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## INVITED ARTICLE

### Variational formulations of the coupled-cluster method in quantum chemistry

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The coupled-cluster method is one of the most successful methods for *ab initio* electronic structure calculations. For computation of expectation values and properties, two equivalent but very different derivations exist: the Lagrangian formulation of Helgaker and Jørgensen [Adv. Quant. Chem. **19**, 183 (1988)] and the bivariational formulation of Arponen [Ann. Phys. **151**, 311 (1983)]. The latter is, apparently, less well known among quantum chemists, and we therefore discuss and contrast the two approaches. Some generalisations of coupled-cluster based on the bivariational principle are discussed, including the so-called extended coupled-cluster method.

**Keywords:** coupled-cluster; bivariational; Lagrangian

#### 1. Introduction

It is a pleasure to dedicate this article to Trygve Helgaker on the occasion of his 60th birthday. His contributions play a key role in a topic which has held great interest for the present author, and which is the subject matter of this paper, namely, the foundations of coupled-cluster (CC) theory as encountered in quantum chemistry.

CC theory has a well known and interesting history since its invention by Coester and Kümmel [1]. Development of the theory has been done in parallel by the theoretical nuclear physicists and by quantum chemists [2,3]. Of particular interest for the present article is the development of the part of CC theory concerning evaluation of expectation values and properties; an important task in quantum chemical calculations. Since the original formulation was not variational, the Hellmann–Feynman theorem did not apply out of the box, and computing energy derivatives and expectation values was therefore not straightforward.

The correct solution to this problem, finding a formulation of CC in terms of critical points of some functional, was actually discovered independently by workers in quantum chemistry (Helgaker and Jørgensen [4,5]) and in theoretical physics (Arponen [6]).

Interestingly, while leading to the same stationary principle, the problem was approached from quite different angles: Helgaker and Jørgensen derived what is today known as ‘the coupled-cluster Lagrangian’ by formulating the CC amplitude equations as a constrained optimisation problem. On the other hand, Arponen derived CC from a guiding variational principle, ‘the bivariational principle’, which is a straight generalisation of the usual variational method for the ground-state problem.

In this article, we contrast these two approaches. A slight emphasis on Arponen’s work is chosen, since Helgaker and Jørgensen’s approach is well known in the quantum chemistry community, while the approach by Arponen is, apparently, less well known. Also, it may shed some additional light on the *origins* of CC theory: where does it come from, why does it work, and, importantly, how can it be generalised and improved in a systematic manner?

The bivariational principle forms the basis of Arponen’s approach. Like a standard variational procedure, improvements can in principle be made by adding degrees of freedom to the bivariational ansatz. Thus, CC is derived by a systematic reduction of the exact many-electron problem, which suggests that improvements of standard CC are accessible. One such extension, first described by Arponen in [6], is so-called extended coupled-cluster (ECC). However, ECC has seen little use due to its apparent complexity, but there are approximations to ECC (apart from standard CC) that may be tractable. We discuss such a set of approximations and demonstrate their size-extensivity.

Another option that presents itself via the bivariational principle is to include the orbitals as variational parameters in the standard CC method, and we consider this briefly in the present paper. Such an approach has been explored theoretically and numerically for *ab initio* dynamics calculations in Ref. [7], which is related to the approach taken in Ref. [8] for response theory.

After Arponen’s initial publication [6], Arponen and coworkers have developed the abstract theory of CC to high sophistication, see, e.g., Refs. [9–15]. Therein, the configuration–interaction method, standard CC, and ECC are treated on largely equal footing. The point of view

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taken is that the methods are mappings of quantum theory into *formally classical* Hamiltonian systems. We shall have opportunity to briefly discuss the classical embedding in Section 5.

In Ref. [15], the approach taken to truncating CC theory is slightly different than what is now prevalent in quantum chemistry, as Hermitian expectation values are enforced. This yields a constrained Hamiltonian formalism in the sense of Dirac [16]. This approach is then closely related to the so-called variational coupled-cluster (VCC) method [17], and out of scope for the present paper.

An important remark regarding nomenclature is in place. The quantum chemistry and theoretical physics communities have, over the years, evolved CC nomenclatures that differ in many respects. As the present article is mainly written with quantum chemists in mind, we will select nomenclature accordingly. For example, the classical Hamiltonian formulation by Arponen and coworkers should not be confused with the CC Lagrangian from quantum chemistry. The reader may be led to think that these are Legendre transforms of each other. This is not the case, as they are in fact, the same objects viewed in different ways.

## 2. A brief recap of CC theory

Suppose a single-particle basis is given, along with an independent-particle model reference state  $|\Phi\rangle = c_1^\dagger c_2^\dagger \cdots c_N^\dagger |\text{vac}\rangle$  with  $N$  particles. Within the Hilbert space defined by the single-particle basis, any state  $|\Psi\rangle$  with  $\langle\Phi|\Psi\rangle = 1$  can be expanded as

$$|\Psi\rangle = (1 + U_1 + U_2 + \cdots + U_N) |\Phi\rangle, \quad (1)$$

where  $U_n$  is an excitation operator, creating  $n$  particle-hole pairs relative to the reference state. One may observe that  $1 + U_1 + \cdots$  can be written in exponential form,

$$|\Psi\rangle = \exp(T) |\Phi\rangle, \quad T = T_1 + T_2 + \cdots, \quad (2)$$

where  $T_n$  is again an excitation operator. All excitations commute,  $[T_n, T_m] = 0$ , and  $\exp(T_n)$  describes all possible *independent excitations* of subsets of particles in  $\Phi$ . The exponential ansatz, therefore, factorises for subsystems  $A$  and  $B$  if they are separated by, say, distance.

