An accurate local model for triple substitutions in fourth order Møller–Plesset theory and in perturbative corrections to singles and doubles coupled cluster methods

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An accurate local model for triple substitutions in fourth order Møller–Plesset theory and in perturbative corrections to singles and doubles coupled cluster methods

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Abstract

Two noniterative local models for evaluating the contribution of triple substitutions to the electron correlation energy (as needed in MP4 and CCSD(T)), are developed. The occupied space is spanned by a minimal basis, and the virtual space by an extended basis of atom-centered functions. The triple substitutions are truncated by an atomic criterion such that either zero or one electrons can be transferred between atoms. The covalent model asymptotically recovers 70% of the triples correlation energy for poly-ynes with a 6-31G* basis, while the singly-ionic model recovers 99%. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

One of the most important achievements of computational quantum chemistry over the past decade is the development of methods for predicting chemical reaction energies to chemical accuracy or better (1–2 kcal/mol) [1]. The electronic structure methods that have made this possible are those that include the effects of triple substitutions as a correction to singles and doubles coupled-cluster-based methods, such as CCSD(T), QCISD(T) and BD(T) [2–5]. All of these methods are based on the triples contribution in fourth and fifth order Møller–Plesset perturbation theory (MP4 and MP5) [6,2], as are the widely used thermochemical methods G2 and G3 [7,8].

The barrier preventing application of these methods to molecules larger than roughly ten first-row atoms (with present computer resources) is the fact that evaluation of the triples scales with the seventh power of molecule size. Simply doubling the size of the molecule requires 128 times more computing. Accordingly, there have been efforts to accelerate the evaluation of the triples contributions, using Laplace transformation methods [9,10] (which reduce the scaling to 6th power), and using auxiliary basis sets [11] (which reduce disk storage and disk transfers).

An alternative approach to permitting triples calculations on larger molecules is to attempt to fundamentally reduce the computational scaling by imposing a local ansatz. Local models based on double substitutions have been actively developed over the last two decades, focussing mainly on the simplest such method, MP2. The seminal work of Pulay and Saebø [12–14] is based on employing localized

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molecular orbitals to span the occupied space. Double substitutions are restricted to a basin defined as the union of the atomic orbitals on the atoms where the two localized orbitals are located. This local MP2 method reduces the number of double substitutions to asymptotically quadratic in the size of the molecule.

Computational efficiency has been demonstrated by several groups [15–17], with recent reports achieving linear scaling [18] by employing an additional multipole approximation and truncation of the most distant interactions. Alternatively, Laplace transform based MP2 [9] can also approach linear scaling [19]. The Pulay and Saebø local correlation method has also been successfully extended to iterative single and double substitution methods, such as CISD [20,21] and CCSD [22]. It has not been successfully extended to triple substitutions hitherto, because the local amplitude equations must be solved iteratively. This requires storage of the triples amplitudes which are very large in number, even in a local model.

Recently we have introduced an alternative approach to local electron correlation, in which substitutions are represented in terms of atom-centered functions [23–25]. The occupied space is spanned by a minimal basis of distorted atomic orbitals (projected into the occupied space) [26] [2], while the virtual space is spanned by the full set of atomic orbitals, projected into the virtual space. Given this overcomplete representation, local models can be developed based on atomic truncations. At the level of double substitutions, the simplest restriction is to require one occupied and one virtual index to be on the same atom – an atomic substitution. Double substitutions then couple three atoms together: a triatomics in molecules (TRIM) model [24]. If both occupied–virtual substitutions are atomic, the result

is a diatomics in molecules (DIM) model [24] in which each double substitution spans at most two atoms. At the MP2 level, DIM recovers 93–95% of the correlation energy, while TRIM recovers around 99.7%. These atomic truncations are desirable for several reasons. First, they yield smooth potential energy surfaces. Second, the resulting local models can be solved non-iteratively, because the space of retained local substitutions can be written as a direct product. There is no need to store the local amplitudes and hence it is feasible to generalize these models to include triple substitutions.

In this work we report the development of atom-centered local triples methods. The primary objective is then to explore the extent to which triples energies reflect spatially localized contributions, rather than attempt to demonstrate computational efficiency, which we view as a separate second stage. The DIM model of double substitutions will be generalized to a covalent triples model that does not explicitly permit electron transfers between atoms [3]. The TRIM doubles model explicitly permits single electron transfers, and will be generalized to a singly-ionic triples model.

