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# Compressive Sampling in Configuration Interaction Wavefunctions

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The problem is considered of generating approximate quantum-mechanical wavefunctions that have as many as possible coefficients held exactly to zero for a given desired accuracy. Two approaches are adopted. In the first, perturbation theory within the Davidson diagonalisation algorithm is used to mask off small coefficients in the wavefunction against a predefined target energy threshold. Secondly, sparsity is introduced by penalty-function optimisation, with a norm-based compressive-sampling penalty function that decreases with increasing sparsity. The first approach is found to be robust and reliable, whereas the second does not succeed in keeping the wavefunction sparse.

## 1. Introduction

It has long been recognised that configuration interaction (CI) wavefunctions for the electrons in molecules typically contain both near-redundant information, and considerable sparsity. The former arises typically from disconnected products of excitations from a dominant reference wavefunction, whilst the latter can originate in the spatial sparsity of the Hamiltonian operator (small couplings between spatially distant orbitals) and the structure of couplings to highly excited states through multiple applications of the two-body Hamiltonian.

Exploitation of such sparsity patterns is very commonly done by choosing a truncated ansatz - for example, restricting excitations from a reference wavefunction to be just those that move one or two electrons, or adopting a formulation like the coupled cluster (CC) method where disconnected excitation products are brought in automatically. Alternatively, one may look for them numerically in a general full configuration interaction (FCI) procedure[1]; there have been numerous adaptive configuration-selection algorithms expored over many years [2–9]. This is also one way of viewing algorithms such as the Density-Matrix Renormalisation Group method (DMRG)[10, 11]

In a pioneering contribution that established the potential for performing FCI calculations [1], Handy showed that it was possible to exploit the sparsity in both the Hamiltonian and the wavefunction to allow reduced-precision representation of those quantities without significant degradation of the quality of the computed wavefunction and energy. The resulting savings in computer storage made possible calculations that at the time would not otherwise have been feasible. Further dynamic pruning of the CI wavefunction can be achieved by selection of configurations on the basis of perturbation theory[6, 8, 12–15].

General data compression technologies are ubiquitous in many areas of science and engineering, and have been applied in this context to seek optimally sparse representations of FCI wavefunctions[16]. An important signal-processing technique

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is that of Compressive Sampling[17, 18], in which signal data patterns that are incoherently spread across a sparse data object can be discovered very efficiently through automatic algorithms.

In this paper, the possibility is explored of automatically and optimally retaining an optimally sparse wavefunction whilst computing a Hamiltonian eigenvalue to within an initially prescribed tolerance.

## 2. Methods

All practical computational electronic structure computations are performed with finite precision and finite accuracy. There is typically an intrinsic error associated with the choice of $N$-electron methodology used to approximate FCI, plus a further systematic 1-electron basis-set truncation error. In addition, there are precision errors that arise from the use of finite-precision computer arithmetic (these are usually insignificant), from the incomplete iterative solution of the defining equation systems, and from finite thresholds that truncate small intermediate quantities. The latter are usually introduced to give rise to a reduced computational effort by avoiding arithmetic and data movement that does not lead to a significant contribution to the result[3, 14, 15, 19]. Thresholds are typically chosen ad-hoc on the basis of experience, although there are notable exceptions where full error control is exercised through a single numerical parameter that specifies the desired maximum error in the result of the computation [20, 21]

In this study, we consider FCI wavefunctions, for which the only systematic error is basis-set truncation, and focus entirely on the control of precision associated with seeking only an incomplete, approximate eigensolution of the hamiltonian matrix. The conclusions are expected to be transferrable in an obvious way to any other correlated wavefunction ansatz.

