Atomic cluster expansion and wave function representations

Ralf Drautz
ICAMS, Ruhr-Universität Bochum, Bochum, Germany

Christoph Ortner
Department of Mathematics, University of British Columbia, Vancouver, BC, Canada V6T 1Z2

(Dated: June 24, 2022)

The atomic cluster expansion (ACE) was recently introduced as a polynomial expansion for the parameterization of the interatomic interaction that explicitly encodes permutation and rotation symmetries. It is a complete representation, which means that any conceivable form of the interaction between atoms can in principle be represented. The framework has been extended to include magnetism and charge transfer, to handle vectorial and tensorial objects as well as message passing networks. Efficient implementations and parameterization software are available. Previously we provided ample evidence that ACE models can compete with and oftentimes outperform far more complex and computationally expensive deep learning models.

Here we explore from a theoretical perspective the deep connections between symmetric and anti-symmetric functions in the context of the ACE framework and how to employ this framework to discretize many-electron wave functions. First, we demonstrate how a number of well-known linear parameterisations naturally arise from a polynomial expansion by adapting the ACE framework. In particular, we propose a single-particle basis that can explicitly or implicitly incorporate electron-electron and electron-nuclei cusp terms to accelerate the convergence of the discretisation.

An important motivation for this work is the remarkable success of recent machine learning representations of wave functions. Therefore, we also review the most important nonlinear parameterisations (Slater-Jastrow, Backflow) and discuss how ACE representations of symmetric functions provide building blocks towards alternative and/or generalized parameterisations of those representations. A potential advantage of our framework is a clear interpretation of natural approximation parameters and a flexible choice of those parameters, which will in particular enable the study of convergence, both theoretically and numerically. In this way we contribute to a systematic interpolation between classical and modern nonlinear parameterisations of wave functions and a systematic exploration of this design space.

I. INTRODUCTION

The state of a particle is described by a variable \( x \), which can include the position and several other features of the particle. For atomistic simulations one chooses \( x = (r, µ) \), where \( r \) is the position and \( µ \) the chemical species, whereas electrons are described by \( x = (r, σ) \) with \( σ \) the spin state. We start from an orthonormal and complete set of one-particle basis functions, \( φ_v(x) \),

\[
\langle v|u \rangle = \sum_σ \int dr \ φ_v^*(x) φ_u(x) = δ_{vu} ,
\]

\[
\sum_v |v⟩⟨v| = \sum_v φ_v^*(x) φ_v(x') = δ(x-x') ,
\]

with basis function indices \( v = 0, 1, 2, \ldots \). Leaving away any symmetry considerations for the moment, it is straightforward to write down a general expansion of a \( N \)-particle function \( Ψ(1, 2, \ldots, N) \) as

\[
Ψ = \sum_v \sum_i c_v^i φ_v(i) + \sum_{v1 v2 i1 i2} c_{v1 v2}^{i1 i2} φ_{v1}(i1) φ_{v2}(i2) + \ldots + \sum_{v1 \ldots vN i1 \ldots iN} c_{v1 \ldots vN}^{i1 \ldots iN} φ_{v1}(i1) φ_{v2}(i2) \cdots φ_{vN}(iN) ,
\]

where \( φ_v(i) = φ_v(x_i) \), \( φ_0(i) = 1 \) and the summation over basis function indices is limited to \( v > 0 \). (Until we impose symmetries, the coefficients \( c_{v1 \ldots vN}^{i1 \ldots iN} \) must carry the labels \( i_1, \ldots, i_N \) of the interacting particles in each term.)

The sum over indices \( i_1 i_2 \ldots \) is unrestricted, i.e. contributions \( i_n = i_m \) are also taken into account. This expansion is referred to as the atomic cluster expansion, see also Ref. [2] for an analogous expansion for discrete variables. One would naturally constrain \( c_{v1 \ldots vN}^{i1 \ldots iN} = 0 \) if any two particles are identical \( i_n = i_m \) to avoid unphysical self-interactions. As the unphysical self-interactions can be removed from the expansion by modified lower order expansion coefficients, we will not enforce this constraint in the following.