Truncating  $T$  at, e.g., singles-and-doubles excitations,  $T \approx T_1 + T_2$ , we obtain an approximate wavefunction allowing all independent excitations of up to two particles, irrespective of whether the other particles are excited or not. Truncation of the linear ansatz (1) cannot achieve this and is therefore not size-extensive.

Instead of attempting a variational minimisation of the energy using  $\exp(T)|\Phi\rangle$  as trial function, Coester and Kümmel [1,2] considered the similarity transformed

Schrödinger equation:

$$\exp(-T)H \exp(T) |\Phi\rangle = E |\Phi\rangle. \quad (3)$$

Projecting against the reference and any excited determinant we obtain the equations

$$E = \langle\Phi| \exp(-T)H \exp(T) |\Phi\rangle, \quad (4a)$$

$$0 = \langle\Phi_\mu| \exp(-T)H \exp(T) |\Phi\rangle. \quad (4b)$$

Here,  $\mu$  is a generic index counting the excitations, i.e.,  $T = \sum_\mu t_\mu X_\mu \equiv tX$ ,  $X_\mu$  being a product of pair creation operators on the form  $c_a^\dagger c_i$ , where  $i$  is occupied in  $|\Phi\rangle$  while  $a$  is unoccupied. Equation (4) forms a set of nonlinear equations that determine the amplitudes  $t_\mu$  of  $T$ , and is *exactly equivalent* to the Schrödinger equation (within the full configuration-interaction (FCI) space) if  $T$  is not truncated.

One chooses Equation (4) over the variational minimisation of the expectation value  $\langle\Psi|H|\Psi\rangle/\langle\Psi|\Psi\rangle$  because of the fundamental fact that the Baker–Campbell–Hausdorff expansion for  $\exp(-T)H\exp(T)$  terminates *identically*, irrespective of  $N$ , after  $2k$  commutators, where  $k = 2$  if  $H$  contains at most two-body operators,  $k = 3$  if it contains three-body operators, etc. This can be proven explicitly by computing  $\exp(-T)c_p^{(\dagger)}\exp(T) = c_p^{(\dagger)} + [c_p^{(\dagger)}, T]$ .

The nonlinear amplitude Equation (4) is complicated and is best derived using computer algebra software. The right-hand sides are  $2k$ th order polynomials in the amplitudes  $t_\mu$ . The standard way to solve these equations are by iterative methods, e.g., quasi-Newton methods [18]. If  $T$  is truncated, the system (4) is over-determined, and the standard solution is to use only the equations corresponding to projections against precisely the excitations included. That is, equation number  $\mu$  is included if and only if  $t_\mu$  is included in  $T$ .

It is standard to interpret Equation (4) in terms of Goldstone diagrams [18]. When a solution is found, the energy (4a) has a straightforward interpretation as a partial summation of certain classes of diagrams entering in many-body perturbation theory. From a perturbation theory point of view, CC is simply a clever way to sum certain classes of Goldstone diagrams to infinite order. One obtains

$$E = \sum_n \frac{1}{n!} \langle\Phi| \{HT^n\}_L |\Phi\rangle, \quad (5)$$

the subscript ‘L’ indicating that only Goldstone diagrams are retained, where each factor  $T$  is linked to  $H$ . Most calculations are done with the singles-and-doubles approximation (CCSD), that is,  $T = T_1 + T_2$  is assumed, neglecting all higher order excitations. Triples are commonly introduced in an approximate way using perturbation

theory (CCSD(T)), because its accuracy is well balanced with the computational effort; the ‘gold standard of quantum chemistry’.

Since the calculation is not variational, the question arises of how to compute expectation values in CC theory. Čížek [19] and Fink [20] used the variational expectation value,

$$\langle O \rangle_{\text{var}} = \frac{\langle \Phi | \exp(T^\dagger) O \exp(T) | \Phi \rangle}{\langle \Phi | \exp(T^\dagger) \exp(T) | \Phi \rangle}, \quad (6)$$

but it is legitimate to ask if this is natural:  $\exp(T)|\Phi\rangle$  is not a variationally optimal wavefunction, i.e., it is *not* computed by optimising Equation (6) variationally (with  $O = H$ ). Thus,  $\langle H \rangle_{\text{var}}$  does not coincide with the computed CC energy.

Another approach, typical for many-body perturbation theory, is to be inspired by Equation (4a), and compute

$$\langle O \rangle_{\text{Goldstone}} = \langle \Phi | \exp(-T) O \exp(T) | \Phi \rangle. \quad (7)$$

This amounts to computing the same diagram series as used for the CC energy, but replacing all the interactions by  $O$  instead of  $H$ .

However, none of these approaches conform with the Hellmann–Feynman theorem [21],

$$\langle \Psi | O | \Psi \rangle = \frac{d}{d\epsilon} E(H + \epsilon O) \Big|_{\epsilon=0}, \quad (8)$$

where  $|\Psi\rangle$  is the exact ground state of  $H$ , and where  $E(H')$  is the exact ground-state energy of  $H'$ . This can be viewed as a restatement of first-order perturbation theory for the energy. Replacing  $E(H')$  by some approximate energy functional, it is natural to define expectation values via Equation (8).

Monkhorst [22] developed a Hellmann–Feynman conforming framework for computing expectation values for the CC method. Thus, writing  $E_{\text{CC}}(H')$  for the CC energy, we define the expectation value to be

$$\langle O \rangle_{\text{H-F}} = \frac{d}{d\epsilon} E_{\text{CC}}(H + \epsilon O) \Big|_{\epsilon=0}. \quad (9)$$

Monkhorst arrived at a linear system of equations that determined the response  $T_O$  to the cluster amplitudes  $T$ , needed for the computation of  $\langle O \rangle_{\text{H-F}}$ . In Section 3, we discuss the Lagrangian approach of Helgaker and Jørgensen, and then make the connection with the approach of Monkhorst. The  $O$ -dependent linear equation is replaced with a *universal* linear equation (in the sense that it is independent of  $O$ ). The solutions are Lagrange multipliers, viewing CC as a constrained minimisation problem.