The CI eigenvalue problem is usually solved using a preconditioned form of the Lanczos algorithm as proposed by Davidson[22, 23]. At iteration $n$, the eigenvector of the $N \times N$ hamiltonian matrix $\mathbf{H}$ is represented as a linear combination of a sequence of expansion vectors $\{\mathbf{c}_m, m \leq n \ll N\}$,

$$\mathbf{c}'_n = \sum_m^n \alpha_m \, \mathbf{c}_m \tag{1}$$

For the lowest eigenvalue, $\boldsymbol{\alpha}$ is obtained by minimising the expectation value of the energy, leading to the reduced eigenvalue problem

$$\overline{\mathbf{H}} \, \boldsymbol{\alpha} = \overline{\mathbf{S}} \, \boldsymbol{\alpha} \, E \tag{2}$$

where

$$\boldsymbol{\sigma}_n = \mathbf{H} \, \mathbf{c}_n \tag{3}$$

$$\overline{H}_{mn} = \mathbf{c}_m^\dagger \, \boldsymbol{\sigma}_n \tag{4}$$

$$\overline{S}_{mn} = \mathbf{c}_m^\dagger \, \mathbf{c}_n \tag{5}$$

We then construct the residual as

$$\mathbf{g}_n = (\mathbf{H} - E) \, \mathbf{c}'_n \tag{6}$$

and the new solution as

$$\mathbf{c}_{n+1} = \mathbf{c}'_n - \mathbf{G}\,\mathbf{g}_n \tag{7}$$

where $\mathbf{G} = (\mathbf{H}^0 - E\mathbf{1})^{-1}$ is a preconditioner usually based on the diagonal elements of $\mathbf{H}$, $\mathbf{H}^0_{IJ} = H_{II}\,\delta_{IJ}$, and motivated by perturbation theory. $\mathbf{c}_{n+1}$ may then be used directly as the next expansion vector, or it can be orthogonalised to the existing vectors.

Once $n$ exceeds half the number of electrons, the successive applications of the two-body hamiltonian result in $\mathbf{c}_{n+1}$ being completely dense, apart from any sparsity that arises from symmetry. There are, however, computational advantages if the expansion vectors can be kept sparse. Not only will storage be saved, but typically the cost of evaluating $\boldsymbol{\sigma}_{n+1}$ can be reduced. We therefore look for strategies to modify $\mathbf{c}_{n+1}$ such that small numbers are replaced by zero, without significant degradation of the quality of the final eigenvalue. Note that provided the truncation strategy supports eventual sampling of the entire space, only the convergence of the iterative process, and not the final eigenvalue, is affected.

We first consider the truncation of expansion vectors according to the estimated contribution of individual coefficient updates to the final eigenvalue. At leading order in $c_{I,n+1} - c'_{I,n}$, the contribution of the update in $c_I$ to the eigenvalue is

$$\Delta_I E = (c_{I,n+1} - c'_{I,n}) g_I = -G_{II}\, g_I^2. \tag{8}$$

In previous work[3, 14, 15], whenever $|\Delta_I E| \le \eta$ where $\eta$ is a prescribed ad-hoc threshold, the update was set to zero; here, we choose $\eta$ dynamically such that

$$\sum_{I}^{|\Delta_I E| \le \eta} |\Delta_I E| = \epsilon \tag{9}$$

where $\epsilon$ is a single parameter expressing the desired precision to which the eigenvalue is computed. The value of $\eta$ that satisfies (9) is determined using a simple bisection algorithm on $\log \eta$ at negligible cost. $\epsilon$ is also used to control the termination of the iterative process, completing when the eigenvalue change in successive iterations is less than $\epsilon$. This algorithm gives automatic compression of the wavefunction based on a single input parameter

A second approach to introducing sparsity is that of Compressive Sampling[17, 18]. The principal algorithmic idea is to minimise a functional that is the energy plus a penalty function that grows with the fractional population of the eigenvector.

We define the $L_k$ norm of the eigenvector as

$$L_k = \sum_{J}^{c_J \ne 0} |c_J|^k = \sum_J c_J^k \,(\operatorname{sgn} c_J)^{-k} \tag{10}$$

where

$$\operatorname{sgn} x = \begin{cases} 1 & x \ge 0 \\ -1 & x < 0 \end{cases} \tag{11}$$

and powers of negative numbers are defined as principal values. We will normally choose $\boldsymbol{\alpha}$ such that $L_2 = 1$. For a fully sparse vector $\mathbf{c} = (1, 0, 0, \dots)$, then $L_k = 1$

for all $k$; for a vector whose components are all equal, $\mathbf{c} = (1, 1, 1, \dots) N^{-1/2}$, leading to $L_k = N^{1-k/2}$.

