of the particles by specifying the position of the kets. The idea of second quantization is to specify the states using operators

$$|\alpha, \beta\rangle = c^\dagger_\beta c^\dagger_\alpha |0\rangle. \tag{12}$$

Now the order of the particles is specified by the order of the operators. To ensure the antisymmetry of the wave function the operators have to change sign when they are reordered

$$|\alpha, \beta\rangle = c^\dagger_\beta c^\dagger_\alpha |0\rangle = -c^\dagger_\alpha c^\dagger_\beta |0\rangle = -|\beta, \alpha\rangle . \tag{13}$$

2.1 Creation and annihilation operators

To arrive at the formalism of second quantization we postulate a set of operators that have certain reasonable properties. We then verify that we can use these operators to represent Slater determinants. But first we consider a few simple states to motivate what properties the new operators ought to have.

To be able to construct many-electron states, we start from the simplest such state: $|0\rangle$ the *vacuum state* with no electron, which we assume to be normalized $\langle 0|0\rangle = 1$. Next we introduce for each single-electron state $|\alpha\rangle$ an operator $c^\dagger_\alpha$ such that $c^\dagger_\alpha|0\rangle = |\alpha\rangle$. We call them *creation operators* since they add an electron (in state α) to the state that they act on: in $c^\dagger_\alpha|0\rangle$ the creation operator adds an electron to the vacuum state (N=0), resulting in a single-electron state (N=1). Applying another creation operator produces a two-electron state $c^\dagger_\beta c^\dagger_\alpha|0\rangle$, ($N$=2). To ensure the antisymmetry of the two electron state, the product of creation operators has to change sign when they are reordered: $c^\dagger_\alpha c^\dagger_\beta = -c^\dagger_\beta c^\dagger_\alpha$. This is more conveniently written as $\{c^\dagger_\alpha,\, c^\dagger_\beta\} = 0$ by introducing the *anti-commutator*

$$\{A,\, B\} := A\,B + B\,A . \tag{14}$$

As we have seen, the simplest state we can produce with the creation operators is the single-electron state $|\alpha\rangle = c_\alpha^\dagger|0\rangle$. When we want to calculate its norm, we have to consider the adjoint of $c_\alpha^\dagger|0\rangle$, formally obtaining $\langle\alpha|\alpha\rangle = \langle 0|c_\alpha c_\alpha^\dagger|0\rangle$, or, more generally, $\langle\alpha|\beta\rangle = \langle 0|c_\alpha c_\beta^\dagger|0\rangle$. This implies that c_α, the adjoint of a creation operator, must remove an electron from the state, otherwise the overlap of $c_\alpha c_\beta^\dagger|0\rangle$ with the vacuum state $\langle 0|$ would vanish. We therefore call the adjoint of the creation operator an *annihilation operator*. We certainly cannot take an electron out of the vacuum state, so $c_\alpha|0\rangle = 0$. To obtain the overlap of one-electron states we postulate the anticommutation relation $\{c_\alpha, c_\beta^\dagger\} = \langle\alpha|\beta\rangle$, giving $\langle 0|c_\alpha c_\beta^\dagger|0\rangle = \langle 0|\{c_\alpha,\, c_\beta^\dagger\} - c_\beta^\dagger c_\alpha|0\rangle = \langle\alpha|\beta\rangle$. For completeness, taking the adjoint of the anticommutation relation for the creation operators, we obtain the corresponding anticommutator of the annihilators: $\{c_\alpha,\, c_\beta\} = 0$.
Thus, we are led to define the vacuum state $|0\rangle$ and the set of operators c_α related to single-electron states $|\alpha\rangle$ with the properties

$$\boxed{\begin{aligned} c_\alpha|0\rangle &= 0 \qquad & \big\{c_\alpha,\, c_\beta\big\} &= 0 = \big\{c_\alpha^\dagger,\, c_\beta^\dagger\big\} \\ \langle 0|0\rangle &= 1 \qquad & \big\{c_\alpha,\, c_\beta^\dagger\big\} &= \langle\alpha|\beta\rangle \end{aligned}} \tag{15}$$

As a direct consequence we obtain the Pauli principle in the form $c_\alpha c_\alpha = 0 = c_\alpha^\dagger c_\alpha^\dagger$.
We note that the creators transform in the same way as the single-electron states they represent

$$|\tilde\alpha_i\rangle = \sum_\mu |\alpha_\mu\rangle\, U_{\mu i} \qquad \rightsquigarrow \quad \tilde c_{\tilde\alpha_i}^\dagger|0\rangle = \sum_\mu c_{\alpha_\mu}^\dagger|0\rangle\, U_{\mu i} = \left(\sum_\mu c_{\alpha_\mu}^\dagger U_{\mu i}\right)|0\rangle. \tag{16}$$

The creators and annihilators are clearly not operators in a Hilbert space, but transfer states from an N-electron to an $N\pm1$-electron Hilbert space, i.e., they are operators defined on *Fock space*. It is also remarkable that the mixed anti-commutator is the only place where the orbitals that distinguish different operators enter.
To make contact with the notation of first quantization, we introduce the *field operators* $\hat\Psi^\dagger(x)$, with $x = (r,\sigma)$, that create an electron of spin σ at position r, i.e., in state $|x\rangle = |r,\sigma\rangle$. Given a complete, orthonormal set of orbitals $\{\varphi_n\}$, we can expand $|x\rangle$

$$\hat\Psi^\dagger(x)|0\rangle = |x\rangle = \sum_n |\varphi_n\rangle\langle\varphi_n|x\rangle = \sum_n c_{\varphi_n}^\dagger|0\rangle\langle\varphi_n|x\rangle \tag{17}$$

from which we obtain

$$\hat\Psi^\dagger(x) = \sum_n \overline{\langle x|\varphi_n\rangle}\, c_{\varphi_n}^\dagger = \sum_n \overline{\varphi_n(x)}\, c_{\varphi_n}^\dagger. \tag{18}$$

The anticommutators then follow from (15) for an orthonormal and complete set, e.g.,

$$\big\{\hat\Psi(x),\, \hat\Psi^\dagger(x')\big\} = \sum_{n,m}\langle x|\varphi_n\rangle \underbrace{\big\{c_{\varphi_n},\, c_{\varphi_m}^\dagger\big\}}_{=\delta_{n,m}}\langle\varphi_m|x'\rangle = \sum_n\langle x|\varphi_n\rangle\langle\varphi_n|x'\rangle = \langle x|x'\rangle = \delta(x-x'),$$

resulting in the anticommutation relations for the field operators

$$\big\{\hat\Psi(x),\, \hat\Psi(x')\big\} = 0 = \big\{\hat\Psi^\dagger(x),\, \hat\Psi^\dagger(x')\big\} \quad \text{and} \quad \big\{\hat\Psi(x),\, \hat\Psi^\dagger(x')\big\} = \langle x|x'\rangle. \tag{19}$$

We can, of course, expand the field operators also in a non-orthogonal set of orbitals $\{|\chi_i\rangle\}$, as long as it is complete, $\sum_{i,j} |\chi_i\rangle (S^{-1})_{ij} \langle\chi_j| = \mathbb{1}$, where $S_{ij} = \langle\chi_i|\chi_j\rangle$ is the overlap matrix,

$$\hat{\Psi}^\dagger(x) = \sum_{i,j} c_i^\dagger \, (S^{-1})_{ij} \, \langle\chi_j|x\rangle. \tag{20}$$

Conversely, given any single-electron wave functions in real space $\varphi(x)$, we can express the corresponding creation operator in terms of the field operators

$$c_\varphi^\dagger = \int dx \, \varphi(x) \, \hat{\Psi}^\dagger(x). \tag{21}$$

Its anticommutator with the field annihilator just gives back the single-electron wave function

$$\left\{\hat{\Psi}(x), \, c_\varphi^\dagger\right\} = \int dx' \, \varphi(x') \left\{\hat{\Psi}(x), \, \hat{\Psi}^\dagger(x')\right\} = \varphi(x) \, . \tag{22}$$

2.2 Representation of Slater determinants

We have now all the tools in place to write the Slater determinant (4) in second quantization, using the creation operators to specify the occupied orbitals and the field operators to give the coordinates for the real-space representation:

$$\Phi_{\alpha_1\alpha_2\ldots\alpha_N}(x_1, x_2, \ldots, x_N) = \frac{1}{\sqrt{N!}} \left\langle 0\right| \hat{\Psi}(x_1)\hat{\Psi}(x_2)\ldots\hat{\Psi}(x_N) \;\; c_{\alpha_N}^\dagger \ldots c_{\alpha_2}^\dagger c_{\alpha_1}^\dagger \left|0\right\rangle. \tag{23}$$

Note how writing the Slater determinant as an expectation value of annihilation and creation operators nicely separates the coordinates on the left from the orbitals on the right. This is just the desired generalization of the Dirac notation $\varphi(x) = \langle x|\varphi\rangle$.

Not surprisingly, the proof of (23) is by induction. As a warm-up we consider the case of a single-electron wave function (N=1). Using the anticommutation relation (22), we see that

$$\left\langle 0\right| \hat{\Psi}(x_1) \, c_{\alpha_1}^\dagger \left|0\right\rangle = \left\langle 0\right| \varphi_{\alpha_1}(x_1) - c_{\alpha_1}^\dagger \hat{\Psi}(x_1) \left|0\right\rangle = \varphi_{\alpha_1}(x_1) \, . \tag{24}$$

For the two-electron state $N = 2$, we anticommute $\hat{\Psi}(x_2)$ in two steps to the right

$$\begin{aligned} \left\langle 0\right| \hat{\Psi}(x_1)\hat{\Psi}(x_2) \, c_{\alpha_2}^\dagger c_{\alpha_1}^\dagger \left|0\right\rangle &= \left\langle 0\right| \hat{\Psi}(x_1) \big(\varphi_{\alpha_2}(x_2) - c_{\alpha_2}^\dagger \hat{\Psi}(x_2)\big) c_{\alpha_1}^\dagger \left|0\right\rangle \\ &= \left\langle 0\right| \hat{\Psi}(x_1) c_{\alpha_1}^\dagger \left|0\right\rangle \varphi_{\alpha_2}(x_2) - \left\langle 0\right| \hat{\Psi}(x_1) c_{\alpha_2}^\dagger \hat{\Psi}(x_2) c_{\alpha_1}^\dagger \left|0\right\rangle \\ &= \varphi_{\alpha_1}(x_1)\varphi_{\alpha_2}(x_2) - \varphi_{\alpha_2}(x_1)\varphi_{\alpha_1}(x_2) \, . \end{aligned} \tag{25}$$

We see how anticommuting automatically produces the appropriate signs for the antisymmetric wave function. Dividing by $\sqrt{2}$, we obtain the desired two-electron Slater determinant.