Because the representation Eq. (3) is constructed from products of single particle functions, one may choose the...
expansion coefficients to be symmetric with respect to simultaneous change of the particle indices \( i_n \leftrightarrow i_m \) and the corresponding basis indices \( v_n \leftrightarrow v_m \), illustrated here for the N-product coefficient,

\[
c_{v_1 d_1 \ldots d_n d_m \ldots d_N} = c_{v_1 d_1 \ldots d_m d_n \ldots d_N},
\]

without limiting the generality of the expansion and we will assume this symmetry for the following. Then in Eq. (4) either the sum over particles \( i \) or basis function \( v \) can be restricted to \( i_1 \leq i_2 \leq \ldots \leq i_N \) or \( v_1 \leq v_2 \leq \ldots \leq v_N \), which leads to different representations of symmetric and anti-symmetric functions that will be discussed in the next sections.

A. Symmetric functions

If the N-particle function is symmetric with respect to exchange of particles

\[
\Psi(1, 2, \ldots, n, \ldots, m, \ldots, N) = \Psi(1, 2, \ldots, m, \ldots, n, \ldots, N),
\]

then the expansion coefficients are symmetric with respect to exchange of particles, which means that the particle index can be dropped from the expansion coefficients and the expansion written as

\[
\Psi = \sum_{i} \sum_{v} c_{v} \phi_{v}(i) + \sum_{v_1 v_2} c_{v_1 v_2} \phi_{v_1}(i_1) \phi_{v_2}(i_2) + \ldots + \sum_{v_1 v_2 \ldots v_N} c_{v_1 v_2 \ldots v_N} \phi_{v_1}(i_1) \phi_{v_2}(i_2) \ldots \phi_{v_N}(i_N).
\]

From Eq. (3) the expansion coefficients are symmetric with respect to exchange of basis function indices \( v_n \leftrightarrow v_m \), that is,

\[
c_{v_1 v_2 \ldots v_n \ldots v_m \ldots v_N} = c_{v_1 v_2 \ldots v_m \ldots v_n \ldots v_N}.
\]

This enables to carry out the summation over particles \( i \) first followed by summation over basis function indices \( v \) (sometimes called "density trick") and enables very efficient expansions for atomistic simulations. To this end the atomic basis is introduced

\[
A_v = \sum_i \phi_v(i),
\]

and the expansion written as

\[
\Psi = \sum_v c_v A_v + \sum_{v_1 v_2} c_{v_1 v_2} A_{v_1} A_{v_2} + \ldots + \sum_{v_1 v_2 \ldots v_N} c_{v_1 v_2 \ldots v_N} A_{v_1} A_{v_2} \ldots A_{v_N}.
\]

From Eq. (7) the summations can be restricted further to ordered \( v_1 \leq v_2 \leq \ldots \).

As representation of functions in high dimension is plagued by the curse of dimensionality, it is particularly interesting to highlight the improvement in computational cost that the ACE expansion affords over a naive tensor product approximation. First, we point out that through a recursive evaluation of the basis functions \( A_{v_1} \cdots A_{v_n} \), the cost, \( \text{COST} \), of evaluating Eq. (4) becomes directly proportional to the number of parameters, \( \text{PARAMS} \); to be precise, \( \text{COST} \sim 2 \times \text{PARAMS} \). Counting the number of parameters requires more information about the basis choice than we have provided so far. However, even in general it is intuitively clear that in the limit of a complete basis set where most tuples \( (v_1 \ldots v_N) \) will be strictly ordered, the number of parameters is reduced approximately by a factor \( N! \).

We can say more if we make a specific choice, and will focus for simplicity on the case of a total degree approximation which is particularly natural when approximation smooth (analytic) \( \Psi \). This results in restricting the sums \( \sum_{v_1 v_2 \ldots v_N} \) in Eq. (6) over the ordered tuples \( (v_1, \ldots, v_n) \) to \( \sum_{i=1}^{N} \deg(\phi_v) \leq D \). Here, \( \deg(\phi_v) \) denotes the degree of the basis function \( \phi_v \), which depends on the choice of basis, but is almost always canonical and \( D \) is the total degree. Bachmayr et al. make this precise and show that for a \( d \)-dimensional discretisation any natural choice of basis will lead to both asymptotic and pre-asymptotic bounds,

\[
\text{PARAMS} \lesssim \begin{cases} \exp(cD^{d(N)^N}), & \text{as } D \to \infty, \\ \exp(cD^{1-1/d}), & \text{uniformly in } N, D. \end{cases}
\]

The first bound makes precise the intuition that the basis size is reduced by a factor \( N! \) compared with an unsymmetrized approximation. The second bound is particularly interesting in two respects: (1) it does not rely on the \( D \to \infty \) asymptotics which cannot be practically reached; (2) It is remains super-algebraic but independent of the number of particles \( N \), which results in the powerful suggestion that approximation quality may be independent of dimensionality.