It is not obvious a priori that triple substitutions can be accurately described by a local model. The equations for the triples amplitudes contain a profusion of non-local exchange terms (which arise from three-body antisymmetrizers), so one might reasonably suspect that the triple substitutions would be more non-local than the doubles. Numerical tests on the linear poly-ynes confirm that the triples energy contains relatively large non-local contributions, and an important conclusion of the numerical study is that the non-local contributions are well described by the singly-ionic model.

The Letter is organized as follows: In Section 2 we review the canonical CCSD(T) energy expression and then transform it to a basis of localised orbitals. Section 3 describes the local truncation of the energy expression, leading to our formula for the local correlation energy (Eq. (15)). Section 4 elaborates on the covalent and singly-ionic triples truncation pro-

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[2] The EPAO algorithm for calculating localised orbitals [26] may be viewed as an extension of the Boys algorithm for localised orthogonal orbitals [32] to allow non-orthogonal, linearly dependent orbitals. Eliminating the orthogonality constraint enables the construction of atom-centred orbitals. The number of EPAO's equals the number of atomic orbitals in a minimal basis and is somewhat greater than the number of orthogonal occupied orbitals. Note that we use the O-EPAO (n) procedure.

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[3] In fact, because we employ redundant atom-centered functions to span occupied and virtual spaces, some local ionic character is implicitly present in even the covalent model.

cedures and Section 5 presents an application to the linear poly-ynes.

2. The CCSD(T) correlation energy

2.1. Canonical expression for the CCSD(T) energy

Our starting point is the energy expression derived by Lee et al. [27,28] [4] for restricted closed-shell CCSD(T) in terms of *canonical spatial orbitals*

$$E_{(T)} = 1/3 \sum_{aibjck} \left(W_{aibjck} + V_{aibjck} \right) X_{aibjck} / D_{aibjck} , \tag{1}$$

where W_{aibjck}, V_{aibjck} and X_{aibjck} are given by

$$W_{aibjck} = P_{aibjck} \left[\sum_d t_{aidj} (bd|ck) \right.$$

$$\left. - \sum_l t_{aibl} (lj|ck) \right], \tag{2}$$

$$V_{aibjck} = t_{ai} (bj|ck) + t_{bj} (ai|ck) + t_{ck} (ai|bj) , \tag{3}$$

$$X_{aibjck} = 4W_{aibjck} + W_{akbicj} + W_{ajbki} - 4W_{akbji}$$

$$- W_{aibkcj} - W_{ajbick} \tag{4}$$

and the permutation operator P_{aibjck} is defined by

$$P_{aibjck}(aibjck) = (aibjck) + (bjckai) + (ckaibj)$$

$$+ (aickbj) + (bjaick) + (ckbjai) . \tag{5}$$

D_{aibjck} is the triples energy denominator

$$D_{aibjck} = f_{aa} - f_{ii} + f_{bb} - f_{jj} + f_{cc} - f_{kk} . \tag{6}$$

The matrix elements f_{ii} etc. are diagonal elements of the closed-shell Fock operator, and the t_{ai}, t_{aibj} amplitudes are those defined by Scuseria et al. [29]. The two-electron integrals are raw, i.e. they do not contain exchange terms. In the above equations indices i,j,k refer to occupied spatial orbitals while a,b,c refer to virtual orbitals. The MP4(T) energy expression may be obtained from Eq. (1) by omitting

V_{aibjck} and using the MP2 amplitudes in place of the CCSD amplitudes.

2.2. Matrix version of the energy expression

If W, V and X are regarded as vectors, and D_{aibjck} as an element of a diagonal matrix $\mathbf{D}$,

$$\mathbf{D}_{aibjck, aibjck} = D_{aibjck} , \tag{7}$$

then the energy may be expressed as a scalar product

$$E_{(T)} = 1/3 (W + V)^t \cdot \mathbf{D}^{-1} \cdot X . \tag{8}$$

This matrix version has a number of advantages. It is not encumbered by a plethora of indices and it may readily be transformed from the canonical basis to a localised basis.