The general case of an N-electron state works just the same. Anti-commuting $\hat{\Psi}(x_N)$ all the way to the right produces $N-1$ terms with alternating sign

$$\begin{aligned} \left\langle 0\right| \hat{\Psi}(x_1) \cdots \hat{\Psi}(x_{N-1})\hat{\Psi}(x_N) \, c_{\alpha_N}^\dagger c_{\alpha_{N-1}}^\dagger \cdots c_{\alpha_1}^\dagger \left|0\right\rangle \; = \\ + \quad \left\langle 0\right| \hat{\Psi}(x_1) \cdots \hat{\Psi}(x_{N-1}) \, c_{\alpha_{N-1}}^\dagger \cdots c_{\alpha_1}^\dagger \left|0\right\rangle \quad \varphi_{\alpha_N}(x_N) \\ - \quad \left\langle 0\right| \hat{\Psi}(x_1) \cdots \hat{\Psi}(x_{N-1}) \textstyle\prod_{n\neq N-1} c_{\alpha_n}^\dagger \left|0\right\rangle \quad \varphi_{\alpha_{N-1}}(x_N) \\ \vdots \\ (-1)^{N-1} \quad \left\langle 0\right| \hat{\Psi}(x_1) \cdots \hat{\Psi}(x_{N-1}) \, c_{\alpha_N}^\dagger \quad \cdots \quad c_{\alpha_2}^\dagger \left|0\right\rangle \quad \varphi_{\alpha_1}(x_N) \, . \end{aligned}$$

Using (23) for the $N-1$-electron states, this is nothing but the Laplace expansion of

$$D = \begin{vmatrix} \varphi_{\alpha_1}(x_1) & \varphi_{\alpha_2}(x_1) & \cdots & \varphi_{\alpha_N}(x_1) \\ \varphi_{\alpha_1}(x_2) & \varphi_{\alpha_2}(x_2) & \cdots & \varphi_{\alpha_N}(x_2) \\ \vdots & \vdots & \ddots & \vdots \\ \varphi_{\alpha_1}(x_N) & \varphi_{\alpha_2}(x_N) & \cdots & \varphi_{\alpha_N}(x_N) \end{vmatrix}$$

along the Nth row. Dividing by $\sqrt{N!}$ we see that we have shown (23) for N-electron states, completing the proof by induction.
Given this representation of Slater determinants it is easy to eliminate the coordinates so we can work with N-electron states rather than N-electron wave functions—just as in Dirac notation. In particular we can rewrite the basis of Slater determinants (6) into a basis of product states

$$\left\{ c^\dagger_{n_N} \cdots c^\dagger_{n_1} |0\rangle \;\middle|\; n_1 < \cdots < n_N \right\}, \tag{26}$$

which allows us to express any N-electron state as

$$|\Psi\rangle = \sum_{n_1<\cdots<n_N} a_{n_1,\ldots,n_N}\, c^\dagger_{n_N} \cdots c^\dagger_{n_1} |0\rangle. \tag{27}$$

2.3 Representation of n-body operators

To work with N-electron states rather than Slater determinants, we also have to rewrite the N-electron operators $M(\boldsymbol{x})$ appropriately. This is easily done by incorporating the coordinates that we have separated from the Slater determinants into the operators such that the expectation values remain unchanged. This is, again, analogous to the Dirac formalism:

$$\int dx\, \overline{\varphi_n(x)}\, M(x)\, \varphi_m(x) = \langle \varphi_n | \underbrace{\int dx\, |x\rangle M(x) \langle x|}_{=:\hat{M}} \varphi_m \rangle = \langle \varphi_n | \hat{M} | \varphi_m \rangle. \tag{28}$$

For N-electron Slater determinants this becomes

$$\begin{aligned} &\int dx_1 \cdots dx_N\, \overline{\Phi_{\beta_1\cdots\beta_N}(x_1,\cdots,x_N)}\, M(x_1,\ldots,x_N)\, \Phi_{\alpha_1\cdots\alpha_N}(x_1,\cdots,x_N) \\ =&\int dx_1 \cdots dx_N \langle 0| c_{\beta_1} \cdots c_{\beta_N} \hat{\Psi}^\dagger(x_N) \cdots \hat{\Psi}^\dagger(x_1) |0\rangle M(x_1,\ldots,x_N) \langle 0| \hat{\Psi}(x_1) \cdots \hat{\Psi}(x_N) c^\dagger_{\alpha_N} \cdots c^\dagger_{\alpha_1} |0\rangle \\ &= \left\langle 0 \right| c_{\beta_1} \cdots c_{\beta_N}\, \hat{M}\, c^\dagger_{\alpha_N} \cdots c^\dagger_{\alpha_1} \left| 0 \right\rangle \end{aligned}$$

with the representation of the n-body operator in terms of field operators

$$\hat{M} := \frac{1}{N!} \int dx_1 \cdots x_N\, \hat{\Psi}^\dagger(x_N) \cdots \hat{\Psi}^\dagger(x_1)\, M(x_1,\cdots,x_N)\, \hat{\Psi}(x_1) \cdots \hat{\Psi}(x_N)\,. \tag{29}$$

Note that this particular form of the operator is only valid when applied to N-electron states, since we have used that the N annihilation operators bring us to the zero-electron space, where $|0\rangle\langle 0| = \mathbb{1}$. Keeping this in mind, we can work entirely in terms of our algebra (15).

To see what (29) means, we look, in turn, at the different n-body parts of $M(\boldsymbol{x})$, (2):

$$M(\mathbf{x}) = M^{(0)} + \sum_i M^{(1)}(x_i) + \sum_{i<j} M^{(2)}(x_i, x_j) + \sum_{i<j<k} M^{(3)}(x_i, x_j, x_k) + \cdots \tag{30}$$

We start with the simplest case, the zero-body operator, which, up to a trivial prefactor, is $M^{(0)} = 1$. Operating on an N-electron wave function, it gives

$$\begin{aligned}
\hat{M}^{(0)} &= \frac{1}{N!} \int dx_1 dx_2 \cdots x_N \, \hat{\Psi}^\dagger(x_N) \cdots \hat{\Psi}^\dagger(x_2) \hat{\Psi}^\dagger(x_1) \, \hat{\Psi}(x_1) \hat{\Psi}(x_2) \cdots \hat{\Psi}(x_N) \\
&= \frac{1}{N!} \int \quad dx_2 \cdots x_N \, \hat{\Psi}^\dagger(x_N) \cdots \hat{\Psi}^\dagger(x_2) \qquad \hat{N} \qquad \hat{\Psi}(x_2) \cdots \hat{\Psi}(x_N) \\
&= \frac{1}{N!} \int \quad dx_2 \cdots x_N \, \hat{\Psi}^\dagger(x_N) \cdots \hat{\Psi}^\dagger(x_2) \qquad 1 \qquad \hat{\Psi}(x_2) \cdots \hat{\Psi}(x_N) \\
&\;\;\vdots \\
&= \frac{1}{N!} \, 1 \cdot 2 \, \cdots \, N = 1 \,,
\end{aligned} \tag{31}$$

where we have used that the operator

$$\int dx \, \hat{\Psi}^\dagger(x) \hat{\Psi}(x) = \hat{N}$$

counts the number of electrons: Applied to the vacuum state it gives $\hat{N} \, |0\rangle = 0$, while its commutator with any creation operator produces that operator

$$[\hat{N}, \, c_n^\dagger] = \int dx \, [\hat{\Psi}^\dagger(x) \hat{\Psi}(x), \, c_n^\dagger] = \int dx \, \hat{\Psi}^\dagger(x) \, \{\hat{\Psi}(x), \, c_n^\dagger\} = \int dx \, \hat{\Psi}^\dagger(x) \, \varphi_n(x) = c_n^\dagger. \tag{32}$$

where we have used the simple relation $[AB, \, C] = A\{B, \, C\} - \{A, \, C\}B$. Commuting with an annihilator we pick up a minus sign $[\hat{N}, \, \hat{\Psi}(x')] = -\hat{\Psi}(x')$. Thus, commuting $\hat{N}$ through a general product state, we obtain for each creation operator that we encounter a copy of the state, while for each annihilator we obtain minus that state, giving in total the original state times the difference in the number of creation and annihilation operators.

Remarkably, while we started from an operator acting on N-electron states, the resulting operator in second quantized form is independent of the number of electrons. We will see that this is an important general feature of operators in second quantization which makes working in Fock spaces amazingly simple.

We note that (31) just means that the overlap of two Slater determinants (7) is equal to that of the corresponding product states

$$\int d\boldsymbol{x} \, \overline{\Phi_{\alpha_1,\ldots,\alpha_N}(\boldsymbol{x})} \, \Phi_{\beta_1,\ldots,\beta_N}(\boldsymbol{x}) = \big\langle 0 \big| \, c_{\alpha_1} \cdots c_{\alpha_N} \, c_{\beta_N}^\dagger \cdots c_{\beta_1}^\dagger \big| 0 \big\rangle. \tag{33}$$

2.3.1 One-body operators

Next we consider one-body operators $\sum_j M^{(1)}(x_j)$

$$\begin{aligned}\hat{M}^{(1)} &= \frac{1}{N!}\int dx_1\cdots dx_N\,\hat\Psi^\dagger(x_N)\cdots\hat\Psi^\dagger(x_1)\sum_j M^{(1)}(x_j)\;\hat\Psi(x_1)\cdots\hat\Psi(x_N)\\ &= \frac{1}{N!}\sum_j\int dx_j\,\hat\Psi^\dagger(x_j)\,M^{(1)}(x_j)\,(N{-}1)!\,\hat\Psi(x_j)\\ &= \frac{1}{N}\sum_j\int dx_j\,\hat\Psi^\dagger(x_j)\,M^{(1)}(x_j)\,\hat\Psi(x_j)\\ &= \int dx\quad\hat\Psi^\dagger(x)\;\;M^{(1)}(x)\;\;\hat\Psi(x)\end{aligned}\tag{34}$$

Here we have first anticommuted $\hat\Psi^\dagger(x_j)$ all the way to the left and $\hat\Psi(x_j)$ to the right. Since these take the same numbers of anticommutations, there is no sign involved. In between these field operators we are left with a zero-body operator for $N{-}1$ electrons, producing, when $\hat M^{(1)}$ acts on an N-electron state, a factor of $(N{-}1)!$. Again we notice that we obtain an operator that no longer depends on the number of electrons, i.e., that is valid in the entire Fock space.
Expanding the field-operators in a complete orthonormal set $\hat\Psi(x)=\sum_n\varphi_n(x)\,c_n$ gives

$$\hat M^{(1)} = \sum_{n,m}\int dx\,\overline{\varphi_n(x)}\,M(x)\,\varphi_m(x)\;c_n^\dagger c_m = \sum_{n,m}\langle\varphi_n|M^{(1)}|\varphi_m\rangle\;c_n^\dagger c_m = \sum_{n,m} c_n^\dagger\,M^{(1)}_{nm}\,c_m.\tag{35}$$

The matrix elements $M^{(1)}_{nm}=\langle\varphi_n|M^{(1)}|\varphi_m\rangle$ transform like a single-electron matrix $\boldsymbol{M}^{(1)}$: From (16) and writing the annihilation operators as a column vector $\boldsymbol{c}$ we see that

$$\hat M^{(1)} = \boldsymbol{c}^\dagger\,\boldsymbol{M}^{(1)}\,\boldsymbol{c}\;\; = \boldsymbol{c}^\dagger\boldsymbol{U}^\dagger\,\boldsymbol{U}\boldsymbol{M}^{(1)}\boldsymbol{U}^\dagger\,\boldsymbol{U}\boldsymbol{c} = \tilde{\boldsymbol{c}}^\dagger\,\tilde{\boldsymbol{M}}^{(1)}\,\tilde{\boldsymbol{c}}\,.\tag{36}$$

Once we have arrived at the representation in terms of orbitals, we can restrict the orbital basis to a non-complete set. This simply gives the operator in the variational (Fock) subspace spanned by the orbitals.
We note that the expression (35) not only works for local operators but also for differential operators like the momentum or kinetic energy: we have taken care not to exchange the order of $M^{(1)}$ and one of its field operators. We can write truly non-local operators in a similar way. As an example, the one-body density operator is given by

$$\hat\Gamma^{(1)}(x;x') = \hat\Psi^\dagger(x)\hat\Psi(x')\tag{37}$$

so that one coordinate is not integrated over, rather setting it to x in the bra and x' in the ket. In an orthonormal basis it becomes

$$\hat\Gamma^{(1)}(x;x') = \sum_{n,m}\overline{\varphi_n(x)}\,\varphi_m(x')\,c_n^\dagger c_m\,.\tag{38}$$