### 3. The Lagrangian formalism

It is now standard to define the value of a property using the Hellmann–Feynman theorem [23]. The advantages are

a clear connection with the exact theory that all expectation values are automatically size-extensive if the underlying model is size-extensive and that the expectation values are well behaved with respect to refinement of the theory to include more correlations, i.e., excitations in the CC theory.

If the ground state problem of the Hamiltonian  $H$  is formulated in terms of a stationary point of some functional  $\mathcal{E}_H$ , the Hellmann–Feynman theorem gives considerable simplifications for evaluating expectation values and higher order properties, most prominently the  $2n + 1$  rule [23].

To this end, suppose that the state of the system is approximated by an element of some smooth submanifold of the density operators, with (local) coordinates  $z$ , i.e.,  $\rho = \rho(z)$  is the state. For simplicity, one may assume that the parameters are  $n$ -dimensional,  $z \in \mathbb{C}^n$ , but this is not a limitation in general; infinite-dimensional parameterisations work as well. Assume that we have an energy functional  $\mathcal{E}_H(z)$  of a system with Hamiltonian  $H$  and assume that  $\mathcal{E}_H$  is linear in  $H$ , i.e.,

$$\mathcal{E}_{\alpha H_1 + \beta H_2}(z) = \alpha \mathcal{E}_{H_1}(z) + \beta \mathcal{E}_{H_2}(z). \quad (10)$$

Suppose the approximate ground state  $\rho_* = \rho(z_*)$  is characterised by the stationary condition:

$$\frac{\partial}{\partial z} \mathcal{E}_H(z) \Big|_{z_*} = 0. \quad (11)$$

Let  $\epsilon \mapsto H(\epsilon)$  be a smooth family of Hamiltonians,  $\epsilon \in \mathbb{R}$  being a small parameter, and let  $z_*(\epsilon)$  be the corresponding ground state, assuming that it exists. It is straightforward to compute

$$\frac{d}{d\epsilon} \mathcal{E}_{H(\epsilon)}(z_*(\epsilon)) = E_{\dot{H}(\epsilon)}(z_*(\epsilon)), \quad (12)$$

where the dot denotes a derivative with respect to  $\epsilon$ . With  $H(\epsilon) = H + \epsilon O$ , we obtain the Hellmann–Feynman expectation value

$$\langle O \rangle_{\text{H-F}} = \mathcal{E}_O(z_*), \quad (13)$$

where  $z_*$  is the stationary point corresponding to the Hamiltonian  $H$ .

Thus, it is natural to *define*  $\mathcal{E}_O(z)$  as an expectation value functional for the operator  $O$  in the state parameterised by  $z$  provided that it is linear in  $O$  and that a stationary state is defined by the stationary condition (11). The Hellmann–Feynman theorem is thus baked into the very definition of an expectation value functional.

Many methods of quantum chemistry, such as CC, are not truly variational methods. The Hermitian expectation value is not a minimum with respect to variations in the state. Configuration interaction (CI) with a single-particle basis determined by, say, the complete active space

self-consistent field (CASSCF) method is another example. The basic observation made by Helgaker and Jørgensen was that the CI energy is a *constrained minimum*, the constraints being that the orbitals solve the CASSCF equations. Therefore, one may introduce a set of Lagrange multipliers  $\lambda$  corresponding to the constraints, replacing the CI energy by a Lagrangian.

Very general non-variational approximations may be replaced by an equivalent Lagrangian. Indeed, assume for simplicity that  $x \in \mathbb{C}^n$ ,  $y \in \mathbb{C}^m$  and that  $\rho(x, y)$  is the state parameterisation. Assume that the quantum chemical method in question is a constrained minimisation of an energy function  $\mathcal{E}_H(x, y)$ , the constraints being a set of (possibly nonlinear) equations  $\mathcal{F}_H(y) = 0 \in \mathbb{R}^m$ . As before,  $\mathcal{E}_H$  and  $\mathcal{F}_H$  are linear in  $H$ . In the CI example above,  $x$  would be the CI wavefunction coefficients, while  $y$  would be a set of orbital rotation parameters.  $\mathcal{F}_H(y) = 0$  is the CASSCF equations. Introducing Lagrange multipliers  $\lambda \in \mathbb{C}^m$ , the constrained minimisation of  $\mathcal{E}_H$  is replaced by an unconstrained extremalisation of the Lagrangian:

$$\mathcal{L}_H(x, y, \lambda) = \mathcal{E}_H(x, y) + \lambda \cdot \mathcal{F}_H(y). \quad (14)$$

Clearly,  $\mathcal{L}_H$  depends linearly on  $H$  and a stationary point  $(x_*, y_*, \lambda_*)$  is determined by the conditions

$$\begin{aligned} \frac{\partial}{\partial \lambda} \mathcal{L}_H(x, y, \lambda) &= \mathcal{F}_H(y) = 0, \\ \frac{\partial}{\partial x} \mathcal{L}_H(x, y, \lambda) &= \frac{\partial}{\partial x} \mathcal{E}_H(x, y) = 0, \end{aligned}$$

and

$$\frac{\partial}{\partial \lambda} \mathcal{L}_H(x, y, \lambda) = 0.$$

Moreover, at the stationary point,  $\mathcal{L}_H(x_*, y_*, \lambda_*) = \mathcal{E}_H(x_*, y_*)$ , so that  $\mathcal{L}_H$  may be considered an expectation value functional for the nonvariational method in the sense defined above. Thus,

$$\langle O \rangle_{\text{H-F}} = \mathcal{L}_O(x_*, y_*, \lambda_*) = \mathcal{E}_O(x_*, y_*) + \lambda_* \cdot \mathcal{F}_O(y_*). \quad (15)$$

In general,  $\mathcal{F}_O(y_*) \neq 0$ .