B. Anti-symmetric functions

The expansion may also be simplified significantly for anti-symmetric N-particle functions. Then,

\[
\Psi(1, 2, \ldots, n, \ldots, m, \ldots, N) = -\Psi(1, 2, \ldots, m, \ldots, n, \ldots, N).
\]

As the exchange of two particles must change the sign of the function, it is clear from the general expansion Eq. (3) that only the N-product may contribute. The expansion coefficient of the N-product has to fulfill

\[
c_{v_1 i_2 \ldots i_n \ldots i_m \ldots i_N} = -c_{v_1 i_2 \ldots i_n \ldots i_m \ldots i_N}
\]

\[
\text{and}
\]

\[
-c_{v_1 i_2 \ldots i_m \ldots i_n \ldots i_N}.
\]
where the second identity follows from Eq. (11). This allows to factorize the expansion coefficient as
\[
\epsilon_{i_1 i_2 \ldots i_N} = \epsilon_{i_1 i_2} \epsilon_{i_3 i_4} \ldots \epsilon_{i_{N-1} i_N} c_{v_{i_1} v_{i_2} \ldots v_{i_N}}, \tag{13}
\]
with \(\epsilon_{i_1 i_2} \ldots i_N = 1\) for an even particle permutation, \(\epsilon_{i_1 i_2} \ldots i_N = -1\) for an odd permutation, and \(\epsilon_{i_1 i_2} \ldots i_N = 0\) if two or more indices are identical. This allows to factorize \(c_{v_{i_1} v_{i_2} \ldots v_{i_N}}\), and where the expansion coefficients are symmetric
\[
c_{v_{i_1} v_{i_2} \ldots v_{i_m} \ldots v_{i_N}} = c_{v_{i_1} v_{i_2} \ldots v_{i_m} \ldots v_{i_N}}. \tag{14}
\]
As for the general expansion Eq. (3) and using Eq. (4), one may limit the sums either to ordered particle indices \(i_1 < i_2 < \cdots < i_N\) or ordered basis function indices \(v_1 < v_2 < \cdots < v_N\). This implies that one has some freedom for anti-symmetric expansions that we discuss next.

1. Restricted basis function summation

We first discuss anti-symmetric expansions when the summation over basis functions is limited \(v_1 < v_2 < \cdots < v_N\), while the particle indices are completely summed over.

a. Configuration interaction With \(v_1 < v_2 < \cdots < v_N\), Eq. (3) reduces to a general anti-symmetric expansion given by
\[
\Psi = \sum_{v_1 < v_2 < \cdots < v_N} c_{v_{i_1} v_{i_2} \ldots v_{i_N}} D_{v_{i_1} v_{i_2} \ldots v_{i_N}}, \tag{15}
\]
with the Slater determinant
\[
D_{v_{i_1} v_{i_2} \ldots v_{i_N}} = \frac{1}{\sqrt{N!}} \sum_{i_1 i_2 \ldots i_N} \epsilon_{i_1 i_2 \ldots i_N} \phi_{v_1}(i_1) \phi_{v_2}(i_2) \ldots \phi_{v_N}(i_N). \tag{16}
\]
The Slater determinant takes the same role as the product basis \(A_{v_1} A_{v_2} \cdots A_{v_N}\) in Eq. (9) for symmetric functions. The linear expansion Eq. (15) is referred to as configuration interaction and the completeness of the configuration interaction representation is shown here by construction.