2.3.2 Two-body operators

For the two-body operators $\sum_{i<j} M^{(2)}(x_i, x_j)$ we proceed in the familiar way, anti-commuting first the operators with the coordinates involved in $M^{(2)}$ all the way to the left and right, respectively. This time we are left with a zero-body operator for $N-2$ electrons:

$$\begin{aligned}
\hat{M}^{(2)} &= \frac{1}{N!} \int dx_1 \cdots dx_N \, \hat{\Psi}^\dagger(x_N) \cdots \hat{\Psi}^\dagger(x_1) \sum_{i<j} M^{(2)}(x_i, x_j) \, \hat{\Psi}(x_1) \cdots \hat{\Psi}(x_N) \\
&= \frac{1}{N!} \sum_{i<j} \int dx_i dx_j \, \hat{\Psi}^\dagger(x_j) \hat{\Psi}^\dagger(x_i) \, M^{(2)}(x_i, x_j) \, (N-2)! \, \hat{\Psi}(x_i) \hat{\Psi}(x_j) \\
&= \frac{1}{N(N-1)} \sum_{i<j} \int dx_i dx_j \, \hat{\Psi}^\dagger(x_j) \hat{\Psi}^\dagger(x_i) \, M^{(2)}(x_i, x_j) \, \hat{\Psi}(x_i) \hat{\Psi}(x_j) \\
&= \frac{1}{2} \int dx \, dx' \, \hat{\Psi}^\dagger(x') \, \hat{\Psi}^\dagger(x) \;\; M^{(2)}(x, x') \;\; \hat{\Psi}(x) \, \hat{\Psi}(x')
\end{aligned}$$

Expanding in an orthonormal basis, we get

$$\begin{aligned}
\hat{M}^{(2)} &= \frac{1}{2} \sum_{n,n',m,m'} \int dx dx' \, \overline{\varphi_{n'}(x') \varphi_n(x)} \, M^{(2)}(x, x') \, \varphi_m(x) \varphi_{m'}(x') \quad c^\dagger_{n'} c^\dagger_n c_m c_{m'} \\
&= \frac{1}{2} \sum_{n,n',m,m'} \qquad \langle \varphi_n \varphi_{n'} | M^{(2)} | \varphi_m \varphi_{m'} \rangle \qquad c^\dagger_{n'} c^\dagger_n c_m c_{m'} \qquad (39)
\end{aligned}$$

where the exchange of the indices in the second line is a consequence of the way the Dirac state for two electrons is usually written: first index for the first coordinate, second index for the second, while taking the adjoint of the operators changes their order. $M_{nn',\,mm'} = \langle \varphi_n \varphi_{n'} | M^{(2)} | \varphi_m \varphi_{m'} \rangle$ transforms like a fourth-order tensor: Transforming to a different basis (16) gives

$$\tilde{M}^{(2)}_{\nu\nu',\mu\mu'} = \sum_{n,n',m,m'} U^\dagger_{\nu n} U^\dagger_{\nu' n'} M_{nn',mm'} U_{m\mu} U_{m'\mu'} \, . \qquad (40)$$

Form the symmetry of the two-body operator $M^{(2)}(x, x') = M^{(2)}(x', x)$ follows $M_{nn',\,mm'} = M_{n'n,\,m'm}$. Moreover, $M_{nn,\,mm'}$ will not contribute to $\hat{M}^{(2)}$ since $c^\dagger_n c^\dagger_n = \{c^\dagger_n,\, c^\dagger_n\}/2 = 0$, and likewise for $M_{nn',\,mm}$.

Note that the representation (39) is not quite as efficient as it could be: The terms with n and n' and/or m and m' exchanged connect the same basis states. Collecting these terms by introducing an ordering of the operators and using the symmetry of the matrix elements we obtain

$$\hat{M}^{(2)} = \sum_{n'>n,\; m'>m} c^\dagger_{n'} c^\dagger_n \underbrace{\left(M^{(2)}_{nn',\,mm'} - M^{(2)}_{n'n,\,mm'} \right)}_{=: \breve{M}^{(2)}_{nn',\,mm'}} c_m c_{m'} \, . \qquad (41)$$

Since the states $\{c^\dagger_{n'} c^\dagger_n |0\rangle \mid n' > n\}$ form a basis of the two-electron Hilbert space, considering nn' as the index of a basis state, the $\breve{M}^{(2)}_{nn',\,mm'}$ form a two-electron matrix $\breve{\boldsymbol{M}}^{(2)}$.

The procedure of rewriting operators in second quantization obviously generalizes to observables acting on more than two electrons in the natural way. We note that, while we started from

a form of the operators (30) that was explicitly formulated in an N-electron Hilbert space, the results (31), (35), and (39) are of the same form no matter what value N takes. Thus these operators are valid not just on some N-electron Hilbert space, but on the entire Fock space. This is a particular strength of the second-quantized formalism.

2.4 Transforming the orbital basis

We noted in (16) that the creators transform in the same way as the orbitals they represent

$$|\beta_i\rangle = U|\alpha_i\rangle = \sum_j |\beta_j\rangle\langle\alpha_j|\alpha_i\rangle = \sum_\mu |\alpha_\mu\rangle \underbrace{\langle\alpha_\mu|U|\alpha_i\rangle}_{=:U_{\mu i}} \quad \rightsquigarrow \quad c^\dagger_{\beta_i} = \sum_\mu c^\dagger_{\alpha_\mu} U_{\mu i}\,, \tag{42}$$

so the "operators" really transform like states. Writing the transformation matrix as $\boldsymbol{U} = e^{\boldsymbol{M}}$, where $\boldsymbol{M}$ is anti-Hermitian, $\boldsymbol{M}^\dagger = -\boldsymbol{M}$ when $\boldsymbol{U}$ is unitary, but can be any matrix when $\boldsymbol{U}$ is merely invertible, we can write the basis transformation in a form appropriate for operators:

$$c^\dagger_{\beta_i} = e^{\boldsymbol{c}^\dagger \boldsymbol{M} \boldsymbol{c}}\, c^\dagger_{\alpha_\mu}\, e^{-\boldsymbol{c}^\dagger \boldsymbol{M} \boldsymbol{c}}\,. \tag{43}$$

To see this, we use the Baker-Campbell-Hausdorff formula in the form

$$e^{\lambda A} B\, e^{-\lambda A} = B + \lambda\,[A,\,B] + \frac{\lambda^2}{2!}\big[A,\,[A,\,B]\big] + \frac{\lambda^3}{3!}\Big[A,\,\big[A,\,[A,\,B]\big]\Big] + \cdots\,, \tag{44}$$

where the expansion coefficients follow by taking the derivatives of the left hand side at $\lambda = 0$, together with the commutator

$$[c^\dagger_{\alpha_\mu} c_{\alpha_\nu},\, c^\dagger_{\alpha_\kappa}] = c^\dagger_{\alpha_\mu}\,\delta_{\nu,\kappa} \tag{45}$$

from which we obtain for the repeated commutators

$$\Big[\sum_{\mu,\nu} M_{\mu\nu} c^\dagger_{\alpha_\mu} c_{\alpha_\nu},\, \sum_\kappa c^\dagger_{\alpha_\kappa} \left(M^n\right)_{\kappa i}\Big] = \sum_{\mu\nu\kappa} c^\dagger_{\alpha_\mu}\, M_{\mu\nu}\,\delta_{\nu,\kappa} \left(M^n\right)_{\kappa i} = \sum_\mu c^\dagger_{\alpha_\mu} \left(M^{n+1}\right)_{\mu i}\,. \tag{46}$$

To keep the derivation simple, we have chosen to transform an operator from the orthonormal basis that we also used to write the exponential operator. Being linear, the transform works, of course, the same for an arbitrary creation operator.

Using this form of the basis transformation and noticing that $e^{-\boldsymbol{c}^\dagger \boldsymbol{M} \boldsymbol{c}}|0\rangle = |0\rangle$, we immediately see that acting with the exponential of a one-body operator on a product state results in another product state

$$e^{\boldsymbol{c}^\dagger \boldsymbol{M} \boldsymbol{c}} \prod c^\dagger_{\alpha_n} |0\rangle = \prod e^{\boldsymbol{c}^\dagger \boldsymbol{M} \boldsymbol{c}}\, c^\dagger_{\alpha_n} e^{-\boldsymbol{c}^\dagger \boldsymbol{M} \boldsymbol{c}} |0\rangle = \prod c^\dagger_{\beta_n} |0\rangle\,. \tag{47}$$

This is, e.g., used when working in the interaction picture. Anticommutators with transformed operators, (42), are simply $\big\{c_{\alpha_j},\, e^{-\boldsymbol{c}^\dagger \boldsymbol{M} \boldsymbol{c}} c^\dagger_{\alpha_i}\, e^{-\boldsymbol{c}^\dagger \boldsymbol{M} \boldsymbol{c}}\big\} = \langle\alpha_j|e^M|\alpha_i\rangle$.

Annihilation operators, being the adjoint of the creators, transform in just the expected way

$$c_{\beta_i} = e^{-\boldsymbol{c}^\dagger \boldsymbol{M}^\dagger \boldsymbol{c}}\, c_{\alpha_\mu}\, e^{\boldsymbol{c}^\dagger \boldsymbol{M}^\dagger \boldsymbol{c}}\,, \tag{48}$$

which means that for unitary transformations, where $\boldsymbol{M}$ is anti-Hermitian, creators and annihilators transform in the same way. Note that in the imaginary-time formalism the annihilators are, via analytic continuation, chosen to transform in the same way as the creators, making them different from the adjoint of the creators.

3 Exact diagonalization

We have worked, so far, with complete, i.e., infinite bases. This is, of course, not possible in actual computer simulations, where we have to confine ourselves to finite basis sets. Such calculations on subspaces are based on the variational principle.

3.1 Variational principles

The variational principle and the Schrödinger equation are equivalent. Consider the energy expectation value as a wave-function functional

$$E[\Psi] = \frac{\langle\Psi|H|\Psi\rangle}{\langle\Psi|\Psi\rangle}\,. \tag{49}$$

Its variation is

$$E[\Psi+\delta\Psi] = E[\Psi] + \frac{\langle\delta\Psi|H|\Psi\rangle + \langle\Psi|H|\delta\Psi\rangle}{\langle\Psi|\Psi\rangle} - \langle\Psi|H|\Psi\rangle\,\frac{\langle\delta\Psi|\Psi\rangle + \langle\Psi|\delta\Psi\rangle}{\langle\Psi|\Psi\rangle^2} + \mathcal{O}^2. \tag{50}$$

The first-order term vanishes for $H|\Psi\rangle = E[\Psi]\,|\Psi\rangle$, which is the Schrödinger equation. Since the eigenfunctions

$$H|\Psi_n\rangle = E_n|\Psi_n\rangle\,, \tag{51}$$

can be chosen to form an orthonormal basis, we can expand any wavefunction as

$$|\Psi\rangle = \sum_n |\Psi_n\rangle\,\langle\Psi_n|\Psi\rangle \tag{52}$$

and determine, as long as $\langle\Psi|\Psi\rangle \neq 0$, its energy expectation value

$$\frac{\langle\Psi|H|\Psi\rangle}{\langle\Psi|\Psi\rangle} = \frac{\sum_{m,n}\langle\Psi|\Psi_m\rangle\langle\Psi_m|H|\Psi_n\rangle\langle\Psi_n|\Psi\rangle}{\sum_{m,n}\langle\Psi|\Psi_m\rangle\langle\Psi_m|\Psi_n\rangle\langle\Psi_n|\Psi\rangle} = \frac{\sum_n E_n\left|\langle\Psi_n|\Psi\rangle\right|^2}{\sum_n\left|\langle\Psi_n|\Psi\rangle\right|^2}\,. \tag{53}$$

Since by definition no eigenenergy can be lower than the ground state energy E_0, we immediately see that the energy expectation value can never drop below the ground state energy

$$\frac{\langle\Psi|H|\Psi\rangle}{\langle\Psi|\Psi\rangle} = \frac{\sum_n E_n\left|\langle\Psi_n|\Psi\rangle\right|^2}{\sum_n\left|\langle\Psi_n|\Psi\rangle\right|^2} \geq \frac{\sum_n E_0\left|\langle\Psi_n|\Psi\rangle\right|^2}{\sum_n\left|\langle\Psi_n|\Psi\rangle\right|^2} = E_0\,. \tag{54}$$

We can use the same argument to generalize this variational principle: Assume we have arranged the eigenenergies in ascending order, $E_0 \leq E_1 \leq \cdots$, then the energy expectation value for a wavefunction that is orthogonal to the n lowest eigenstates, can not drop below E_n

$$\frac{\langle\Psi_{\perp_n}|H|\Psi_{\perp_n}\rangle}{\langle\Psi_{\perp_n}|\Psi_{\perp_n}\rangle} \geq E_n \quad \text{if } \langle\Psi_i|\Psi_{\perp_n}\rangle = 0 \text{ for } i = 0,\ldots,n{-}1. \tag{55}$$

This generalized variational principle is, of course, only of practical use if we know something about the eigenstates, e.g., when we can use symmetries to ensure orthogonality.