The CC method can be formulated using this framework, leading to the CC Lagrangian. Indeed, let  $y = (t_\mu)$  be the CC amplitudes,  $T = \sum_\mu t_\mu$ , and define the de-excitation operator

$$\Lambda = \sum_\mu \lambda_\mu X_\mu^\dagger \quad (16)$$

where  $\lambda = (\lambda_\mu)$  is the set of Lagrange multipliers, truncated at the same level as  $T$ . (The set of variables  $x$  is here empty.)

The Lagrangian becomes

$$\begin{aligned} \mathcal{L}_H(T, \Lambda) &= \langle \Phi | e^{-T} H e^T | \Phi \rangle + \sum_\mu \lambda_\mu \langle \Phi | X_\mu^\dagger e^{-T} H e^T | \Phi \rangle \\ &= \langle \Phi | (1 + \Lambda) e^{-T} H e^T | \Phi \rangle, \end{aligned} \quad (17)$$

which is precisely the CC Lagrangian [4,5]. Thus, the CC ground state energy is a constrained minimum of  $\langle \Phi | \exp(-T) H \exp(T) | \Phi \rangle$ , the constraints being  $\langle \Phi | X_\mu^\dagger \exp(-T) H \exp(T) | \Phi \rangle = 0$ .

Let us contrast the Lagrangian formulation of the generic constrained minimisation problem with a direct approach to calculating the Hellmann–Feynman expectation value  $\langle V \rangle_{\text{H-F}}$ . Then applied to CC, this is the approach taken early in Ref. [22]. A necessary condition for  $\lambda_*$  to be unique is that the Jacobian  $\mathcal{J}_H(y) \equiv \partial \mathcal{F}_H / \partial y$  is invertible. In that case, we have the formula

$$\lambda_* = -\mathcal{J}_H(y_*)^{-1} \frac{\partial \mathcal{E}_H}{\partial y} \Big|_{y_*}. \quad (18)$$

Given  $H(\epsilon) = H + \epsilon O$ , the condition that  $\mathcal{J}_{H(\epsilon)}$  is invertible also means that the implicit function theorem can be applied to  $\mathcal{F}_{H(\epsilon)}(y) = 0$  to determine  $\epsilon \mapsto y_*(\epsilon)$  such that  $\mathcal{F}_{H(\epsilon)}(y_*(\epsilon)) \equiv 0$  in a neighbourhood of  $\epsilon = 0$ ,  $y_*(0) = y_*$ . The solution is given by integrating the differential equation

$$\dot{y}_*(\epsilon) = -\mathcal{J}_{H(\epsilon)}(y_*(\epsilon))^{-1} \mathcal{F}_O(y_*(\epsilon)). \quad (19)$$

In particular,

$$y_O \equiv \dot{y}_*(0) = -\mathcal{J}_H(y_*)^{-1} \mathcal{F}_O(y_*), \quad (20)$$

giving the response of  $y_*$  to the perturbation  $V$ . Compare Equation (20) with (18). We now have

$$\langle V \rangle_{\text{H-F}} = \mathcal{E}_V(x_*, y_*) + y_V \cdot \frac{\partial \mathcal{E}_H}{\partial y} \Big|_{y_*}. \quad (21)$$

Equation (21) requires the calculation of  $y_O$  for *every operator*  $O$  for which we desire the expectation value. Clearly, as the computation of the  $O$ -independent  $\lambda_*$  amounts to solving a linear system with the *same* coefficient matrix  $\mathcal{J}_H(y_*)$  as for the computation of  $y_O$ , the Lagrangian formulation has much lower cost if more than one expectation value is required.

It is interesting to note that according to the definition of the general Lagrangian  $\mathcal{L}_H(x, y, \lambda)$  as an expectation value functional, *both*  $(x, y)$  and  $\lambda$  can be interpreted as parts of the description of the state of the system: the energy is parameterised by  $(x, y, \lambda)$ . Indeed,  $\varphi(\cdot) = \mathcal{L}(\cdot, x, y, \lambda)$  is a linear functional on the space of operators. By the Riesz representation theorem there exists an operator  $\sigma_\varphi$  representing this functional such that  $\varphi(H) = \text{Tr} H \sigma_\varphi$ . But

$\varphi$  is uniquely determined by  $(x, y, \lambda)$ . Hence, there exists an operator-valued map  $(x, y, \lambda) \mapsto \sigma(x, y, \lambda)$  such that

$$\mathcal{L}_H(x, y, \lambda) = \text{Tr} H \sigma(x, y, \lambda). \quad (22)$$

This suggests the hypothesis that *all* expectation value functionals can be represented by the actual expectation value of some density operator  $\sigma$ .

However, it is not necessarily true that  $\sigma(x, y, \lambda)$  is actually a density operator, i.e., we do not know whether the representability conditions  $\sigma \geq 0$ ,  $\sigma = \sigma^\dagger$ ,  $\text{Tr} \sigma = 1$ , and  $\sigma^2 \leq \sigma$  hold.

On the other hand, for the CC method we can say more. We have

$$\mathcal{L}_H(T, \Lambda) = \text{Tr}(H\rho), \quad \rho = e^T |\Phi\rangle \langle \Phi| (1 + \Lambda) e^{-T}. \quad (23)$$

It is straightforward to see that  $\text{Tr} \rho = 1$  and that  $\rho^2 = \rho$ . However,  $\rho \neq \rho^\dagger$ . Thus, in standard CC we describe the system using a pure state density operator, albeit a non-Hermitian one, i.e., the bra and ket states are different, and  $T$  and  $\Lambda$  together parameterises the bra and ket part of  $\rho$ .

These observations foreshadow the treatment of CC given by Arponen, using the bivariational principle, which we discuss in Section 4.