We then introduce a penalised energy functional that is invariant to overall scaling of $\mathbf{c}$:

$$F_{k,l} = E + \mu \, P_{k,l} \tag{12}$$

$$P_{k,l} = \operatorname{sgn}(l(2-k)) \, k^{-1} \, l^{-1} \, (L_k^l L_2^{-k\,l/2} - 1) \tag{13}$$

The penalty function $P_{k,l}$ is positive semidefinite, and is zero for the special case of all but one of the components of $\mathbf{c}$ being zero, or of $k = 2$, and otherwise for $l = 0$ is equal to unity. In general, the sparser is $\mathbf{c}$, the smaller is $P_{k,l}$. Thus for a penalty strength $\mu > 0$, minimisation of $F_{k,l}$ tends to increase larger and decrease smaller components of $\mathbf{c}$. $k = 1, l = 1$ corresponds to the previously-considered choice[17, 18] of enhancing sparsity by penalising with the $L_1$ norm. Note that $P_{k,l}$ is continuous for $k \geq 0$, but can have discontinuous derivatives wherever any coefficient is zero. Its differential properties are

$$\frac{\partial P_{k,l}}{\partial c_I} = \operatorname{sgn}(l(2-k)) \, (L_k L_2^{-k/2})^l \, \left( c_I^{k-1} \, (\operatorname{sgn} c_I)^{-k} L_k^{-1} - c_I \, L_2^{-1} \right) \tag{14}$$

$$= \operatorname{sgn}(l(2-k)) \, (L_k L_2^{-k/2})^l \, \left( |c_I|^{k-2} \, L_k^{-1} - L_2^{-1} \right) \, c_I \tag{15}$$

$$\begin{aligned} \frac{\partial^2 P_{k,l}}{\partial c_I \, \partial c_J} &= \operatorname{sgn}(l(2-k)) \, (L_k L_2^{-k/2})^l \\ &\times \Big\{ l \, k \, \left( c_I^{k-1} \, (\operatorname{sgn} c_I)^k L_k^{-1} - c_I \, L_2^{-1} \right) \left( c_J^{k-1} \, (\operatorname{sgn} c_J)^k L_k^{-1} - c_J \, L_2^{-1} \right) \\ &+ \delta_{IJ} \left( (k-1) c_I^{k-2} \, (\operatorname{sgn} c_I)^k L_k^{-1} - L_2^{-1} \right) \\ &- k L_k^{-2} c_I^{k-1} (\operatorname{sgn} c_I)^k c_J^{k-1} (\operatorname{sgn} c_J)^k + 2 L_2^{-2} \, c_I \, c_J \Big\} \end{aligned} \tag{16}$$

Thus $P_{k,l}$ has continuous slope only if $k \geq 2$, and for $k < 1$ the gradient diverges as any $c_I$ tends to zero. Note that it is still possible to carry out minimisation when $k < 2$, since zero coefficients are explicitly excluded in the definition (10) of $L_k$, but convergence might be erratic.

We then approach the choice of penalty strength $\mu$ by making it consistent with the threshold $\epsilon$ for error in the eigenvalue. We seek $E_{kl}(\mu) - E(0) \leq \epsilon$, where $E_{kl}(\mu)$ denotes the value of $E$ obtained by minimising $F_{kl}$ for a given value of $\mu$. $E_k(\mu)$ can be calculated using perturbation theory. The minimum condition $\mathbf{g} = 0$,

$$g_I \equiv \tfrac{1}{2} \frac{\partial F_{k,l}}{\partial c_I} = \tfrac{1}{2} \frac{\partial E}{\partial c_I} + \tfrac{1}{2} \mu \, \frac{\partial P_{k,l}}{\partial c_I} \quad , \tag{17}$$

is satisfied for all $\mu$, and so

$$0 = \frac{\partial^2 E}{\partial c_I \partial c_J} \frac{d c_J}{d\mu} + \mu \frac{\partial^2 P_{k,l}}{\partial c_I \partial c_J} \frac{d c_J}{d\mu} + \frac{\partial P_{k,l}}{\partial c_I} \tag{18}$$

$$= \frac{\partial^2 E}{\partial c_I \partial c_J} \frac{d c_J}{d\mu} + \frac{\partial P_{k,l}}{\partial c_I} + O(\mu) \tag{19}$$

which are a set of linear equations for $d\mathbf{c}/d\mu$.