For an ab-initio Hamiltonian of N electrons in the field of nuclei of charge Z_α at position $\boldsymbol{R}_\alpha$,

$$H = -\frac{1}{2}\sum_i \Delta_i - \sum_{i,\alpha} \frac{Z_\alpha}{|\boldsymbol{r}_i - \boldsymbol{R}_\alpha|} + \sum_{i<j} \frac{1}{|\boldsymbol{r}_i - \boldsymbol{r}_j|} + \sum_{\alpha<\beta} \frac{Z_\alpha Z_\beta}{|\boldsymbol{R}_\alpha - \boldsymbol{R}_\beta|}\,, \tag{56}$$

the Schrödinger equation is a partial differential equation. In second quantization it becomes a linear-algebra problem: We introduce an orbital basis set $\{\varphi_k \,|\, k\}$, which for simplicity we assume here to be orthonormal, from which we construct an orthonormal basis of N-electron product states, $\{\Phi_{k_1,\dots,k_N} \,|\, k_1<\cdots<k_N\}$. To simplify the notation we sort the basis states, e.g., lexicographically in the orbital indices $\boldsymbol{k} = (k_1,\dots,k_N)$ and define the row vector of basis states $|\boldsymbol{\Phi}\rangle := \big(|\Phi_{\boldsymbol{1}}\rangle, |\Phi_{\boldsymbol{2}}\rangle, \dots\big)$. The expansion of a state $|\Psi\rangle$ in this basis can then be written as

$$|\Psi\rangle = \sum_{k_1<\cdots<k_N} a_{k_1,\dots,k_N}\,|\Phi_{k_1,\dots,k_N}\rangle = \sum_i a_i\,|\Phi_i\rangle = |\boldsymbol{\Phi}\rangle\,\boldsymbol{a}\,, \tag{57}$$

where $\boldsymbol{a}$ is the vector of expansion coefficients. Likewise we can write the Schrödinger equation as a matrix eigenvalue problem

$$\boldsymbol{H}\boldsymbol{a} = \langle\boldsymbol{\Phi}|\hat{H}|\boldsymbol{\Phi}\rangle\boldsymbol{a} = \begin{pmatrix} \langle\Phi_1|\hat{H}|\Phi_1\rangle & \langle\Phi_1|\hat{H}|\Phi_2\rangle\cdots \\ \langle\Phi_2|\hat{H}|\Phi_1\rangle & \langle\Phi_2|\hat{H}|\Phi_2\rangle\cdots \\ \vdots & \vdots \quad \ddots \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ \vdots \end{pmatrix} = E \begin{pmatrix} a_1 \\ a_2 \\ \vdots \end{pmatrix} = E\boldsymbol{a}\,. \tag{58}$$

From the eigenvectors of the matrix $\boldsymbol{H}$ we easily recover the eigenstates of the Hamiltonian

$$\boldsymbol{H}\boldsymbol{a}_n = E_n\boldsymbol{a}_n \quad \rightsquigarrow \quad \hat{H}|\Psi_n\rangle = E_n|\Psi_n\rangle \quad \text{with } |\Psi_n\rangle = |\boldsymbol{\Phi}\rangle\,\boldsymbol{a}_n\,. \tag{59}$$

Unfortunately, for an ab-initio Hamiltonian like (56) we need an infinite orbital basis set, so that the Hamiltonian matrix $\boldsymbol{H}$ is infinite dimensional. A pragmatic approach to allow for computer simulations is to simply restrict the calculation to a finite basis $|\tilde{\boldsymbol{\Phi}}\rangle := \big(|\Phi_1\rangle,\dots,|\Phi_{\tilde{L}}\rangle\big)$, i.e., work with a finite matrix $\tilde{\boldsymbol{H}} := \langle\tilde{\boldsymbol{\Phi}}|\hat{H}|\tilde{\boldsymbol{\Phi}}\rangle$ of dimension $\tilde{L}$. The crucial question is then how the eigenvectors

$$\tilde{\boldsymbol{H}}\tilde{\boldsymbol{a}}_n = \tilde{E}_n\tilde{\boldsymbol{a}}_n \quad \rightsquigarrow \quad |\tilde{\Psi}_n\rangle := |\tilde{\boldsymbol{\Phi}}\rangle\,\tilde{\boldsymbol{a}}_n \tag{60}$$

are related to those of H. The answer is surprisingly simple [7]: The eigenvalues of $\tilde{\boldsymbol{H}}$, ordered as $\tilde{E}_0 \le \tilde{E}_1 \le \cdots \le \tilde{E}_{\tilde{L}-1}$, are variational with respect to those of H:

$$E_n \le \tilde{E}_n \quad \text{for } n \in \{0,\dots,\tilde{L}{-}1\}\,. \tag{61}$$

To show this, we construct a state in $\operatorname{span}\big(|\tilde{\Psi}_0\rangle,\dots,|\tilde{\Psi}_n\rangle\big)$, which by construction has an energy expectation value $\le \tilde{E}_n$, that is orthogonal to the exact eigenstates $|\Psi_0\rangle,\dots,|\Psi_{n-1}\rangle$, so that by the generalized variational principle its expectation value is $\ge E_n$. Being the non-zero solution of $n{-}1$ linear equations with n variables, such a state certainly exists, hence $E_n \le \tilde{E}_n$.
To get reliable results, we simply have to systematically increase the basis until the change in the desired eigenvalues becomes smaller than the accuracy required by the physical problem. The art is, of course, to devise clever basis sets such that this is achieved already for bases of low dimensions.

The convergence of the matrix eigenvalues with increasing basis size is surprisingly regular. Let us extend our original basis of $\tilde{L}$ states by an additional $L-\tilde{L}$ states. Then, repeating the above argument with the L-dimensional problem taking the role of $\hat{H}$, we obtain (61) with E_n being the eigenvalues of the L-dimensional Hamiltonian matrix $\boldsymbol{H}$. Since $\boldsymbol{H}$ now is finite, we can use the same argument for $-\boldsymbol{H}$, obtaining

$$-E_{L-i} \leq -\tilde{E}_{\tilde{L}-i} \quad \text{for } i \in \{1,\ldots,\tilde{L}\}. \tag{62}$$

Taking the two inequalities together we obtain

$$E_n \leq \tilde{E}_n \leq E_{n+(L-\tilde{L})} \quad \text{for } n \in \{0,\ldots,\tilde{L}-1\}. \tag{63}$$

For the special case $L=\tilde{L}+1$ of adding a single basis state, this is the Hylleraas-Undheim/MacDonald nesting property for eigenvalues in successive approximations

$$E_1 \leq \tilde{E}_1 \leq E_2 \leq \tilde{E}_2 \leq \cdots \leq \tilde{E}_L \leq E_{L+1}\,. \tag{64}$$

3.2 Matrix eigenvalue problem

For practical calculations we have to set up the Hamiltonian matrix $\tilde{\boldsymbol{H}} = \langle\tilde{\boldsymbol{\Phi}}|\hat{H}|\tilde{\boldsymbol{\Phi}}\rangle$ and the state vectors $\tilde{\boldsymbol{a}}$ for the chosen basis. This is particularly easy for a basis of Slater determinants constructed from a basis set of K orbitals $\{\varphi_k \,|\, k=0,\ldots,K-1\}$. The basis states are then the N-electron product states of $|\Phi_{k_1,\ldots,k_N}\rangle = c^\dagger_{k_N}\cdots c^\dagger_{k_1}|0\rangle$ with $k_1 < \cdots k_N$. We can write them more computer friendly as

$$|n_{K-1},\ldots,n_0\rangle = \prod_{k=0}^{K-1} \left(c^\dagger_k\right)^{n_k} |0\rangle \tag{65}$$

which is the occupation number representation with $n_k \in \{0,1\}$ and $\sum n_k = N$. It is natural to interpret the vector of occupation numbers as the binary representation of the integer $\sum_k 2^{n_k}$. This implies a natural ordering of the basis functions $|\Phi_l\rangle$. For the simple case of K=4 orbitals and N=2 electrons we obtain

i	(n_3,n_2,n_1,n_0)	state	l
0	0000		
1	0001		
2	0010		
3	0011	$c^\dagger_1 c^\dagger_0|0\rangle = |\Phi_1\rangle$	1
4	0100		
5	0101	$c^\dagger_2 c^\dagger_0|0\rangle = |\Phi_2\rangle$	2
6	0110	$c^\dagger_2 c^\dagger_1|0\rangle = |\Phi_3\rangle$	3
7	0111		
8	1000		
9	1001	$c^\dagger_3 c^\dagger_0|0\rangle = |\Phi_4\rangle$	4
10	1010	$c^\dagger_3 c^\dagger_1|0\rangle = |\Phi_5\rangle$	5
11	1011		
12	1100	$c^\dagger_3 c^\dagger_2|0\rangle = |\Phi_6\rangle$	6
13	1101		
14	1110		
15	1111		

The bit representation of the basis states also simplifies setting up the Hamiltonian matrix. Given the Hamiltonian in second quantization

$$\hat{H} = \sum_{n,m} T_{nm}\, c_n^\dagger c_m + \sum_{n'>n,\, m'>m} \underbrace{\big(U_{nn',mm'} - U_{n'n,mm'}\big)}_{=\breve{U}_{nn',mm'}} c_{n'}^\dagger c_n^\dagger c_m c_{m'} \tag{66}$$

the matrix element $\langle\Phi_l|\hat{H}|\Phi'_l\rangle$, with $|\Phi_{l'}\rangle = c_{l'_N}^\dagger \cdots c_{l'_1}^\dagger|0\rangle$, is given by

$$\sum_{n,m} T_{nm}\langle 0|c_{l_1}\cdots c_{l_N}\, c_n^\dagger c_m\, c_{l'_N}^\dagger \cdots c_{l'_1}^\dagger|0\rangle + \sum_{\substack{n'>n\\ m'>m}} \breve{U}_{nn',mm'}\langle 0|c_{l_1}\cdots c_{l_N}\, c_{n'}^\dagger c_n^\dagger c_m c_{m'}\, c_{l'_N}^\dagger \cdots c_{l'_1}^\dagger|0\rangle.$$

Anticommuting the operators coming from the Hamiltonian, the matrix elements become overlaps of $N{+}1$ and $N{+}2$-electron product states, which, by (33) and (7), are just the determinants of the overlap matrices of the corresponding orbitals. When $\hat{H}$ is written in the same orbitals as these $|\Phi_l\rangle$, the overlap matrices simplify to permutation matrices with determinant ± 1. In the occupation number representation, calculating this Fermi sign reduces to counting set bits. As an example we consider a simple hopping of an electron:

$$\begin{aligned}
c_6^\dagger c_2|\Phi_{l(181)}\rangle &= c_6^\dagger c_2\, c_7^\dagger c_5^\dagger c_4^\dagger c_2^\dagger c_0^\dagger|0\rangle \\
&= (-1)^3 c_6^\dagger c_7^\dagger c_5^\dagger c_4^\dagger c_2 c_2^\dagger c_0^\dagger|0\rangle \\
&= (-1)^3 c_6^\dagger c_7^\dagger c_5^\dagger c_4^\dagger\big(1 - c_2^\dagger c_2\big) c_0^\dagger|0\rangle \\
&= (-1)^3 c_6^\dagger c_7^\dagger c_5^\dagger c_4^\dagger \cdot c_0^\dagger|0\rangle \\
= +|\Phi_{l(241)}\rangle &= (-1)^2 c_7^\dagger c_6^\dagger c_5^\dagger c_4^\dagger \cdot c_0^\dagger|0\rangle
\end{aligned}$$

In the occupation number representation this becomes

$$1\,0\,1\,1\,0\,1\,0\,1 \quad = \quad (-1)^c \;\; 1\,1\,1\,1\,0\,0\,0\,1$$

where c is the count of set bits between the orbitals of the electron hop. Note that a dedicated machine instruction, `popcnt`, for counting set bits is part of the x86 SSE4 instruction set, see also [8].