#### 4. The bivariational principle

Arponen approached the CC problem using a very general bivariational principle which we now describe [6]. The result is a systematic development of CC that also allows for straightforward generalisations. To the best of the present author's knowledge, the bivariational principle is not discussed at length in the literature, except for Ref. [7]. We therefore discuss the topic in some detail here.

In this section, we compute as if all Hilbert spaces were finite dimensional, for simplicity. For a mathematically rigorous treatment one must be much more careful. We also assume a complex Hilbert space, but all observables (including the Hamiltonian) are assumed Hermitian (i.e., self-adjoint).

A straightforward generalisation of the usual variational functional is the *bivariational functional* defined by

$$S_H[\tilde{\psi}, \psi] = \frac{\langle \tilde{\psi} | H | \psi \rangle}{\langle \tilde{\psi} | \psi \rangle} = \frac{\text{Tr}(H\rho)}{\text{Tr}(\rho)}, \quad (24)$$

where, unlike in the standard variational principle,  $\langle \tilde{\psi} |$  and  $|\psi\rangle$  are *independent* elements of Hilbert space, and where  $\rho = |\psi\rangle \langle \tilde{\psi}|$ . In the usual variational principle,  $\langle \tilde{\psi} | \equiv |\psi\rangle^\dagger$  which are *not* independent. (However, since  $H$  is *Hermitian*,  $|\psi\rangle$  and  $\langle \tilde{\psi}|$  can be treated as formally independent in the derivations of the stationary conditions.) The key point here

is that  $S_H$  generalises the variational principle to operators that are not necessarily Hermitian. To see this, a straightforward formal calculation yields the following conditions for a stationary  $S_H$ :

$$H |\psi\rangle = E |\psi\rangle, \quad \langle \tilde{\psi} | H = E \langle \tilde{\psi} |. \quad (25)$$

Here,  $E = S_H[\tilde{\psi}, \psi]$  is the value at the stationary point. Thus,  $\langle \tilde{\psi} |$  and  $|\psi\rangle$  are, respectively, left and right eigenvectors of  $H$  with eigenvalue  $E$ .

The bivariational functional is an expectation value functional in the sense defined in Section 3. If we introduce a parameterisation  $z \mapsto (|\psi(z)\rangle, \langle \tilde{\psi}(z)|)$ , we obtain a generalisation of the usual variational principle. This opens up the possibility of more general approximation schemes than the standard variational principle does.

We note that a time-dependent generalisation is readily available, similar to the usual time-dependent action functional in the Hermitian case [6,7]. The time-dependent Schrödinger equation (and its complex conjugate) can be recovered from the stationary conditions of the action-like functional

$$\begin{aligned} \mathcal{A}_H[\tilde{\psi}(\cdot), \psi(\cdot)] \\ = \int_0^T \langle \tilde{\psi}(t) | \psi(t) \rangle^{-1} \langle \tilde{\psi}(t) | \left( i \frac{\partial}{\partial t} - H \right) | \psi(t) \rangle dt, \end{aligned} \quad (26)$$

depending on the whole history of the system for  $t \in [0, T]$ .

Turning to CC theory, the ket ground-state wavefunction is written as  $|\psi\rangle = \exp(T) |\Phi\rangle$ , and the bra ground-state wavefunction as  $\langle \tilde{\psi} | = \langle \Phi | \exp(\tilde{T})$ , where  $\tilde{T} = \tilde{t} X^\dagger$  is a de-excitation operator. This is a completely general parameterisation, assuming  $\langle \Phi | \psi \rangle \neq 0$  and  $\langle \tilde{\psi} | \Phi \rangle \neq 0$ . Plugging into  $S_H$  we get

$$S_H[T, \tilde{T}] = \frac{\langle \Phi | \exp(\tilde{T}) H \exp(T) | \Phi \rangle}{\langle \Phi | \exp(\tilde{T}) \exp(T) | \Phi \rangle}. \quad (27)$$

(The abuse of notation on the left-hand side is deliberate, and hopefully causes no confusion.) If  $(T, \tilde{T})$  is allowed to vary over all untruncated (de)excitation operators,  $S_H[T, \tilde{T}]$  is the exact functional employed in VCC [17]: it is equivalent to FCI within the chosen single-particle basis.

We now do a change of variables  $(T, \tilde{T}) \mapsto (T, S)$ , where  $S$  is a de-excitation operator. We start by noting that

$$\langle \omega | = \frac{\langle \tilde{\psi} | \exp(T)}{\langle \tilde{\psi} | \psi \rangle} \quad (28)$$

satisfies  $\langle \omega | \Phi \rangle = 1$ , so that there exists an  $S = sX^\dagger$  such that  $\langle \omega | = \langle \Phi | \exp(S)$ . Rearranging, we get

$$\langle \tilde{\psi} | = \langle \tilde{\psi} | \psi \rangle \langle \Phi | \exp(S) \exp(-T). \quad (29)$$

This implicitly defines  $S$  as a function of  $(T, \tilde{T})$ . Inserting Equation (29) into Equation (27) yields

$$S_H^{\text{ECC}}[T, S] = \langle \Phi | \exp(S) \exp(-T) H \exp(T) | \Phi \rangle. \quad (30)$$

This functional is exact if  $(T, S)$  is not truncated. Compared to the CC Lagrangian in Equation (17), we see that  $\exp(S) = 1 + \Lambda$ , i.e., we introduce a linked parameterisation of  $1 + \Lambda$ . Truncating  $(T, S)$  at, say, singles and doubles, we obtain the ECC method defined by Arponen [6,9] (hence the superscript). Clearly, standard CC can be viewed as a first-order approximation to ECC in the sense that  $\exp(S) \approx 1 + S$ . At the untruncated limit,  $\exp(S) = 1 + \Lambda$  can be inverted, and ECC and standard CC are equivalent.