The computational cost of evaluating the configuration interaction representation can be analyzed similarly as in the symmetric case. The cost of evaluating a single Slater determinant \(D_\rho\) or its gradient \(\nabla D_\rho\) is of the order \(O(N^3)\) (the dominant contribution is the factorisation of \(\phi_{a_i}(i_b)\)). Since we are restricting the basis to \(v_1 < v_2 < \cdots\) the number of terms can be bounded exactly as in the symmetric case; cf. Eq. (10). Thus, for a total degree discretisation we obtain
\[
\text{COST} \lesssim N^3 \cdot \text{PARAMS}.
\]
The strict ordering \(v_1 < v_2 < \cdots\) as opposed to non-strict ordering \(v_1 \leq v_2 \leq \cdots\) in the symmetric case only marginally improves the bound in the pre-asymptotic regime.

b. Effective single determinant A complete expansion may also be obtained from a single determinant-like object by simply changing the summation over particles and basis function indices. Consider a general product expansion that is not symmetric with respect to exchange of particles,
\[
\Phi(123 \ldots N) = \sum_{v_1 < v_2 < \cdots < v_N} c_{v_{i_1} v_{i_2} \ldots v_{i_N}} \phi_{v_1}(1) \phi_{v_2}(2) \cdots \phi_{v_N}(N), \tag{17}
\]
then
\[
\Psi = \frac{1}{\sqrt{N!}} \sum_{i_1 i_2 \ldots i_N} \epsilon_{i_1 i_2 \ldots i_N} \Phi(i_1 i_2 \ldots i_N) \tag{18}
\]
is again a complete parameterisation of anti-symmetric functions. Despite having to anti-symmetrize only the function \(\Phi(123 \ldots N)\), this representation appears computationally highly inefficient since the \(O(N!)\) explicit anti-symmetrisation cannot be avoided as in the case of a Slater determinant. We will discuss the related backflow parameterizations in the following but already here hint that this representation may be viewed as an effective single Slater determinant by representing \(\Phi(123 \ldots N)\) as a product of \(N\) effective single-particle functions \(\varphi_i\), where effective means that \(\varphi_i\) depends on all particle coordinates, which allows one to achieve efficient representations.

2. Restricted particle summation

Alternatively, one may limit the sum over particle indices to \(i_1 < i_2 < \cdots < i_N\). As for \(N\) particles for an anti-symmetric expansion there is only one contribution to the summation that fulfills \(i_1 < i_2 < \cdots < i_N\), the sum over particle indices may be dropped and the expansion written as
\[
\Psi = \sum_{v_{i_1} v_{i_2} \ldots v_{i_N}} c_{v_{i_1} v_{i_2} \ldots v_{i_N}} \epsilon_{i_1 i_2 \ldots i_N} \phi_{v_1}(1) \phi_{v_2}(2) \ldots \phi_{v_N}(N). \tag{19}
\]
This corresponds to the occupation number representation of second quantization. Because of \(\epsilon_{v_1 v_2 \ldots v_N}\) two electrons cannot be in the same orbital and exchanging two electrons means exchanging their one particle orbitals which leads to a sign change of the expansion.

III. EFFECTIVE SINGLE PARTICLE FUNCTIONS

In practise the configuration interaction and related representations need to be improved for accurate wave function representations. A basic idea is to regard the single electron wave functions \(\phi_i(x)\) as quasi-particle wave functions that depend not only on the coordinate of particle \(i\) but on the coordinates of other particles, too.
This will also become a crucial ingredient in our review of nonlinear parameterisations below.

We use the ACE to construct a general representation of a generalized single-particle function

\[ \varphi(i) = \varphi_i(x_i; x_{\neq i}) \]

that is associated to particle \( i \) but depends on all other particles, too. The notation \( \varphi_i(x_i; x_{\neq i}) \) indicates that \( \varphi_i \) is required to be symmetric with respect to exchange of all particles except \( i \). A complete expansion using the ACE formalism then reads

\[ \varphi(i) = \sum_v c_v \phi_{v1}(i) \prod_{t=2}^\nu A_{v_t}, \quad (20) \]

with

\[ A_v = \sum_{j \neq i} \phi_v(j). \quad (21) \]

While Eq. (20) provides a completely general expansion of an effective single-particle function, it still lacks two fundamental properties. First, the Hamiltonian is invariant under simultaneous translation of the positions of electrons and nuclei and the same should hold for the wave function, while the single electron basis functions \( \phi_v(i) \) and the effective single electron functions \( \varphi_i \) are not. A simple method to generate translationally invariant representations is to work with difference vectors between particles only. This was employed, for example, in ACE models for interatomic potentials to achieve translational invariance of general interatomic interactions. In addition the ACE representation provides a complete representation for functions with permutation symmetries. Secondly, the many-electron wave function has characteristic cusps when two electrons come close. It is computationally demanding to provide accurate representations of the cusps with smooth polynomial single electron basis functions. By working with basis functions that depend on difference vectors the cusps can be naturally injected into the wavefunction.