2.3. Invariance of the energy expression under basis transformations

The energy expression (Eq. (8)) is invariant under many-particle transformations $\mathbf{T}$ from the canonical basis $(aibjck)$ to a localised basis $(a'i'b'j'c'k')$

$$a'(1)i'(1')b'(2)j'(2')c'(3)k'(3')$$

$$= \sum_{aibjck} a(1)i(1')b(2)j(2')c(3)k(3')$$

$$\times T_{aibjck, a'i'b'j'c'k'} . \tag{9}$$

To see this we note that the vector W transforms as

$$W \rightarrow \mathbf{T}^t \cdot W \tag{10}$$

and similarly for the vectors V and X, while the matrix $\mathbf{D}$ transforms as

$$\mathbf{D} \rightarrow \mathbf{T}^t \mathbf{D} \mathbf{T} . \tag{11}$$

The energy transforms as

$$E_{(T)} \rightarrow 1/3 (W + V)^t \mathbf{T} \cdot \mathbf{T}^{-1} \mathbf{D}^{-1} \mathbf{T}^{t-1} \cdot \mathbf{T}^t X$$

$$= 1/3 (W + V)^t \cdot \mathbf{P} \mathbf{D}^{-1} \mathbf{P}^t \cdot X , \tag{12}$$

where the projection operator $\mathbf{P}$ is defined as

$$\mathbf{P} \equiv \mathbf{T} \mathbf{T}^{-1} . \tag{13}$$

$\mathbf{P}$ represents a projection onto the space spanned by the localised basis. If the localised basis spans the same space as the canonical basis then $\mathbf{P}$ is simply the identity operator and hence does not affect the energy.

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[4] The derivation of this energy expression assumes that the reference state satisfies Brillouin's theorem, i.e. the Fock matrix elements f_{ai} coupling the occupied and virtual orbitals are zero. If the reference state does not obey Brillouin's theorem then the energy expression contains additional terms.

3. Local truncation of the CCSD(T) energy

If the localised basis only spans a local subspace of the canonical space then $\mathbf{T}$ is singular and $\mathbf{P}$ is no longer the identity operator, though it is still Hermitean and idempotent. In this case Eq. (12) defines a local approximation to the energy. $\mathbf{P}$ plays the role of a resolution of the identity in the local triples space, and one can generate a variety of local energy expressions by inserting or removing resolutions of the identity. Two alternatives are;

$$E_{(T)} = 1/3 \, (\mathbf{W} + \mathbf{V})^t \cdot \mathbf{PD}^{-1} \cdot \mathbf{X}, \qquad (14)$$

and

$$E_{(T)} = 1/3 \, (\mathbf{W} + \mathbf{V})^t \cdot \mathbf{D}^{-1} \mathbf{P}^t \cdot \mathbf{X}. \qquad (15)$$

All the local energy expressions (Eqs. (12), (14) and (15)) yield very similar energies. We have found it most convenient to use Eq. (15) to calculate the local energies reported in this Letter.

4. Evaluation of the local correlation energy

Local truncation introduces a systematic error into the correlation energy, and we have assessed the magnitude of this error by adapting a canonical code to produce the locally truncated energy.

The canonical code evaluates the untruncated energy as the scalar product of a row-vector $\left((\mathbf{W} + \mathbf{V})^t \cdot \mathbf{D}^{-1}\right)$ with a column-vector $\mathbf{X}$. The local correlation energy (Eq. (15)) may be evaluated in a similar fashion, as the scalar product of the transformed row-vector $\left((\mathbf{W} + \mathbf{V})^t \cdot \mathbf{D}^{-1}\right) \cdot \mathbf{T}^{-1t}$ with the transformed column vector $\mathbf{T}^t \cdot \mathbf{X}$. The canonical and local algorithms were implemented in a research version of the QChem software package [30].

Transformation of the canonical row-vector requires $\mathbf{T}^{-1}$, the inverse of a very large matrix (Eq. (9)). While more efficient algorithms avoid the canonical basis altogether, every algorithm that we have investigated requires the inverse of a matrix with the same number of indices as $\mathbf{T}$, typically the overlap matrix in the local many-particle basis. In the next section we examine the connection between the structure of the local space and the cost of evaluating the matrix inverse.