Note, however, that  $S_H[T, S]$  is not a Lagrangian as discussed in Section 3. It is, however, an expectation value functional.

As usual in CC theory, one may write

$$S_H^{\text{ECC}}[T, S] = \sum_n \frac{1}{n!} \langle \Phi | \exp(S) \{H T^n\}_L | \Phi \rangle, \quad (31)$$

meaning that, when the expression is expanded using Wick's theorem [18], only those terms survive that contains at least one contraction between each factor  $T$  and  $H$ . A further coordinate transformation  $(S, T) \mapsto (S, \Sigma)$ ,  $\Sigma = \sigma X$ , is given by

$$\sigma_\mu = \langle \Phi | X_\mu^\dagger e^T S | \Phi \rangle \quad (32)$$

with inverse

$$t_\mu = \langle \Phi | X_\mu^\dagger e^{-T} \Sigma | \Phi \rangle. \quad (33)$$

Arponen showed that [6,9,10] parameterising ECC in terms of the new variables gives a considerable simplification, namely,

$$S_H^{\text{ECC,DL}}[\Sigma, S] = \sum_n \frac{1}{n!} \langle \Phi | \exp(S) \{H \Sigma^n\}_L | \Phi \rangle_{\text{DL}}, \quad (34)$$

where 'DL' stands for 'doubly linked'. It indicates that only those terms survive, where each power of  $S$  is linked to either two different  $\sigma_\mu$ s, unless it is linked to  $H$ . In effect, this eliminates some of the terms that would appear in the algebraic expansion (31) and its derivatives.

A couple of observations can be made. Starting from the bivariational expectation value functional, a systematic reduction produces the CC Lagrangian. The main difference in *interpretation* compared to the Lagrangian approach is that  $\Lambda$  is not a Lagrange multiplier, but rather part of the parameterisation of the state  $\rho$ , in a stronger sense than was indicated in Section 3.

A second observation is that, since we started with a completely general variational principle, one could add further degrees of freedom such as the single-particle orbitals themselves. (This would not be identical to the so-called orbital-optimised CC, since in the latter case the orbitals are fixed by auxiliary conditions and not via the bivariational principle.) The method, which we call orbital-adaptive coupled-cluster (OACC) has been suggested and in an earlier paper, and also implemented in the time domain [7].

Thirdly, to the best of this author's knowledge, extended CC has not been implemented for molecular electronic structure calculations. There are, however, some approximations that have been applied, e.g., truncation of the doubly linked ECC functional (34) to finite order in all the amplitudes, see, e.g., Ref. [24]. In Section 6, we discuss OACC, ECC, and truncations that preserve size-consistency.

## 5. Choosing bivariational approximations

The general bivariational expectation value functional  $S_H[\tilde{\psi}, \psi]$  is not real valued in general. Indeed, it takes values in the whole of  $\mathbb{C}$ . It is, therefore, legitimate to ask to what extent it makes sense to introduce arbitrary variational parameters in the bivariational functional. The standard variational principle is, to paraphrase Kramer and Saraceno, a 'blind and dumb procedure that always provides an answer' [25]. This might not be the case with bivariational formulations: it is intuitively clear that some care must be taken when choosing a state parameterisation since we are not, in general, provided with an upper bound to the ground state energy. We now discuss this in terms of a number of claims. We stress that this section deals with *general* uses of the bivariational functional, and is not directly concerned with CC theory, except where noted.

In this section,  $z \in \mathbb{C}^n$ ,  $\rho(z) = |\psi(z)\rangle \langle \tilde{\psi}(z)|$ , where  $\langle \tilde{\psi}(z) | \psi(z) \rangle \equiv 1$  for simplicity. (Note, that by setting  $z = (u, v)$  it is perfectly possible for  $\langle \tilde{\psi} |$  to be completely independent from  $|\psi\rangle$  even if the notation suggests otherwise.) This induces an approximate functional  $S_H[z]$ ,

$$S_H[z] = \text{Tr} (H |\psi(z)\rangle \langle \tilde{\psi}(z)|) = \langle \tilde{\psi}(z) | H | \psi(z) \rangle. \quad (35)$$

The time-dependent action-like functional (26) becomes

$$\begin{aligned} \mathcal{A}_H[z(\cdot)] &= \int_0^T \left\{ i \langle \tilde{\psi}(z(t)) | \frac{\partial}{\partial z} \psi(z(t)) \rangle \cdot \dot{z}(t) - S_H[z(t)] \right\} dt, \end{aligned} \quad (36)$$

where  $\dot{z} = dz/dt$ .

Claim 1: *The state parameterisation  $\rho(z)$  must be complex analytic.*

Suppose to the contrary that it is not.  $S_H[z]$  then effectively depends on  $2n$  real parameters. The stationary

conditions are complex-valued equations numbering  $2n$ , in total  $4n$  equations for  $2n$  variables. Unless the equations are in fact complex analytic ( $n$  complex equations in  $n$  complex variables), a solution cannot be expected to be found.

*Claim 2: The dimension  $n$  in  $z \in \mathbb{C}^n$  must be even.*

Suppose  $z$ , and hence  $|\psi\rangle$  and  $\langle\tilde{\psi}|$ , is not uniquely determined. Then the expectation value functional  $O \mapsto \mathcal{S}_O[z]$ , and hence the physical interpretation, is not unique. If  $n$  is odd, it is straightforward to compute, using the time-dependent formulation (36), that  $z(t)$  is not unique.