In the specific case of CI, $E$ is the Rayleigh quotient

$$E = \frac{\mathbf{c}^\dagger \mathbf{H} \mathbf{c}}{\mathbf{c}^\dagger \mathbf{c}} \tag{20}$$

$$\frac{\partial E}{\partial c_I} = 2(\mathbf{c}^\dagger \mathbf{c})^{-1}(\mathbf{H}\mathbf{c} - E\mathbf{c})_I = O(\mu) \tag{21}$$

$$\frac{\partial^2 E}{\partial c_I \partial c_J} = 2(\mathbf{c}^\dagger \mathbf{c})^{-1}(H_{IJ} - E\delta_{IJ}) - 4(\mathbf{c}^\dagger \mathbf{c})^{-2}(\mathbf{H}\mathbf{c} - E\mathbf{c})_I \, c_J - 4(\mathbf{c}^\dagger \mathbf{c})^{-2}(\mathbf{H}\mathbf{c} - E\mathbf{c})_J \, c_I \tag{22}$$

$$= 2(\mathbf{c}^\dagger \mathbf{c})^{-1}(H_{IJ} - E\delta_{IJ}) + O(\mu) \quad . \tag{23}$$

The first-order variation of $E$ with $\mu$ is then

$$\frac{\partial E}{\partial \mu} = \frac{\partial E}{\partial c_I}\frac{dc_I}{d\mu} \tag{24}$$

$$= \frac{\partial P_{kl}}{\partial c_I} c_I \tag{25}$$

$$= \left( \operatorname{sgn}(l(2-k)) \left(L_k L_2^{-k/2}\right)^l \sum_I \left( |c_I|^{k-2} L_k^{-1} - L_2^{-1} \right) c_I^2 \right) \tag{26}$$

$$= 0, \tag{27}$$

which is consistent with the variational principle, and so we have to consider $d^2E/d\mu^2$ in establishing how the energy rises in response to the penalty strength $\mu$.

Note also that the principal effect of $l$ is to scale the value of $\partial P_{k,l}/\partial c_I$, but this scaling is immediately undone by its incorporation in the value of $\mu$. Thus there is no obvious reason to choose any value of $l$ other than 1.

The second differential of the variational condition is

$$0 = \frac{\partial^3 E}{\partial c_I \partial c_J \partial c_K}\frac{dc_J}{d\mu}\frac{dc_K}{d\mu} + \frac{\partial^2 E}{\partial c_I \partial c_J}\frac{d^2 c_J}{d\mu^2} + 2\frac{\partial^2 P_{k,l}}{\partial c_I \partial c_J}\frac{dc_J}{d\mu} + O(\mu) \tag{28}$$

$$\begin{aligned} &= 2(\mathbf{c}^\dagger \mathbf{c})^{-1}(H_{IJ} - E\delta_{IJ})\frac{d^2 c_J}{d\mu^2} \\ &- 4c_I(\mathbf{c}^\dagger \mathbf{c})^{-2}(H_{JK} - E\delta_{JK})\frac{dc_J}{d\mu}\frac{dc_K}{d\mu} \\ &+ 2\frac{\partial^2 P_{k,l}}{\partial c_I \partial c_J}\frac{dc_J}{d\mu} + O(\mu) \end{aligned} \tag{29}$$

From this, we can synthesise

$$\frac{d^2 E}{d\mu^2} = \frac{\partial^2 c_I}{d\mu^2}\frac{\partial E}{\partial c_I} + \frac{\partial c_I}{d\mu}\frac{\partial c_J}{d\mu}\frac{\partial^2 E}{\partial c_I \partial c_J} \tag{30}$$

which after some manipulation reduces to

$$\frac{d^2 E}{d\mu^2} = -\sum_I \frac{dc_I}{d\mu}\frac{\partial P_{k,l}}{\partial c_I} \tag{31}$$