4.1. The direct product structure of the local space

Many choices of $\mathbf{T}$ have special structure, enabling $\mathbf{T}^{-1}$ to be formed without difficulty. For example if $\mathbf{T}$ is a product of occupied and virtual orbital transformations $C_{ii'}$ and $C_{aa'}$

$$|i'\rangle = \sum_i |i\rangle C_{ii'}, \qquad (16)$$

$$|a'\rangle = \sum_a |a\rangle C_{aa'}, \qquad (17)$$

$$T_{aibjck, \, a'i'b'j'c'k'} = C_{aa'}C_{ii'}C_{bb'}C_{jj'}C_{cc'}C_{kk'} \qquad (18)$$

or symbolically,

$$\mathbf{T} = \mathbf{C} \otimes \mathbf{C} \otimes \mathbf{C} \otimes \mathbf{C} \otimes \mathbf{C} \otimes \mathbf{C}, \qquad (19)$$

then $\mathbf{T}^{-1}$ may be expressed in terms of the much smaller matrix $\mathbf{C}^{-1}$

$$\mathbf{T}^{-1} = \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} \otimes \mathbf{C}^{-1}. \qquad (20)$$

Local truncation generally destroys the direct-product structure of $\mathbf{T}$ and formation of $\mathbf{T}^{-1}$ is then a formidable problem, unless $\mathbf{T}$ is an orthogonal transformation. In order to surmount this problem we have selected local truncation schemes which preserve some of the direct-product structure. In the next section we consider two particular truncation schemes, one of which is a three-fold direct product of a pair matrix $p_{ai, \, a'i'}$

$$\mathbf{T} = \mathbf{p} \otimes \mathbf{p} \otimes \mathbf{p}, \qquad (21)$$

where

$$p_{ai, \, a'i'} = C_{aa'}C_{ii'} \qquad (22)$$

and the other a four-fold direct product of C and p

$$\mathbf{T} = \mathbf{C} \otimes \mathbf{C} \otimes \mathbf{p} \otimes \mathbf{p}. \qquad (23)$$

4.2. The covalent and singly-ionic local correlation spaces

In this section we define the triple substitutions retained in our local correlation spaces. The triple substitutions are expressed as products of three lo-

calised occupied orbitals and three localised virtual orbitals, so we begin by specifying the choice of localised orbitals.

The localised virtual orbitals $\{a'\}$ are obtained by projecting all the atomic orbitals onto the virtual space. Each virtual orbital can thus be identified with a particular atom, and we subsequently exploit this atom label when constructing the local correlation space. The number of localised virtual orbitals is somewhat larger than the number of canonical virtual orbitals, but the spaces spanned by the two sets are the same, implying that the non-orthogonal localised orbitals contain a number of linear dependencies.

A minimal basis of localised occupied orbitals $\{i'\}$ is obtained for each atom via the EPAO algorithm [26] [5], and by construction each localised orbital has a natural atom label. This non-orthogonal set of localised orbitals spans the same space as the canonical orbitals and contains a number of linear dependencies.

As mentioned in the previous section, the complete set of triple substitutions is a six-fold direct product

$$\{a'i'b'j'c'k'\}_{\text{full}} = \{a'\} \otimes \{i'\} \otimes \{b'\} \otimes \{j'\}$$
$$\otimes \{c'\} \otimes \{k'\}. \qquad (24)$$

Each triple substitution $a'i'b'j'c'k'$ is the product of three single substitutions, $a' \leftarrow i'$, $b' \leftarrow j'$ and $c' \leftarrow k'$. In the most severe local truncation procedure, referred to as the *covalent* model, only *atomic single substitutions are retained*. That is, a' and i' must have the same atom label. We denote an atomic single substitution by $(a'i')$ and the set of all atomic single substitutions by $\{(a'i')\}$. The set of covalent triple substitutions is then a three-fold direct product of atomic single substitutions

$$\{a'i'b'j'c'k'\}_{\text{Covalent}} = \{(a'i')\} \otimes \{(b'j')\}$$
$$\otimes \{(c'k')\}. \qquad (25)$$

The corresponding many-particle transformation $\mathbf{T}_{\text{Covalent}}$ has the same direct-product structure. It may be conveniently expressed in terms of the local pair

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[5] See footnote [2].

transformation (22) $p_{ai,(a'i')}$ where a' and i' have the same atom label

Covalent: $T_{aibjck,(a'i')(b'j')(c'k')}$

$$= p_{ai,(a'i')} p_{bj,(b'j')} p_{ck,(c'k')}. \qquad (26)$$

The Covalent space includes long-range correlation (dispersion) since no restriction is placed on the distance between pairs of atomic substitutions $(a'i')$ and $(b'j')$. The main failing of the Covalent ansatz is that it excludes charge-transfer substitutions, since each single substitution is largely confined to an atom. Charge-transfer substitutions are an important component of chemical bonding, and this has prompted us to develop a more accurate singly-ionic space which includes these substitutions [24].