Indeed, the stationary condition of the action  $\mathcal{A}_H$  in Equation (26) can be computed [7] to be given by the system of ODEs

$$iC(z(t))\dot{z}(t) = \frac{\partial}{\partial z} \mathcal{S}_H(z(t)), \quad (37)$$

where  $C(z(t))$  is a complex anti-symmetric matrix, namely,

$$C(z)_{jk} = \left\langle \frac{\partial}{\partial z_k} \tilde{\psi} \left| \frac{\partial}{\partial z_j} \psi \right. \right\rangle - \left\langle \frac{\partial}{\partial z_j} \tilde{\psi} \left| \frac{\partial}{\partial z_k} \psi \right. \right\rangle. \quad (38)$$

Any square anti-symmetric matrix is singular if the number of rows and columns is an odd number.

*Claim 3: An approximation must be systematically improvable toward the exact state.*

The bivariational principle does not, in general, prove an upper bound for the energy due to the fact that the expectation value is explicitly non-Hermitian. In order to prove convergence of a bivariational method to the exact ground state, functional analytic considerations must be made that heavily rests on the fact that  $\rho(z)$  systematically approximates exact states  $|\psi\rangle\langle\psi|$ . This is in contrast to truly variational methods, where additional variational parameters always improve the energy.

We note here that a mathematical analysis of the standard CC method, using the CC Lagrangian, has recently been carried out [26], demonstrating that it is indeed convergent, and that the error in the energy is quadratic in the error in  $T$ , similar to variational approximations in the Hermitian sense.

At this point, it is interesting to note that the time-dependent action functional for standard CC becomes

$$\mathcal{A}_H[T(\cdot), \Lambda(\cdot)] = \int_0^T \left[ i \sum_{\mu} \lambda_{\mu} \dot{t}_{\mu} - \mathcal{L}_H[T(t), \Lambda(t)] \right] dt, \quad (39)$$

where  $\mathcal{L}_H$  is the CC Lagrangian (17), i.e., the CC expectation value functional. A straightforward calculation gives the stationary conditions

$$i\dot{t}_{\mu} = \frac{\partial \mathcal{L}_H[T, \Lambda]}{\partial \lambda_{\mu}}, \quad (40a)$$

$$-i\dot{\lambda}_{\mu} = \frac{\partial \mathcal{L}_H[T, \Lambda]}{\partial t_{\mu}}, \quad (40b)$$

which is very similar to Hamilton's equations of motion on complex form [6,9–13,15], and familiar from response theory in quantum chemistry [27,28]. Indeed, one may define real amplitudes  $q_{i\mu}$  and  $p_{i\mu}$ ,  $i = 1, 2$ , via

$$t_{\mu} = q_{1\mu} + ip_{2\mu}, \quad \lambda_{\mu} = q_{2\mu} - ip_{1\mu}.$$

Define  $\mathcal{H}(q, p) = \mathcal{L}_H[T(q, p), \Lambda(q, p)]$ . The equations of motion now read

$$\dot{q}_{i\mu} = \frac{\partial \mathcal{H}(q, p)}{\partial p_{i\mu}}, \quad -\dot{p}_{i\mu} = \frac{\partial \mathcal{H}(q, p)}{\partial q_{i\mu}},$$

which are precisely Hamilton's equations of motion for the Hamiltonian  $\mathcal{H}(q, p)$ . Thus, a symplectic structure exists on the CC parameterisation manifold. In particular, the number of coordinates and momenta separately is an even number, in accordance with Claim 2.

Suppose we are given some arbitrary even-dimensional bivariational approximation (not necessarily of CC type) for which we can find a symplectic structure on the approximation manifold, and for which the resulting Hamiltonian function  $\mathcal{H}$  is smooth. Clearly, all the above Claims 1–3 are satisfied. This indicates that mathematical topics like approximation errors are more easily approached for such approximations, since the machinery of classical mechanics is available.

We remark that the above symplectic structure found for standard CC is found to exist for extended CC as well. In fact, the CI, standard CC and ECC all have such structures derived in the same fashion. This has been studied extensively by Arponen and Bishop [13–15].

## 6. Generalisations of standard CC

The present author is not aware of any implementation of the full ECC method for molecular electronic-structure theory, despite its apparent usefulness. The likely explanation is its higher computational complexity compared to standard CC.

To compute this complexity, suppose that  $H$  is at most a  $k$  body operator, and that we truncate the cluster operators at  $n$ -fold (de)excitations.  $\exp(-T)H\exp(T)$  contains excitations up to order  $(2n-1)k$ , so it is clear that  $\mathcal{S}_H^{\text{ECC}}$  is of finite order in both  $T$  (order  $2k$ ) and  $S$  (at most order  $(2n-1)k$ ). The zeroth and first-order terms are precisely the terms contained in the standard CC Lagrangian, while the higher order terms contain progressively more complicated diagrams.

To explore the structure of the ECC method with a possible numerical computer implementation in mind, a computer algebra program for CC written in Python using the library SymPy [29] has been developed. Using this, we have computed the ECC diagrams to leading order in  $S$  for the doubles only model. The number of diagrams for the



first powers of  $S$  is 3 for  $S^0$ , 13 for  $S^1$ , and 57 for  $S^2$ . Some of the terms can be eliminated via diagram factorisation, as is usually done for CC [18]. Even if  $\mathcal{S}_H^{\text{ECC}}$  truncates at a finite power, the number of Goldstone diagrams is clearly very large compared to standard CC. Even if ECC is probably much more accurate, the complexity of implementation probably outweighs the benefits.

ECC is size-extensive: if  $(T_A, S_A)$  and  $(T_B, S_B)$  are critical points for two non-interacting subsystems  $A$  and  $B$ , then  $(T, S) = (T_A + T_B, S_A + S_B)$  is a critical point for the combined system, and the energy of the whole is the sum of the parts. Moreover, both wavefunctions are multiplicatively separable.