We therefore propose to replace the atomic base \( A_v \) in Eq. (21) and \( \phi_{v1}(i) \) in Eq. (20), respectively, with

\[ A_v = \sum_j^{\text{electrons}} \phi^{\text{el}}_v(\mathbf{r}_j - \mathbf{r}_i) + \sum_j^{\text{nuclei}} \phi^{\text{nu}}_v(\mathbf{R}_j - \mathbf{r}_i), \quad (22) \]

and

\[ \phi_v(i) = \sum_j^{\text{nuclei}} \varphi_{vj}^{\text{nu}}(\mathbf{R}_j - \mathbf{r}_i), \quad (23) \]

where \( \mu_j \) denotes the chemical species of atom \( j \). The linear combination of atomic orbitals appears to be a natural choice for representing the pairwise function,

\[ \phi^{\text{el}}_v(\mathbf{r}) = R^{\text{el}}_{nl}(r)Y_{\ell m}(\hat{\mathbf{r}}), \quad (24) \]

\[ \phi^{\text{nu}}_{vj}(\mathbf{r}) = R^{\text{nu}}_{nl}(r)Y_{\ell m}(\hat{\mathbf{r}}), \quad (25) \]

where \( v = (nlm) \) is now a multi-index. Different from the usual reasoning, the orbitals are carried by the electrons and not the nuclei. In this representation it is also straightforward to integrate cusp conditions in the limit \( r_{ji} \to 0 \) through the choice of the radial basis \( R^{\text{el}}_{nl}, R^{\text{nu}}_{nl} \).

The pairwise functions (or, equivalently, the atomic base) could be adapted,

\[ A_v^{\text{opt}} = \sum_k u_{vk} A_k, \quad \text{and} \quad \phi_v^{\text{opt}}(x_i) = \sum_k u_{1,vk} \phi_k(x_i). \quad (26) \]

Replacing \( A_v \) with \( A_v^{\text{opt}} \) and \( \phi_{v1} \) with \( \phi_v^{\text{opt}} \) in Eq. (20) results in a nonlinear representation of \( \varphi_i \) known as a Tucker tensor format. Such a format has been used with considerable success for optimizing only the radial basis in the original atomic cluster expansion.

The effective single particle functions could be extended and refined further by adapting recent extensions of ACE in natural ways to include message passing to electrons along similar lines as in the PauliNet architecture.

For some applications covariance of the wave function and also the generalized single-particle functions \( \varphi(i) \) under joint rotation and inversion of electron and nuclei positions may be required. Such covariance is readily incorporated into the parameterisation, using the techniques outlined in Refs. [1] and [2] and results in

\[ \varphi(i) = \sum_v c_v \sum_k C_{k,v} \phi_{v1}(i) \prod_{t=2}^\nu A_{v_t}, \quad (27) \]

where the (sparse) matrix \( C \) contains generalized Clebsch–Gordan coefficients that specify all possible covariant couplings between the \( \phi_{v1}, A_v \). In particular, this symmetrisation results in significantly fewer parameters \( c_k \) than parameters \( c_v \) in (20).

### IV. NONLINEAR PARAMETRISATIONS

Since evaluating general anti-symmetric functions is significantly more costly than evaluating general symmetric functions, there are numerous approaches to represent anti-symmetric functions in terms of symmetric contributions, normally leading to nonlinear parameterisations of anti-symmetric functions. In Eq. (26), we introduce a canonical mechanism how nonlinearities arise. Here we discuss how several other important approaches can be related to the atomic cluster expansion, or, how the atomic cluster expansion may be employed to represent both the symmetric and anti-symmetric contributions in these representations.
A. Slater-Jastrow ansatz

Trivially, any anti-symmetric function $\Psi$ may be decomposed into

$$
\Psi(1 \cdots N) = \sum_{q=1}^{Q} U_q(1 \cdots N) \Psi_q(1 \cdots N),
$$

(28)

where $U_q$ are symmetric and $\Psi_q$ again anti-symmetric. The challenge then is to impose restrictions on $\Psi_q$ that achieve desired accuracy or qualitative physical properties at an overall much lower cost than fully resolving the wave function directly using, e.g., the naive configuration interaction.