The *singly-ionic* space is constructed from three local spaces, each of which allows charge-transfer in one of the single substitutions $a' \leftarrow i'$, $b' \leftarrow j'$ or $c' \leftarrow k'$

$$\{a'i'b'j'c'k'\}_{\text{Ionic1}} = \{a'\} \otimes \{i'\} \otimes \{(b'j')\}$$
$$\otimes \{(c'k')\}, \qquad (27)$$
$$\{a'i'b'j'c'k'\}_{\text{Ionic2}} = \{(a'i')\} \otimes \{b'\} \otimes \{j'\}$$
$$\otimes \{(c'k')\}, \qquad (28)$$
$$\{a'i'b'j'c'k'\}_{\text{Ionic3}} = \{(a'i')\} \otimes \{(b'j')\}$$
$$\otimes \{c'\} \otimes \{k'\}. \qquad (29)$$

Thus Ionic1 does not place any restrictions on the substitution $a' \leftarrow i'$, but confines $b' \leftarrow j'$ and $c' \leftarrow k'$ to be atomic substitutions. The corresponding many-particle transformation is

Ionic1: $T_{aibjck, a'i'(b'j')(c'k')}$

$$= C_{aa'} C_{i,i'} p_{bj,(b'j')} p_{ck,(c'k')} \qquad (30)$$

and similarly for Ionic2 and Ionic3. The union of the three spaces

$$\{\text{Ionic}\} = \{\text{Ionic1}\} \cup \{\text{Ionic2}\} \cup \{\text{Ionic3}\} \qquad (31)$$

contains all triple substitutions involving at most one charge-transfer substitution.

The Ionic space includes all the substitutions which we wish to include in the local correlation space, but it is not a simple direct-product space and so formation of $\mathbf{T}^{-1}$ is not feasible. A viable approximation consists of evaluating the Ionic1, Ionic2 and Ionic3 energies, and then extracting their con-

tributions to the charge-transfer energy, ($E_{\text{Ionic1}} - E_{\text{Covalent}}$), ($E_{\text{Ionic2}} - E_{\text{Covalent}}$) and ($E_{\text{Ionic3}} - E_{\text{Covalent}}$). The total Ionic energy is then taken to be the sum of the Covalent energy and the three charge-transfer contributions

$$
\begin{aligned}
E_{\text{Ionic}} &= E_{\text{Covalent}} + E_{\text{charge-transfer}} \\
&= E_{\text{Covalent}} + (E_{\text{Ionic1}} - E_{\text{Covalent}}) \\
&\quad + (E_{\text{Ionic2}} - E_{\text{Covalent}}) + (E_{\text{Ionic3}} - E_{\text{Covalent}}) \\
&= E_{\text{Ionic1}} + E_{\text{Ionic2}} + E_{\text{Ionic3}} - 2 E_{\text{Covalent}} .
\end{aligned} \quad (32)
$$

5. Asymptotic correlation energy recovery in the linear poly-ynes

A standard measure of the accuracy of local correlation is the fraction of the untruncated energy recovered. Generally the recovery rate drops from 100% for single atoms towards an asymptotic limit for large molecules. In order to probe the asymptotic limit we have calculated the triples contribution to the MP4 energy as a function of chain length for the linear poly-ynes $C_n H_2$, using a 6-31G* basis. The triple bonds in the poly-yne chain make a sizeable contribution to the triples energy, and their delocalised nature provides a stringent test of the local truncation procedure.

In Fig. 1 the percentage of the untruncated MP4(T) energy recovered by the covalent and singly-ionic methods is plotted for each homologous increment to the poly-yne chain.

It is interesting to compare the covalent triples results with our earlier results for the covalent doubles energy [24]. Previously we found that the covalent ansatz recovers 93–95% of the MP2 doubles energy for the linear alkanes and polyenes, and additional calculations show that it recovers 96% of the MP2 doubles energy for the poly-ynes. In contrast the present results indicate that the covalent ansatz only recovers 70% of the MP4 triples energy. Evidently the charge-transfer substitutions neglected by the covalent ansatz are much more important for the triples energy than for the doubles energy.