3.3 Dimension of the Hilbert space and sparseness

Setting up basis states and Hamiltonian matrix in this way, we can easily solve the many-body problem on our variational space by using any linear algebra library. This is the exact diagonalization approach. As discussed above, it gives us variational estimates of the ground and excited states. But there is a serious practical problem: the dimension of the many-body Hilbert space. For an N-electron problem with a basis set of K orbitals there are $K\,(K{-}1)\,(K{-}2)\cdots(K-(N{-}1))$ ways of picking N occupied orbitals out of K. Since we only use one specific ordering of these indices, we have to divide by $N!$ to obtain the number of such determinants:

$$\dim \mathcal{H}_K^{(N)} = \frac{K!}{N!(K{-}N)!} = \binom{K}{N}. \tag{67}$$

Using Stirling's formula we see that for an N-electron problem this increases faster than exponentially with the size K of the basis set. This is the problem we face when converging the basis set for a finite system, e.g., a molecule. For solids we usually keep the number of orbitals per lattice site fixed, but scale to the thermodynamic limit, increasing the system size M while keeping the electron density N/M fixed. Also here the Hilbert space increases faster than exponentially. To give an impression of the problem we note that for $N = 25$ electrons and $K = 100$ orbitals the dimension already exceeds 10^{23}.

For exact diagonalization the problem gets even worse. Assuming we have a machine with 1 TeraBytes $= 2^{40}$ Bytes of RAM available. Using single precision (4 bytes) for the matrix elements, storing a matrix of dimension $(2^{40}/4)^{1/2} = 524\,288$ would already use up all memory. The dimension problem can be somewhat mitigated by exploiting symmetries: When the Hamiltonian commutes with the projection of the total spin, the number of up- and down-spin electrons is conserved separately. The N-electron Hamiltonian is then block diagonal in the sectors with fixed $N_\uparrow$ and $N_\downarrow$. The dimension of these blocks is significantly smaller than that of the full N-electron Hilbert space. Using the same orbital basis for each spin

$$\dim \mathcal{H}_{2K}^{(N_\uparrow,N_\downarrow)} = \binom{K}{N_\uparrow} \times \binom{K}{N_\downarrow}. \tag{68}$$

The S_z symmetry can be very easily implemented using the same ideas as introduced for the general case: just use bit representations for the up- and down-spin electrons separately. In fact, when the total spin projection is conserved, we can distinguish electrons of different spin. Still, the Hilbert space of the single-band, half-filled Hubbard model with just 12 sites has dimension $853\,776$. Using further symmetries, if they exist, we could bring down the dimension somewhat further, however at the expense of considerable and problem-specific effort.

The key to going to larger systems is the realization that the vast majority of the elements of the Hamiltonian matrix is zero. This is quite easy to see. For the ab-initio Hamiltonian (56) with electron-electron repulsion, matrix elements between configurations that differ in more than two electron occupations vanish. Thus, for each configuration there may only be the diagonal element, $N \times (K-N)$ hopping terms, and $N(N-1)/2 \times (K-N)(K-N-1)/2$ pair-hopping terms. Thus the fraction of non-zero matrix elements of $\tilde{\boldsymbol{H}}$ to the total number is

$$\left(1 + N\left(1 + \frac{N-1}{2}\,\frac{(K-N-1)}{2}\right)(K-N)\right) \Big/ \binom{K}{N} \tag{69}$$

which, with increasing problem size, rapidly approaches zero. For the example of $N = 25$ electrons in $K = 100$ orbitals only 834 376 of the (over 10^{23}) matrix elements per row can be non-zero. This is the worst case. The sparsity of many-body Hamiltonians is even more pronounced when working in a tight-binding basis with short-ranged hopping and local electron-electron repulsion. Thus, many-body Hamiltonians are exceedingly sparse and the more so the larger the problem. They are therefore ideally suited for approaches like the Lanczos method, that are based on matrix-vector products, which for the sparse matrices scale close to linearly in the matrix dimension.

4 Lanczos Method

As we have seen, we can find the ground-state $|\Psi_0\rangle$ and its energy E_0 for a Hamiltonian H from the variational principle. The wavefunction-functional

$$E[\Psi] = \frac{\langle\Psi|H|\Psi\rangle}{\langle\Psi|\Psi\rangle} \tag{70}$$

is minimized for $\Psi = \Psi_0$, with $E[\Psi_0] = E_0$. The functional gradient

$$\frac{\delta E[\Psi]}{\delta\langle\Psi|} = \frac{H|\Psi\rangle - E[\Psi]|\Psi\rangle}{\langle\Psi|\Psi\rangle} = |\Psi_a\rangle \tag{71}$$

gives the direction of steepest-ascent of the functional from the point $|\Psi\rangle$. Moving in the opposite direction will thus result in a wavefunction with lower energy expectation value: $E[\Psi - \alpha\Psi_a] < E[\Psi]$ for small, positive α.
To find the optimum value of α, we minimize the quadratic form $E[\Psi - \alpha\Psi_a]$. For this, it is convenient to introduce an orthogonal basis in the space spanned by the two vectors $|\Psi\rangle$ and $|\Psi_a\rangle$. From (71) we see that $\mathrm{span}\,(|\Psi\rangle, |\Psi_a\rangle) = \mathrm{span}\,(|\Psi\rangle, H|\Psi\rangle)$. As first basis vector, we normalize $|\Psi\rangle$

$$|v_0\rangle = |\Psi\rangle/\sqrt{\langle\Psi|\Psi\rangle}\,,$$

for the second vector we orthogonalize $H|v_0\rangle$ to $|v_0\rangle$

$$|\tilde{v}_1\rangle = H|v_0\rangle - |v_0\rangle\langle v_0|H|v_0\rangle \tag{72}$$

and normalize to obtain $|v_1\rangle$. With $a_n := \langle v_n|H|v_n\rangle$ and $b_1^2 := \langle\tilde{v}_1|\tilde{v}_1\rangle$, eq. (72) becomes

$$H|v_0\rangle = b_1|v_1\rangle + a_0|v_0\rangle \tag{73}$$

from which we see that $\langle v_1|H|v_0\rangle = b_1$.
We can then write any normalized wavefunction in $\mathrm{span}\,(|\Psi\rangle, H|\Psi\rangle) = \mathrm{span}\,(|v_0\rangle, |v_1\rangle)$ as

$$|v\rangle = \cos(\theta)|v_0\rangle + \sin(\theta)|v_1\rangle\,. \tag{74}$$

Minimizing the expectation value

$$\langle v|H|v\rangle = a_0\cos^2(\theta) + 2b_1\sin(\theta)\cos(\theta) + a_1\sin^2(\theta)\,, \tag{75}$$

with respect to θ, we obtain, dividing by $\cos^2(\theta)$, the quadratic equation

$$b_1\tan^2(\theta) + (a_0 - a_1)\tan(\theta) - b_1 = 0\,. \tag{76}$$

Solving for θ we find the lowest-energy state on the subspace spanned by $|v_0\rangle$ and $H|v_0\rangle$. Alternatively, we can diagonalize the Hamiltonian matrix on the two-dimensional subspace, which in the basis $|v_0\rangle$, $|v_1\rangle$ is given by

$$H_{\mathrm{span}(|\Psi\rangle, H|\Psi\rangle)} = \begin{pmatrix} a_0 & b_1 \\ b_1 & a_1 \end{pmatrix}\,. \tag{77}$$

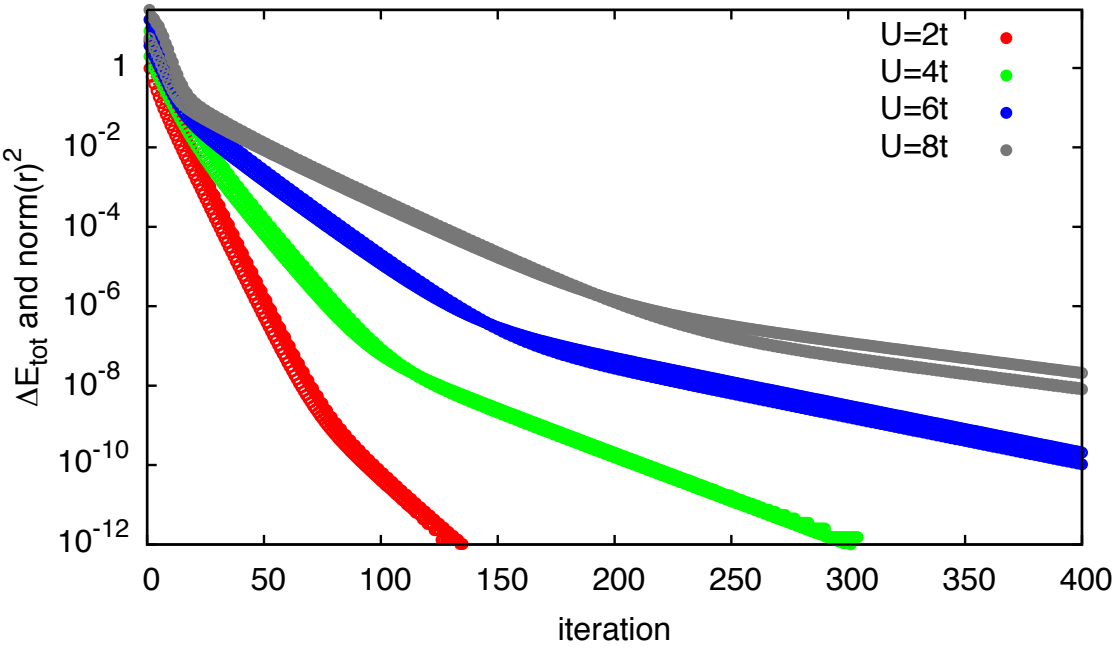

Fig. 1: *Convergence of the residual (filled circles) and the corresponding lowest eigenvalue (open circles) for a steepest-descent minimization of a Hubbard-chain of 10 sites at half-filling, starting from a random initial vector.*

Naturally, we can use the variational state of lowest energy

$$|\Psi^{(2)}\rangle = \cos(\theta_{\min})|v_0\rangle + \sin(\theta_{\min})|v_1\rangle \tag{78}$$

as the starting point for another steepest-descent minimization. Doing this repeatedly, we obtain a series of vectors with decreasing energy expectation value, which rapidly converge to a minimum. For a generic functional such a steepest-descent minimization would usually end up in a local, not the global minimum, which makes the optimization of high-dimensional functions such a hard problem. The energy functional (70), however, has only minima for the ground-states, all other stationary points are saddle points. We can thus expect rapid convergence to the ground state, examples given in figure 1, except when the starting vector $|v_0\rangle$ is orthogonal to the ground state. In this case also $\langle\Psi_0|\, H|v_0\rangle = 0$ so that we only converge to the lowest state that overlaps with $|v_0\rangle$. Should $|v_0\rangle$ happen to be an exact eigenvector, $H|v_n\rangle$ does not add a new dimension so that the algorithm terminates with $b_1 = 0$.