The original Jastrow (or, Slater–Jastrow) wave function ansatz\textsuperscript{26,27} takes the form

$$
\psi(1 \cdots N) = \exp\left(\sum_{i<j} u_{2}(r_{ij})\right) \Psi_1(1 \cdots N),
$$

(29)

where $\Psi_1$ is an anti-symmetric function often chosen to be a classical Slater determinant. For example, if $\Psi_1$ is chosen to be a Slater determinant containing the standard atomic orbitals, and $u_2(r_{ij}) = a_{ij} r_{ij} / (1 + b_{ij} r_{ij})$ with suitably chosen parameters $a_{ij}, b_{ij}$, then the ansatz incorporates the correct electron-electron cusps. By incorporating the nuclei into the expression the electron-nuclei cusps can be resolved well. An in-depth review is given by Foulkes et al.\textsuperscript{27}

The atomic cluster expansion provides a computationally cheap and general mechanism to generalise the two-body form of the Jastrow factor to incorporate terms of arbitrary body-order,

$$
\Psi(1 \cdots N) = \exp\left(\sum_{i=1}^{N} \phi^{(\nu)}(i)\right) \Psi_1(1 \cdots N),
$$

(30)

where $\phi^{(\nu)}(i)$ is a generalized single particle function with ACE representation of the form Eq.\textsuperscript{[20]}, possibly with restricted correlation-order $\nu$, and centered at electron $i$.

As discussed above, the computational cost associated with symmetric functions of higher correlation order is quite moderate compared to the cost of the configuration interaction approach. It is particularly interesting in this context to explore how the accuracy of this ACE-Jastrow-Slater ansatz improves with increasing correlation order. For example, whether higher correlation orders can be employed to resolve higher-order electron cusps.

B. Vandermonde ansatz

The Slater-Jastrow ansatz (with a single term, i.e. $Q = 1$) has many desirable physical and computational properties but is in general understood to be incomplete. Han et al.\textsuperscript{28} introduce a specific class of Slater determinants that further simplify it and render it a complete (universal) representation. If we choose $\Psi_q$ in Eq.\textsuperscript{[28]} to be a generalised Vandermonde determinant,

$$
\Psi_q := \mathcal{V}(\varphi_q) := \prod_{i<j}(\varphi_q(i) - \varphi_q(j)),
$$

(31)

where $\varphi_q = (\varphi_q(i))_{i=1}^{N}$ is a permutation covariant vector, and $U_q$ are general symmetric functions, then they establish that Eq.\textsuperscript{[28]}, which now reads

$$
\Psi = \sum_{q=1}^{Q} U_q \mathcal{V}(\varphi_q),
$$

(32)

can be converged to a general anti-symmetric function.

The ACE representation can obviously be used both to parameterise the symmetric part $U_q$, either directly or as a generalized Jastrow factor as in Eq.\textsuperscript{[30]}. In addition, ACE is also eminently suited to parameterise $\varphi_q$; namely, a general permutation covariant vector can be written again as a generalized single particle function,

$$
[\varphi_q]_i = \varphi_q(i) = \varphi_q(x; x \neq i),
$$

where $\varphi_q$ is symmetric in the second argument $x \neq i$ and hence represented in ACE format by Eq.\textsuperscript{[29]}. The results of Han et al.\textsuperscript{28} imply that this representation is complete in the limit of an infinite basis and $Q \to \infty$. We are unaware of any attempt to employ this representation in practical simulations.

C. Backflow ansatz

The backflow ansatz originally suggested by Feynman and Cohen\textsuperscript{29} has increased in prominence due to the phenomenal success when incorporating deep learning strategies\textsuperscript{10,11}. It employs a generalized Slater determinant,

$$
\Psi = \mathcal{B}(\varphi_1, \ldots, \varphi_N) = \det\begin{pmatrix}
\varphi_1(1) & \varphi_2(1) & \cdots & \varphi_N(1) \\
\varphi_1(2) & \varphi_2(2) & \cdots & \varphi_N(2) \\
\vdots & \vdots & \ddots & \vdots \\
\varphi_1(N) & \varphi_2(N) & \cdots & \varphi_N(N)
\end{pmatrix},
$$