The performance of the covalent triples method varies somewhat with the size of the atomic-orbital basis set. The covalent triples method becomes exact
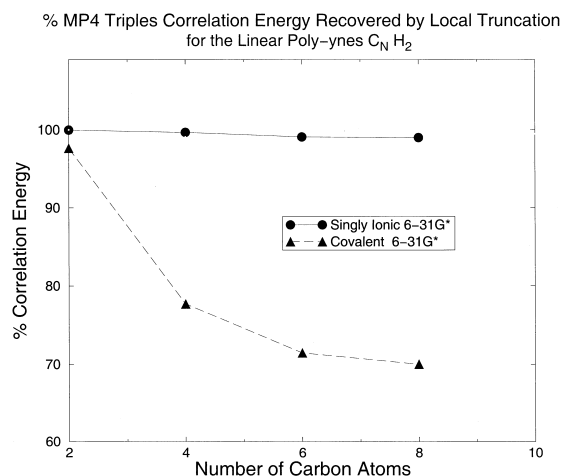

Fig. 1. Percentage of the untruncated MP4(T) correlation energy recovered by the covalent and singly-ionic local correlation spaces for linear poly-yne chains $C_n H_2$ of increasing length. In order to exhibit the asymptotic limit we have plotted the fractional energy recovery for each new increment to the chain length. The 6-31G* basis was used, and all electrons are correlated, including the 1s orbitals. The geometries are not fully optimised, but are close to the equilibrium structures. The C–H bonds are 1.0 Å, the C–C single bonds are 1.4 Å and the triple bonds are 1.2 Å. The singly-ionic space captures 99.0% of the correlation energy in the asymptotic limit.

in the infinite basis limit, while at the other extreme local truncation causes larger errors for very small basis sets. Indeed, the covalent ansatz performs very poorly in an STO-3G basis, recovering only 29% of the triples energy. Interestingly, if the atomic basin of single substitutions is expanded to include nearest-neighbour substitutions ($nn = 1$, in the terminology of an earlier paper [31]), the covalent ansatz recovers 99.9% of the STO-3G triples energy. Thus it appears that the charge-transfer substitutions are very short-range.

The singly-ionic space asymptotically recovers 99.0% of the untruncated 6-31G* triples energy, a remarkable improvement over the covalent results. This performance is particularly pleasing because the singly-ionic space neglects all triple substitutions involving two or three charge-transfer substitutions. The covalent and singly-ionic results indicate that atomic substitutions account for roughly 70% of the triples energy, single-charge-transfer substitutions account for 30%, and double and triple charge-trans-

fer substitutions account for just 1%. Thus the singly-ionic space appears to include the important single-charge-transfer substitutions while retaining only $\mathscr{O}(N^4)$ of the $\mathscr{O}(N^6)$ triples amplitudes.

The singly-ionic space appears to be sufficiently accurate for many chemical applications. For example thermo-chemical applications generally require an accuracy of 1 kcal/mol for energy differences. For C_8H_2, the largest poly-yne considered in our study, the canonical MP4(T) energy is 38.9 kcal/mol. The singly-ionic ansatz introduces an absolute error of just 0.2 kcal/mol, well within the desired range.

6. Conclusion

(1) Two local models for the contribution of triple substitutions to the MP4 and CCSD(T) correlation energy have been developed and tested. These models use redundant atom-centered occupied and virtual orbitals, together with atom-based local truncations. The two models restrict transfer of electrons between atomic regions; the covalent model excludes charge-transfer substitutions while the singly-ionic model permits a single charge-transfer.

(2) These atomic local triples models have two attractive features as local correlation procedures. First, they may be evaluated noniteratively, so that storage of triples is not necessary. The number of local triples amplitudes grows cubically with system size for the covalent model and quartically for the singly-ionic model, versus a sixth-order increase for the untruncated amplitudes. The second advantage is that the atomic local truncation is parameter-free and gives inherently smooth potential energy surfaces.

(3) We have developed a pilot code to implement these local methods, and have tested what fraction of the full (nonlocal) triples correlation energy they recover. For poly-ynes using the 6-31G* basis, the covalent model recovers asymptotically only 70% of the triples correlation energy. Interestingly, the covalent model does much better for doubles correlation, recovering 96% of the MP2 energy. The singly-ionic model recovers 99% of the triples correlation energy, which indicates that the contribution of two and three-electron transfers may be neglected without significant error.

(4) The present results establish that an efficient implementation of the singly-ionic model should be very useful for chemical applications. It remains to demonstrate computational efficiency of the singly-ionic model in a production code, a task in which we are presently engaged. The fact that our present results demonstrate the inherent localizability of triples correlation energies for the first time will also be motivation for seeking other novel local triples methods.

Acknowledgements

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