For checking convergence of this steepest-descent method, introduced by Kantorovich [9] and, independently, by Hestenes and Karush [10], we can monitor the change in the energy expectation value or determine when the residual

$$r[\Psi] = \big|\big|\, (H - E[\Psi])|\Psi\rangle \,\big|\big|^2 = \langle\Psi|H^2|\Psi\rangle - E[\Psi]^2\,, \tag{79}$$

which measures the quality of the eigenstate, becomes sufficiently small. As shown in Fig. 1, both are closely related.

4.1 Krylov space

If we apply the method of steepest-descent L times, starting from a vector $|v_0\rangle$, the resulting vector will lie in

$$\mathcal{K}^L\big(|v_0\rangle\big) = \text{span}\left(|v_0\rangle, H|v_0\rangle, H^2|v_0\rangle, \ldots, H^L|v_0\rangle\right), \tag{80}$$

the $L{+}1$-dimensional *Krylov space* [11] of H over $|v_0\rangle$. Instead of repeatedly minimizing the energy in two-dimensional subspaces, we could directly find the state of lowest energy in $\mathcal{K}^L\big(|v_0\rangle\big)$. Having more degrees of freedom for the minimization will lead to even faster convergence.

To implement this idea, we construct an orthonormal basis $|v_n\rangle$ of the Krylov space. We start with the normalized vector $|v_0\rangle$. The second basis vector $|v_1\rangle$ is constructed as in the steepest-descent method (72):

$$b_1|v_1\rangle = |\tilde{v}_1\rangle = H|v_0\rangle - a_0|v_0\rangle\,. \tag{81}$$

The next basis vector is likewise constructed as $H|v_n\rangle$ orthogonalized to all previous vectors, and normalized

$$b_2|v_2\rangle = |\tilde{v}_2\rangle = H|v_1\rangle - \sum_{i=0}^{1}|v_i\rangle\langle v_i|H|v_1\rangle = H|v_1\rangle - a_1|v_1\rangle - b_1|v_0\rangle\,. \tag{82}$$

where $a_n = \langle v_n|H|v_n\rangle$ and $b_n^2 = \langle\tilde{v}_n|\tilde{v}_n\rangle$. The fourth basis vector is

$$b_3|v_3\rangle = |\tilde{v}_3\rangle = H|v_2\rangle - \sum_{i=0}^{2}|v_i\rangle\langle v_i|H|v_2\rangle = H|v_2\rangle - a_2|v_2\rangle - b_2|v_1\rangle\,. \tag{83}$$

The last term in the orthogonalization vanishes, because (81) together with the orthogonality of the already constructed basis vectors for $n < 3$ implies $\langle v_2|H|v_0\rangle = b_1\langle v_2|v_1\rangle + a_0\langle v_2|v_0\rangle = 0$. The construction of the further basis vectors follows the same scheme

$$b_{n+1}|v_{n+1}\rangle = |\tilde{v}_{n+1}\rangle = H|v_n\rangle - \sum_{i=0}^{n}|v_i\rangle\langle v_i|H|v_n\rangle = H|v_n\rangle - a_n|v_n\rangle - b_n|v_{n-1}\rangle$$

with $a_n = \langle v_n|H|v_n\rangle$ and $b_n^2 = \langle\tilde{v}_n|\tilde{v}_n\rangle$. Rearranging shows that H is tridiagonalized

$$H|v_n\rangle = b_n|v_{n-1}\rangle + a_n|v_n\rangle + b_{n+1}|v_{n+1}\rangle$$

which in turn implies that $H|v_i\rangle$ is orthogonal to all basis states, except $|v_i\rangle$ and $|v_{i\pm1}\rangle$. This tridiagonalization of H is the essence of the *Lanczos method* [12].

After L steps the Hamiltonian on the $L{+}1$-dimensional Krylov space is given by

$$H_{\mathcal{K}^L(|v_0\rangle)} = \begin{pmatrix} a_0 & b_1 & 0 & 0 & & 0 & 0 \\ b_1 & a_1 & b_2 & 0 & \cdots & 0 & 0 \\ 0 & b_2 & a_2 & b_3 & & 0 & 0 \\ 0 & 0 & b_3 & a_3 & & 0 & 0 \\ & \vdots & & & \ddots & \vdots & \\ 0 & 0 & 0 & 0 & & a_{L-1} & b_L \\ 0 & 0 & 0 & 0 & \cdots & b_L & a_L \end{pmatrix} \tag{84}$$

`v=init`	
`b0=norm2(v)`	not part of tridiagonal matrix
`scal(1/b0,v)`	v= $\|v_0\rangle$
`w=0`	
`w=w+H*v`	w= $H\|v_0\rangle$
`a[0]=dot(v,w)`	
`axpy(-a[0],v,w)`	w= $\|\tilde{v}_1\rangle = H\|v_0\rangle - a_0\|v_0\rangle$
`b[1]=norm2(w)`	
`for n=1,2,...`	
`  if abs(b[n])<eps then exit`	invariant subspace
`  scal(1/b[n],w)`	w= $\|v_n\rangle$
`  scal( -b[n],v)`	v= $-b_n\|v_{n-1}\rangle$
`  swap(v,w)`	
`  w=w+H*v`	w= $H\|v_n\rangle - b_n\|v_{n-1}\rangle$
`  a[n]=dot(v,w)`	a[n]= $\langle v_n\|H\|v_n\rangle - b_n\langle v_n\|v_{n-1}\rangle$
`  axpy(-a[n],v,w)`	w= $\|\tilde{v}_{n+1}\rangle$
`  b[n+1]=norm2(w)`	
`  diag(a[0]..a[n], b[1]..b[n])`	getting a_{n+1} needs another $H\|v\rangle$
`  if converged then exit`	
`end`	

Table 1: *The implementation of the Lanczos iteration requires only two N-dimensional vectors for tridiagonalizing H and thus for calculating the ground-state energy. Constructing the Lanczos-approximation of the ground-state vector requires a second iteration and one additional N-dimensional vector. The by far most expensive operation is the matrix-vector product, which requires a problem specific implementation, while the vector operations use the BLAS.*

If we do not normalize the basis vectors, we obtain an iteration of the form

$$|\Phi_{n+1}\rangle = H\,|\Phi_n\rangle - \frac{\langle\Phi_n|H|\Phi_n\rangle}{\langle\Phi_n|\Phi_n\rangle}\,|\Phi_n\rangle - \frac{\langle\Phi_n|\Phi_n\rangle}{\langle\Phi_{n-1}|\Phi_{n-1}\rangle}\,|\Phi_{n-1}\rangle \tag{85}$$

where $|\Phi_n\rangle = \prod_{i=1}^{n} b_i\,|v_n\rangle$ in terms of which we have

$$a_n = \frac{\langle\Phi_n|H|\Phi_n\rangle}{\langle\Phi_n|\Phi_n\rangle}\;, \quad b_n^2 = \frac{\langle\Phi_n|\Phi_n\rangle}{\langle\Phi_{n-1}|\Phi_{n-1}\rangle}\;. \tag{86}$$

In this unnormalized basis the Hamiltonian appears non-Hermitian

$$H\,|\Phi_n\rangle = b_n^2\,|\Phi_{n-1}\rangle + a_n\,|\Phi_n\rangle + |\Phi_{n+1}\rangle\;, \tag{87}$$

but, of course, it actually is

$$\langle\Phi_{n+1}|H|\Phi_n\rangle = \langle\Phi_{n+1}|\Phi_{n+1}\rangle = b_{n+1}^2\,\langle\Phi_n|\Phi_n\rangle = \langle\Phi_n|H|\Phi_{n+1}\rangle\,. \tag{88}$$

The numerical implementation only requires keeping two N-dimensional vectors in memory. It is shown in table 1.

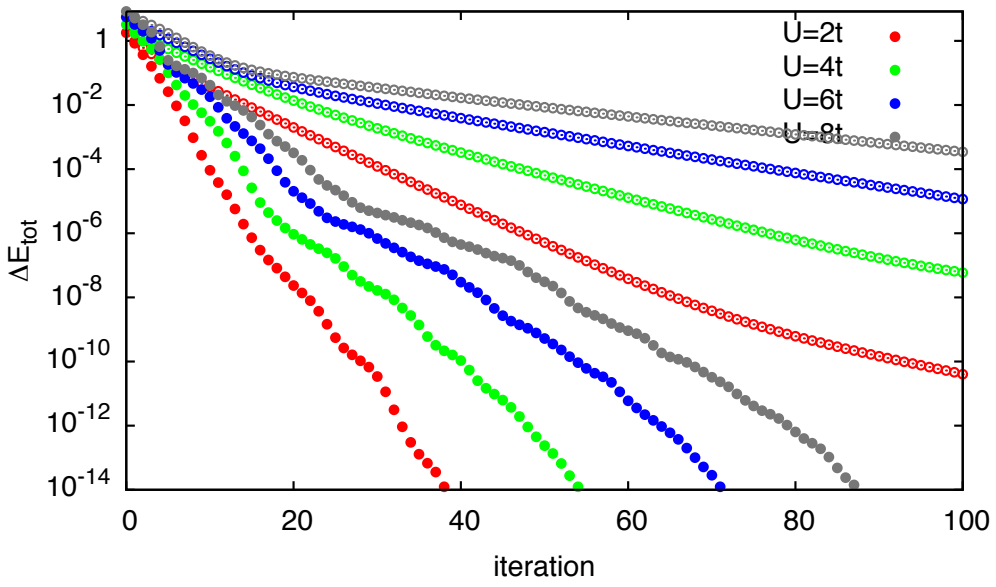

Fig. 2: *Convergence of the lowest eigenvalue for a Lanczos iteration (full circles) compared to steepest-descent minimization (open circles) of a 10-site Hubbard-chain at half-filling, starting from a random initial vector. Due to the additional variational degrees of freedom, Lanczos converges significantly faster. Overall, convergence for the half-filled system gets harder for larger U, as the distance to the lowest excited states is reduced ($\sim t^2/U$) and the spectrum widens ($\sim U$). In all cases, convergence is reached after less than $L \approx 100$ Lanczos iterations, to be compared to the dimension N=63 504 of the Hilbert space.*

Diagonalizing (84), after a few tens to hundred iterations, the lowest eigenvalue of the tridiagonal representation of H on the Krylov space gives an excellent approximation to the ground-state energy of H in the full Hilbert space (Fig. 2). A formal estimate of the convergence was given by Kaniel and Paige [13]: For an $N{+}1$-dimensional, symmetric matrix H with eigenvalues E_n, the lowest eigenvalue $\check{E}_0$ of the tridiagonal representation of H on the $(L{+}1)$-dimensional Krylov space over $|v_0\rangle$ fulfills

$$\frac{\check{E}_0 - E_0}{E_N - E_0} \leq \left(\frac{\tan(\arccos(\langle\check{\Psi}_0|\Psi_0\rangle))}{T_L\left(1 + 2\,\frac{E_1 - E_0}{E_N - E_1}\right)} \right)^2 \tag{89}$$

where $T_L(x)$ is the Chebyshev polynomial of order L and $\langle\check{\Psi}_0|\Psi_0\rangle$ the overlap of the Lanczos approximation to the ground-state $\check{\Psi}_0$ with the ground-state of H. Thus, if the initial state $|v_0\rangle$ is not orthogonal to the non-degenerate ground-state, convergence is exponential with a rate roughly increasing with the square root of the gap to the first excited state measured in units of the width of the spectrum.