Inspired by the fact that the first-order expansion in  $S$  gives standard CC, which is size-extensive in spite of  $\langle \tilde{\psi} |$  not being multiplicatively separable, we may try higher order expansions in  $S$ , and examine whether we obtain a size-consistent method. This is indeed so. We may even truncate the expansion in powers of  $T$ . We thus obtain a hierarchy of size-extensive methods  $\text{ECC}(n, m)$  with functionals

$$\mathcal{S}_H^{\text{ECC}(n,m)}[T, S] = \langle \Phi | \exp_n(S) \exp_m(\mathcal{C}_T) H | \Phi \rangle, \quad (41)$$

with  $\exp_n(A) \equiv \sum_{i=0}^n \frac{1}{i!} A^i$ , and where  $\mathcal{C}_T H \equiv [H, T]$  is the commutator, which acts as a linear operator on  $H$ . That is, we truncate the BCH expansion for  $\exp(-T) H \exp(T)$  after  $m$  terms.

Let us prove size-extensivity of  $\text{ECC}(n, m)$ . Suppose two subsystems  $A$  and  $B$ , with reference states  $|\Phi_A\rangle$  and  $|\Phi_B\rangle$ , respectively, are separated by distance such that the total Hamiltonian is  $H = H_A + H_B$  and that all subsystem orbitals are orthogonal:  $|\Phi_{AB}\rangle$  is the determinant given by the union of the occupied orbitals in  $|\Phi_A\rangle$  and  $|\Phi_B\rangle$ . Each subsystem has a ground state given by  $(T_A, S_A)$  and  $(T_B, S_B)$ , and  $[S_A, T_B] = [S_B, T_A] = 0$ . We note that  $(T, S) = (T_A + T_B, S_A + S_B)$  describes the totality. We have

$$[H, T] = [H_A, T_A] + [H_B, T_B], \quad (42)$$

which implies that

$$\{HT^j\}_L = \{H_A T_A^j\}_L + \{H_B T_B^j\}_L. \quad (43)$$

We now get

$$\begin{aligned} \mathcal{S}_{H_A+H_B}^{\text{ECC}(n,m)}[T_A + T_B, S_A + S_B] \\ &= \sum_{ij} \frac{1}{i!j!} \langle \Phi_{AB} | (S_A + S_B)^i \{H_A T_A^j + H_B T_B^j\}_L | \Phi_{AB} \rangle \\ &= \sum_{ij} \frac{1}{i!j!} \langle \Phi_{AB} | (S_A^i + S_B^i) \{H_A T_A^j + H_B T_B^j\}_L | \Phi_{AB} \rangle \\ &= \mathcal{S}_{H_A}^{\text{ECC}(n,m)}[T_A, S_A] + \mathcal{S}_{H_B}^{\text{ECC}(n,m)}[T_B, S_B]. \end{aligned}$$

In the transition from the first to the second line, we use that all mixed terms of  $(S_A + S_B)^i$  on the form  $S_A^p S_B^{i-p}$  gives zero contribution. This concludes the proof.

While the full ECC model is probably too expensive, the truncations  $\text{ECC}(n, m)$  may be worthwhile to investigate further. This is an ongoing work in progress, of which the mentioned Python program is a part.

We remark that in Refs. [24,30], a similar approach is taken: the ECC functional is truncated after terms proportional to  $S^4 T^{4-i}$ , i.e., a truncation in the total order. It is unclear whether this preserves size-extensivity, however.

We now turn to OACC. In light of the bivariational nature of CC, it is natural to attempt at adding the orbitals as variational parameters. However, since we must require  $\mathcal{S}_H$  to be complex analytic, we must introduce separate variables for the orbitals  $\phi_p$  and the complex conjugates  $\phi_p^*$ , i.e., we introduce dual orbitals  $\tilde{\phi}_p$  instead of the latter. Orthogonality is replaced by biorthogonality [7], which preserves the fundamental anticommutator,

$$\{\tilde{c}_q, c_p^\dagger\} = \langle \tilde{\phi}_q, \phi_p \rangle = \delta_{qp}, \quad (44)$$

from which it follows that Wick's theorem is unchanged. Thus, the algebraic expressions for every CC expression is unchanged, except that one- and two-electron integrals are basis dependent. As a consequence of the orbital relaxation, the singles amplitudes are redundant.

In the OACC model, the CC amplitude Equations (4) are supplemented with a set of coupled nonlinear eigenvalue-type equations for determining  $\phi_p$  and  $\tilde{\phi}_q$ , similar to Hartree–Fock. In Ref. [7], the time-dependent OATDCCD model (using doubles only, OATDCCD) is considered in detail. Deriving these nonlinear eigenvalue equations is out of scope for the present article, but the derivation of the time-dependent equations in Ref. [7] can readily be adapted.

The orbital-adaptive CC method is a systematic hierarchy of size-extensive approximations to MCTDHF (multiconfigurational time-dependent Hartree–Fock, a special case of time-dependent CASSCF where no orbitals are inactive) which is reproduced in the untruncated limit. It is, therefore, expected that OACC can describe static correlation to some extent.

OACC has not been implemented so far. In Ref. [7] an implementation of OATDCCD tested and shown to produce results that compare very well with a MCTDHF implementation. We comment that in Ref. [8] a similar but not equivalent approach, using biorthogonal orbitals, is taken to linear response for CC.

## 7. Concluding remarks

Two different but equivalent approaches to CC theory in quantum chemistry are taken by Helgaker and Jørgensen [4,5] and Arponen [6]. We have contrasted these

approaches, highlighting their differences and how they are related. A discussion of the bivariational principle, in general, has also been given.

It was shown how the bivariational principle of Arponen arrives at the CC Lagrangian in a top-down manner, with intermediate steps giving extensions of standard CC theory. Adding degrees of freedom, such as the orbitals, gives other generalisations that may improve accuracy.

We have outlined the ECC method [6], OACC [7], and proposed some truncated ECC schemes that may be computationally affordable. These are shown to be size-extensive, and a computer implementation is a work in progress.

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