(33)

where each $\varphi_j(i) = \varphi_j(x; x \neq i)$ is permutation-invariant in the second argument. It is easy to see that this generalizes both the Slater-Jastrow ansatz Eq.\textsuperscript{[29]} as well as the generalised Vandermonde representation Eq.\textsuperscript{[28]}. The backflow ansatz can be practically implemented using the ACE parameterisation of the components $\varphi$ by employing again the ACE parameterisations of Sec. III. Similarly as in the Slater–Jastrow approach, if ACE is employed to parameterise the $\varphi_j$ components, it becomes a both mathematically and physically interesting question to explore how the accuracy of the parameterisation improves with increasing correlation order.
Hutter showed that a single term of this form suffices to represent a general anti-symmetric function. In particular, this establishes that also the ACE variant of Eq. is not general, hence this does not constitute a proof of completeness of the backflow ansatz, but a possible route towards identifying low-dimensional structures that could explain its significant practical success.

V. SUMMARY

We demonstrate how the atomic cluster expansion naturally reproduces and relates the most common and most important representations of many-electron wave functions. It remains to be seen how this framework can be leveraged for the development of more efficient computational methods, or a deeper theoretical understanding of existing methods. Numerous routes exist that must be explored, such as the optimization of effective single particle functions to incorporate cusp conditions while retaining the regularity and low-dimensionality of sparse polynomial spaces, combined with a systematically improvable backflow ansatz.

For modelling atomic interactions, the polynomial ACE basis was shown to improve over machine learning representations. Analogous improvements for wave function representations may also be facilitated by ACE.

ACKNOWLEDGMENTS

RD acknowledges helpful discussions with Yury Lysogorskiy, Anton Bochkarev and Matteo Rinaldi. CO thanks Huajie Chen, Dexuan Zhou and Zeno Schätzle for inspiring conversations on the topic of this article.

CO is supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) [funding reference number IDGR019381].

1 R. Drautz, Phys. Rev. B 99, 014104 (2019).
2 M. Bachmayr, G. Csányi, G. Duduson, R. Drautz, S. Etter, C. van der Oord, and C. Ortner, J. Comp. Phys. 454 (2022), https://doi.org/10.1016/j.jcp.2022.110946.
3 R. Drautz, Phys. Rev. B 102, 024104 (2020).
4 A. Bochkarev, Y. Lysogorskiy, C. Ortner, G. Csányi, and R. Drautz, *Multilayer atomic cluster expansion for semi-local interactions* (2022).
5 I. Batatia, S. Batzner, D. P. Kovács, A. Musaelian, G. N. C. Simm, R. Drautz, C. Ortner, B. Kozinsky, and G. Csányi, *The design space of $\rho^3$-equivariant atom-centered interatomic potentials* (2022).
6 Y. Lysogorskiy, C. v. d. Oord, A. Bochkarev, S. Menon, M. Rinaldi, T. Hammerschmidt, M. Mrovec, A. Thompson, G. Csányi, C. Ortner, and R. Drautz, npj Comput Mater 7 (2021), 10.1038/s41524-021-00559-9.
7 A. Bochkarev, Y. Lysogorskiy, S. Menon, M. Qamar, M. Mrovec, and R. Drautz, Phys. Rev. Materials 6 (2022), 10.1103/physrevmaterials.6.013804.
8 https://acsuit.github.io.
9 https://github.com/FitSNAP/FitSNAP.
10 J. Hermann, Z. Schätzle, and F. Noé, Nat. Chem. 12, 891 (2020).
11 D. Pfau, J. S. Spencer, A. G. D. G. Matthews, and W. M. C. Foulkes, Phys. Rev. Research 2 (2020).
12 J. Han, J. Lu, and M. Zhou, Journal of Computational Physics 423, 109792 (2020).
13 Y. Rath, A. Glielmo, and G. H. Booth, Phys. Rev. X 10, 041026 (2020).
14 Y. Rath, A. Glielmo, and G. H. Booth, J. Chem. Phys.
For simplicity of presentation we do not discuss spins explicitly, but note that they can be treated entirely analogously as chemical species in the original ACE model, replacing a position variable \( x = r \) with a position-spin pair \( x = (r, \sigma) \).