The approximate ground-state vector is given by the linear combination

$$|\check{\Psi}_0\rangle = \sum_{n=0}^{L} \check{\psi}_{0,n}|v_n\rangle\,, \tag{90}$$

where $\check{\psi}_0$ is the ground-state vector of the $L{+}1$-dimensional tridiagonal matrix (84). Instead of storing all $L{+}1$ basis vectors $|v_n\rangle$, we can restart the Lanczos iteration from the same $|v_0\rangle$,

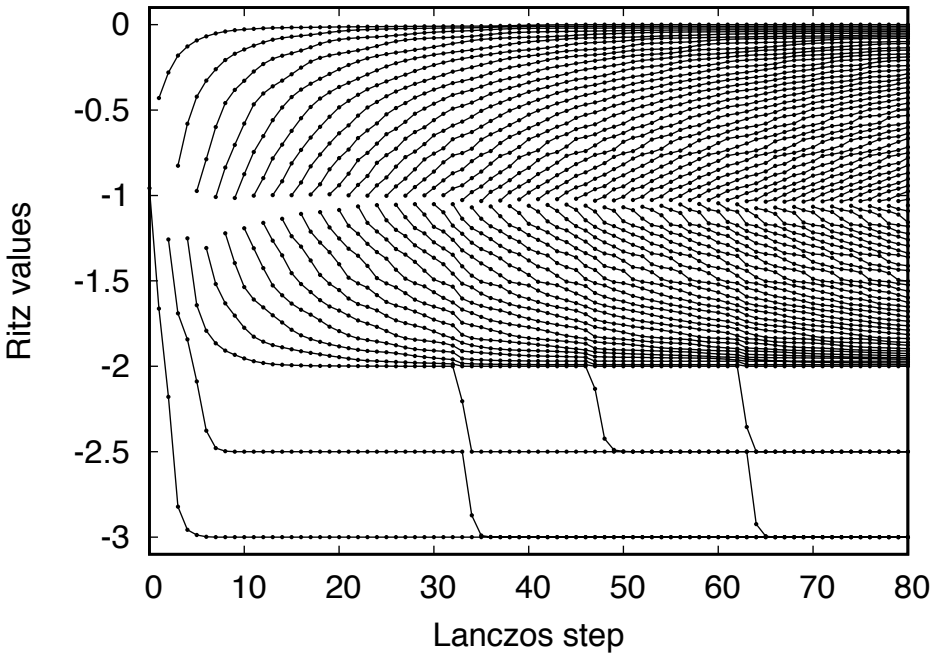

Fig. 3: *Lanczos method for a matrix with eigenvalues* -3*,* -3*,* -2.5*,* -2*,* -1.99*,* -1.98*, . . . ,* -0.01*,* 0*. For clarity the* n*-th lowest/highest Lanczos eigenvalues are connected by lines. Note how the eigenvalues of successive steps are nested as described by* (64)*. Because of the gap the lowest eigenvalue is reached much faster than the highest. For the degenerate eigenspace of value* -3 *only the state proportional to the projection of* $|v_0\rangle$ *on that space is found. Well after convergence of the low eigenvalues orthogonality is lost and ghost states appear.*

accumulating the sum (90) iteration by iteration. This only requires keeping one additional N-dimensional vector in memory.

So far we have tacitly assumed that the Krylov vectors $H^n|v_0\rangle$ are linearly independent. If not, there will be a vector $H|\tilde{v}_m\rangle$ that vanishes when orthogonalized to the previous states, i.e., $b_n = 0$. This means that the Krylov space $\mathrm{span}\,(|v_0\rangle, |v_1\rangle, \ldots, |v_m\rangle)$ is invariant under H, i.e., we have found an exact eigenspace of H. For a large matrix H it is quite unlikely to be that lucky. Still, as the Lanczos iteration approaches the ground-state, we encounter a similar situation: Close to an eigenstate, the functional (70) becomes almost stationary, i.e., the coefficients b_n almost vanish. Normalization of the very short vector $|\tilde{v}_n\rangle$ then amplifies numerical noise in that vector. This makes the numerical $|v_n\rangle$, which in theory should automatically be orthogonal to all $|v_m\rangle$ with $m < n-2$, actually have finite overlaps with these vectors. This loss of orthogonality manifests itself in the appearance of multiple copies of eigenvectors (ghost states) which are unrelated to the actual multiplicities of the eigenvalues. This makes the Lanczos method unpractical for tridiagonalizing dense matrices. For the ground-state the variational principle prevents severe problem from the loss of orthogonality. An example of the appearance of ghost states is shown in figure 3.

If we want to reliably obtain excited states, we need to explicitly orthogonalize to the previous basis states. This leads to the Lanczos method with complete reorthogonalization [13]. A similar orthogonalization is performed in the Arnoldi method [14], which, however, is devised for unsymmetric matrices.

4.2 Spectral functions

The Lanczos method gives excellent approximations to the largest and smallest eigenvalues, but, as seen in Fig. 3, not for the bulk of the spectrum. It therefore looks ill suited for determining functions that depend on the excited states, like the Lehmann representation

$$G_c(z) = \left\langle \Psi_c \left| \frac{1}{z-H} \right| \Psi_c \right\rangle = \sum_{n=0}^{N} \frac{\langle\Psi_c|\Psi_n\rangle\,\langle\Psi_n|\Psi_c\rangle}{z-E_n} \tag{91}$$

which, in terms of the eigenstates on the Krylov space $\mathcal{K}^L(|\Psi_c\rangle)$, would be written as

$$\check{G}_c(z) = \left\langle \Psi_c \left| \frac{1}{z-\check{H}_c} \right| \Psi_c \right\rangle = \sum_{n=0}^{L} \frac{\langle\Psi_c|\check{\Psi}_n\rangle\,\langle\check{\Psi}_n|\Psi_c\rangle}{z-\check{E}_n}\,. \tag{92}$$

This is straightforward to calculate: We run L Lanczos iterations, starting from the (normalized) vector $|\Psi_c\rangle$, to create the tridiagonal $\check{H}_c$. The matrix element of the resolvent is the top left matrix element of the inverse of

$$z-\check{H}_c = \left(\begin{array}{c|cccccc} z-a_0 & -b_1 & 0 & 0 & \cdots & 0 & 0 \\ \hline -b_1 & z-a_1 & -b_2 & 0 & \cdots & 0 & 0 \\ 0 & -b_2 & z-a_2 & -b_3 & \cdots & 0 & 0 \\ 0 & 0 & -b_3 & z-a_3 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \cdots & z-a_{L-1} & -b_L \\ 0 & 0 & 0 & 0 & \cdots & -b_L & z-a_L \end{array}\right). \tag{93}$$

This is easily determined, partitioning the matrix as indicated

$$z-\check{H}_c = \begin{pmatrix} z-a_0 & B^{(1)^T} \\ B^{(1)} & z-\check{H}_c^{(1)} \end{pmatrix} \tag{94}$$

and inverting the block-matrix, giving, see appendix A.3,

$$\left[(z-\check{H}_c)^{-1}\right]_{00} = \left(z-a_0-B^{(1)^T}(z-\check{H}_c^{(1)})^{-1}B^{(1)}\right)^{-1} = \left(z-a_0-b_1^2\left[(z-\check{H}_c^{(1)})^{-1}\right]_{00}\right)^{-1}.$$

Repeating inversion by partitioning for the submatrices $\check{H}^{(n)}$ we obtain the continued fraction

$$\check{G}_c(z) = \left[(z-\check{H}_c)^{-1}\right]_{00} = \cfrac{1}{z-a_0-\cfrac{b_1^2}{z-a_1-\cfrac{b_2^2}{z-a_2-\cdots}}}, \tag{95}$$

which terminates with $-b_L^2/(z-a_L)$. We find the spectral representation (92) by diagonalizing the Lanczos matrix $\check{H}_c$ giving us the $L{+}1$ eigenvalues $\check{E}_n$ and eigenvectors $\check{\psi}_n$. Since

$$|\check{\Psi}_n\rangle = \sum_{l=0}^{L} \check{\psi}_{n,l}|v_l\rangle \tag{96}$$

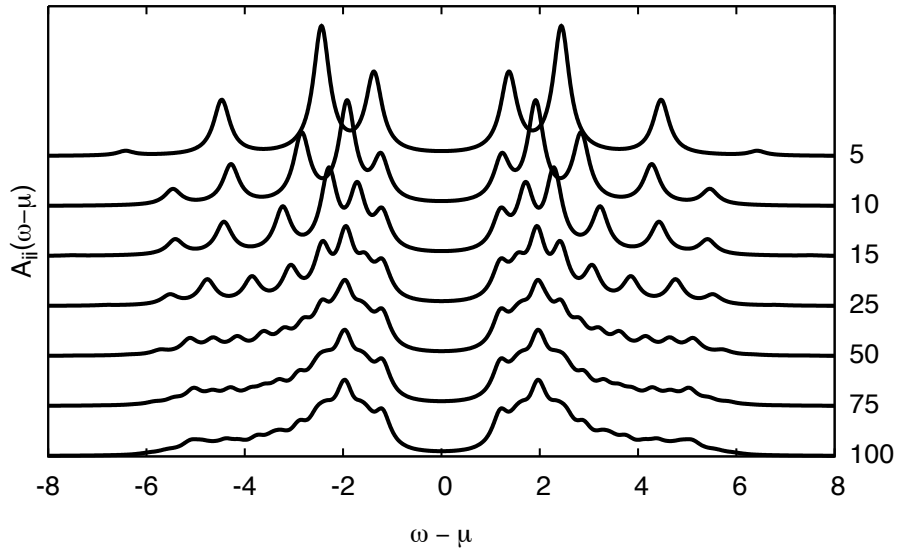

Fig. 4: *Convergence of the spectral function with increasing number of Lanczos steps, L=5, 10, 15, 25, 50, 75, and 100, for a 14-site Hubbard chain with $U = 5t$ at half filling. With increasing L, more and more moments of the photoemission and inverse photoemission part of the spectrum are reproduced correctly.*

the matrix elements are given by $\langle \check{\Psi}_n | \Psi_c \rangle = \check{\psi}_{n,0}$. Thus

$$\check{G}_c(z) = \sum_{n=0}^{L} \frac{|\check{\psi}_{n,0}|^2}{z - \check{E}_n} \,. \tag{97}$$

The spectral function $\check{A}(\omega \pm i\eta) = \mp \frac{1}{\pi} \operatorname{Im} \check{G}(\omega \pm i\eta)$ obtained this way, surprisingly, converges very quickly. An example is shown in figure 4.

To understand how the $L+1$ eigenstates of $\check{H}$ can represent the full spectrum so well, we consider the moments of the spectral function

$$\int_{-\infty}^{\infty} d\omega\, \omega^m \check{A}(\omega) = \sum_{n=0}^{L} |\check{\psi}_{n,0}|^2 \check{E}_n^m = \sum_{n=0}^{L} \langle \Psi_c | \check{\Psi}_n \rangle \langle \check{\Psi}_n | \Psi_c \rangle \, \check{E}_n^m = \langle \Psi_c | \check{H}^m | \Psi_c \rangle. \tag{98}$$

Since $\check{H}$ is the projection of H onto the Krylov space $\mathcal{K}^L(|\Psi_c\rangle)$, we have $\check{H}^m|\Psi_c\rangle = H^m|\Psi_c\rangle$ for $m \leq L$. Thus the Lanczos representation $\check{A}(z)$ correctly reproduces the first $2L+1$ moments of the spectral function $A(z)$. A further Lanczos step adds one new level to the continued fraction (95), leaving all previous terms unchanged. $b_m^2 = 0$ then implies that the continued fraction terminates, and all moments are given correctly. A near vanishing $b_m^2 \approx 0$, which gives rise to the loss of orthogonality of the Lanczos vectors, for the spectral function merely means that further terms in the continued fraction hardly contribute any more.

So far we have considered diagonal elements of the resolvent. Off-diagonal matrix elements

$$G_{c_1,c_2}(z) = \left\langle \Psi_{c_2} \left| \frac{1}{z - H} \right| \Psi_{c_1} \right\rangle \tag{99}$$

are easily obtained by considering the diagonal elements for the linear combinations

$$\left\langle \Psi_{c_1} \pm \Psi_{c2} \left| \frac{1}{z - H} \right| \Psi_{c_1} \pm \Psi_{c_2} \right\rangle = G_{c_1,c_1}(z) \pm G_{c_1,c_2}(z) \pm G_{c_2,c_1}(z) + G_{c_2,c_2}(z) \,. \tag{100}$$

A Appendices

A.1 Non-orthonormal basis

A general one-electron basis of functions $|\chi_n\rangle$ will have an overlap matrix $S_{nm} = \langle\chi_n|\chi_m\rangle$ that is positive definite (and hence invertible) and hermitian. The completeness relation is

$$\mathbb{1} = \textstyle\sum_{k,l} |\chi_k\rangle (S^{-1})_{kl} \langle\chi_l| \,. \tag{101}$$

With it we can easily write the Schrödinger equation $\hat{H}|v\rangle = \varepsilon|v\rangle$ in matrix form

$$\sum_k \underbrace{\langle\chi_i|H|\chi_k\rangle}_{=:H_{ik}} \underbrace{\sum_l (S^{-1})_{kl}\langle\chi_l|v\rangle}_{=:v_k} \langle\chi_i|\hat{H}|v\rangle = \varepsilon\langle\chi_i|v\rangle = \varepsilon \sum_k \underbrace{\langle\chi_i|\chi_k\rangle}_{=S_{ik}} \underbrace{\sum_l (S^{-1})_{kl}\langle\chi_l|v\rangle}_{=v_k} \,. \tag{102}$$

Collecting all components this becomes the generalized eigenvalue problem $\boldsymbol{H}\boldsymbol{v} = \varepsilon\boldsymbol{S}\boldsymbol{v}$. From the solution $\boldsymbol{v}$ we can easily construct $|v\rangle = \sum v_k|\chi_k\rangle$ [15]. It is, however, often more convenient to have an orthonormal basis, so that we do not have to deal with the overlap matrices in the definition of the second quantized operators or the generalized eigenvalue problem.
To orthonormalize the basis $\{|\chi_n\rangle\}$, we need to find a basis transformation $\boldsymbol{T}$ such that

$$|\varphi_n\rangle := \textstyle\sum_m |\chi_m\rangle\, T_{mn} \qquad \text{with} \quad \langle\varphi_n|\varphi_m\rangle = \delta_{mn} \,. \tag{103}$$

This implies that $\boldsymbol{T}^\dagger\boldsymbol{S}\boldsymbol{T} = \mathbb{1}$, or equivalently $\boldsymbol{S}^{-1} = \boldsymbol{T}\boldsymbol{T}^\dagger$. This condition does not uniquely determine $\boldsymbol{T}$. In fact there are many orthonormalization techniques, e.g., Gram-Schmidt orthonormalization or Cholesky decomposition.
Usually we will have chosen the basis functions $|\chi_n\rangle$ for a physical reason, e.g., atomic orbitals, so that we would like the orthonormal basis functions to be as close to the original basis as possible, i.e, we ask for the basis transformation $\boldsymbol{T}$ that minimizes

$$\begin{aligned}\sum_n \big\||\varphi_n\rangle - |\chi_n\rangle\big\|^2 &= \sum_n \Big\| \sum_m |\chi_m\rangle (T_{mn} - \delta_{mn}) \Big\|^2 \\ &= \operatorname{Tr}(\boldsymbol{T}^\dagger - \mathbb{1})\,\boldsymbol{S}\,(\boldsymbol{T} - \mathbb{1}) = \operatorname{Tr}(\underbrace{\boldsymbol{T}^\dagger\boldsymbol{S}\boldsymbol{T}}_{=\mathbb{1}} - \boldsymbol{T}^\dagger\boldsymbol{S} - \boldsymbol{S}\boldsymbol{T} + \boldsymbol{S}) \,.\end{aligned} \tag{104}$$

Given an orthonormalization $\boldsymbol{T}$, we can obtain any other orthonormalization $\tilde{\boldsymbol{T}}$ by performing a unitary transformation, i.e., $\tilde{\boldsymbol{T}} = \boldsymbol{T}\boldsymbol{U}$. Writing $\boldsymbol{U} = \exp(i\lambda\boldsymbol{M})$ with $\boldsymbol{M}$ a Hermitian matrix, we obtain the variational condition

$$0 \stackrel{!}{=} \operatorname{Tr}(+i\boldsymbol{M}\boldsymbol{T}^\dagger\boldsymbol{S} - i\boldsymbol{S}\boldsymbol{T}\boldsymbol{M}) = i\operatorname{Tr}(\boldsymbol{T}^\dagger\boldsymbol{S} - \boldsymbol{S}\boldsymbol{T})\boldsymbol{M}, \tag{105}$$

which is fulfilled for $\boldsymbol{S}\boldsymbol{T} = \boldsymbol{T}^\dagger\boldsymbol{S}$, i.e., $\boldsymbol{S}\boldsymbol{T}^2 = \boldsymbol{T}^\dagger\boldsymbol{S}\boldsymbol{T} = \mathbb{1}$. The second variation at $\boldsymbol{T} = \boldsymbol{S}^{-1/2}$

$$\frac{1}{2}\operatorname{Tr}(\boldsymbol{M}^2\boldsymbol{S}^{1/2} + \boldsymbol{S}^{1/2}\boldsymbol{M}^2) > 0 \tag{106}$$

is positive, since $\boldsymbol{S}$ and the square of the hermitian matrix $\boldsymbol{M}$ are both positive definite. Hence the Löwdin symmetric orthogonalization [16]

$$\boldsymbol{T}_{\text{Löwdin}} = \boldsymbol{S}^{-1/2} \tag{107}$$

minimizes the modification of the basis vectors due to orthogonalization.

A.2 Useful commutation relations

Expressions of commutators of products of operators can be derived by adding and subtracting terms that differ only in the position of one operator, e.g.,

$$\begin{aligned}
[A_1 A_2 \cdots A_N,\, B] = &\quad A_1 A_2 \cdots A_N B - B A_1 A_2 \cdots A_N \\
= &\quad A_1 A_2 \cdots A_N B - A_1 A_2 \cdots B A_N \\
&+ A_1 A_2 \cdots B A_N - A_1 \cdots B A_{N-1} A_N \\
&+ \qquad\qquad \cdots \\
&+ A_1 B A_2 \cdots A_N - B A_1 A_2 \cdots A_N \\
= &\sum_i A_1 \cdots A_{i-1}\, [A_i,\, B]\, A_{i+1} \cdots A_N
\end{aligned}$$

The following special cases are particularly useful

$$\begin{aligned}
[AB,\, C] &= A\,[B,\, C] \,+\, [A,\, C]\,B \\
&= A\{B,\, C\} - \{A,\, C\}B
\end{aligned}$$

$$\begin{aligned}
[A,\, BC] &= B\,[A,\, C] \,+\, [A,\, B]\,C \\
&= [A,\, B]\,C \,+\, B\,[A,\, C] \\
&= \{A,\, B\}C - B\{A,\, C\}
\end{aligned}$$

$$\begin{aligned}
[AB,\, CD] &= A\,[B,\, C]\,D + AC\,[B,\, D] \,+\, [A,C]\,DB + C\,[A,\, D]\,B \\
&= A\{B,\, C\}D - AC\{B,\, D\} + \{A,C\}DB - C\{A,\, D\}B
\end{aligned}$$

Important examples are

$$\begin{aligned}
\left[c_i^\dagger c_j,\, c_\gamma^\dagger\right] &= \quad \langle j|\gamma\rangle\, c_i^\dagger \\
\left[c_i^\dagger c_j,\, c_\gamma\right] &= -\langle i\,|\gamma\rangle\, c_j
\end{aligned}$$

For the commutator of products of creation and annihilation operators appearing in one- and two-body operators we find

$$\left[c_i^\dagger c_j,\, c_\alpha^\dagger c_\beta\right] = \left[c_i^\dagger c_j,\, c_\alpha^\dagger\right] c_\beta + c_\alpha^\dagger \left[c_i^\dagger c_j,\, c_\beta\right] = \langle j|\alpha\rangle\, c_i^\dagger c_\beta - \langle \beta|i\rangle\, c_\alpha^\dagger c_j$$

and

$$\left[c_i^\dagger c_j^\dagger c_k c_l\,,\, c_\alpha^\dagger c_\beta\right] = \langle l|\alpha\rangle\, c_i^\dagger c_j^\dagger c_k c_\beta + \langle k|\alpha\rangle\, c_i^\dagger c_j^\dagger c_\beta c_l - \langle \beta|j\rangle\, c_i^\dagger c_\alpha^\dagger c_k c_l - \langle \beta|i\rangle\, c_\alpha^\dagger c_j^\dagger c_k c_l$$

A.3 Downfolding

To integrate-out high-energy degrees of freedom, we partition the Hilbert space of the full system into states of interest (low-energy states) and 'other' states, which will be integrated out. The Hamiltonian is then written in blocks

$$H = \begin{pmatrix} H_{00} & T_{01} \\ T_{10} & H_{11} \end{pmatrix}, \tag{108}$$

where H_{00} is the Hamiltonian restricted to the states of interest (reduced Hilbert space), H_{11} the Hamiltonian for the 'other' states, and the T matrices describe transitions between the two subspaces. The resolvent is partitioned likewise

$$G(\omega) = (\omega - H)^{-1} = \begin{pmatrix} \omega - H_{00} & -T_{01} \\ -T_{10} & \omega - H_{11} \end{pmatrix}^{-1}. \tag{109}$$

Its elements are easily determined by solving the system of two linear matrix equations

$$\begin{pmatrix} \omega - H_{00} & -T_{01} \\ -T_{10} & \omega - H_{11} \end{pmatrix} \begin{pmatrix} G_{00} & G_{01} \\ G_{10} & G_{11} \end{pmatrix} = \begin{pmatrix} \mathbb{1} & \mathbb{0} \\ \mathbb{0} & \mathbb{1} \end{pmatrix}, \tag{110}$$

keeping track of the order of the sub-matrix products. The resolvent on the reduced Hilbert space is thus given by

$$G_{00}(\omega) = \Big(\omega - \big(\underbrace{H_{00} + T_{01}(\omega - H_{11})^{-1} T_{10}}_{=H_{\text{eff}}(\omega)}\big)\Big)^{-1}. \tag{111}$$

This expression looks just like the resolvent for a Hamiltonian H_{eff} on the reduced Hilbert space. This effective Hamiltonian describes the physics of the full system, but operates only on the small reduced Hilbert space: For an eigenvector $H|\Psi\rangle = E|\Psi\rangle$ on the full Hilbert space

$$H|\Psi\rangle = \begin{pmatrix} H_{00} & T_{01} \\ T_{10} & H_{11} \end{pmatrix} \begin{pmatrix} |\Psi_0\rangle \\ |\Psi_1\rangle \end{pmatrix} = E \begin{pmatrix} |\Psi_0\rangle \\ |\Psi_1\rangle \end{pmatrix} \tag{112}$$

its projection $|\Psi_0\rangle$ onto the reduced Hilbert space is an eigenstate of $H_{\text{eff}}(E)$. On the other hand, we can construct the full eigenstate from a solution $H_{\text{eff}}(E)|\Psi_0\rangle = E|\Psi_0\rangle$ on the reduced Hilbert space by upfolding $|\Psi\rangle \propto (\mathbb{1} + (E - H_{11})^{-1}T_{10})|\Psi_0\rangle$.

Of course, this drastic simplification comes at a price: the effective Hamiltonian is energy dependent. If the hopping matrix elements in T_{01} are small, and/or the states in the part of the Hilbert space that has been integrated out are energetically well-separated from the states that are explicitly considered, this energy dependence can, to a good approximation, be neglected. We can then replace ω by some characteristic energy ε_0 for the states in the reduced Hilbert space to obtain an energy-independent Hamiltonian

$$H_{\text{eff}}(\omega) = H_{00} + T_{01}(\omega - H_{11})^{-1} T_{10} \approx H_{00} + T_{01}(\varepsilon_0 - H_{11})^{-1} T_{10} = H_{\text{eff}}(\varepsilon_0) \tag{113}$$

that gives a good description of the electrons in the reduced Hilbert space, i.e., the states with an energy close to ε_0. Expanding $(\omega - H_{11})^{-1}$ about ε_0, we can systematically improve the approximation (linear and higher-order methods).

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