Table 1. Truncation of FCI wavefunction for $C_2$, cc-pVDZ basis set, as a function of energy truncation threshold $\epsilon$, with no compressive sampling penalty. $\Delta E$ is the error in the energy (Hartree), and $q$ is the fraction of CI coefficients that are non-zero. The molecular orbital basis is the set of ground-state Hartree-Fock orbitals.

| $\epsilon$ | $\Delta E$ | $q$ |
|---|---|---|
| 0.01000000 | 0.00954013 | 0.00082 |
| 0.00300000 | 0.00298976 | 0.00329 |
| 0.00100000 | 0.00119440 | 0.01173 |
| 0.00030000 | 0.00029657 | 0.02338 |
| 0.00010000 | 0.00010243 | 0.05689 |
| 0.00003000 | 0.00003136 | 0.10216 |
| 0.00001000 | 0.00001041 | 0.18938 |
| 0.00000300 | 0.00000301 | 0.29849 |
| 0.00000100 | 0.00000112 | 0.40036 |
| 0.00000030 | 0.00000032 | 0.51379 |
| 0.00000010 | 0.00000009 | 0.62313 |
| 0.00000003 | 0.00000003 | 0.73109 |
| 0.00000001 | 0.00000001 | 0.81625 |

The coupled linear first-order equations (19) need to be solved to get $dc_I/d\mu$. We do this by projecting onto the subspace of expansion vectors, in a generalisation of the Davidson eigensolution algorithm. The projected form of (19) gives $d\alpha/d\mu$ at $\mu = 0$, which in turn leads to a subspace approximation to $d^2E/d\mu^2$. We then choose

$$\mu = \sqrt{\frac{2\epsilon}{d^2E/d\mu^2}} \tag{32}$$

such that the compressive sampling is expected to degrade the final expectation value of the energy $E(\mu)$ by at most $\epsilon$. Once $\mu$ is determined, we seek the $\boldsymbol{\alpha}$ that gives zero projection of (17) onto the subspace,

$$\overline{g}_m = \mathbf{c}_m^\dagger \, \mathbf{g}_n \qquad \text{for all } m \leq n \tag{33}$$

where the residual is now non-linear:

$$\mathbf{g}_n = (\mathbf{H} - E)\,\mathbf{c}'_n + \tfrac{1}{2}\mu \left.\frac{\partial P_{k,l}}{\partial c_I}\right|_{\mathbf{c}'_n} \qquad E = \frac{\mathbf{c}'^\dagger_n \, \mathbf{H} \, \mathbf{c}'_n}{\mathbf{c}'^\dagger_n \, \mathbf{c}'_n} \tag{34}$$

$$= \sum_m^n \alpha_m(\boldsymbol{\sigma}_m - E\,\mathbf{c}_m) + \tfrac{1}{2}\mu \left.\frac{\partial P_{k,l}}{\partial c_I}\right|_{\mathbf{c}'_n} \qquad E = \frac{\boldsymbol{\alpha}^\dagger \, \overline{\mathbf{H}} \, \boldsymbol{\alpha}}{\boldsymbol{\alpha}^\dagger \, \overline{\mathbf{S}} \, \boldsymbol{\alpha}} \tag{35}$$

These subspace equations have to be iterated to convergence, constructing $\mathbf{c}'_n$ and evaluating the projected penalty derivative in each microiteration.

## 3. Results

Table 1 shows the results of FCI calculations on $C_2$ at a bond length of 1.1 Å. The ${}^1\Sigma_g^+$ Hartree-Fock orbitals of the cc-pVDZ basis set[24] were used, with the two lowest energy orbitals omitted, yielding a FCI energy of −75.67189968 Hartree. Energy-directed truncation of Davidson expansion vectors following equation (9) was used, without compressive sampling. It is seen that the actual errors in the

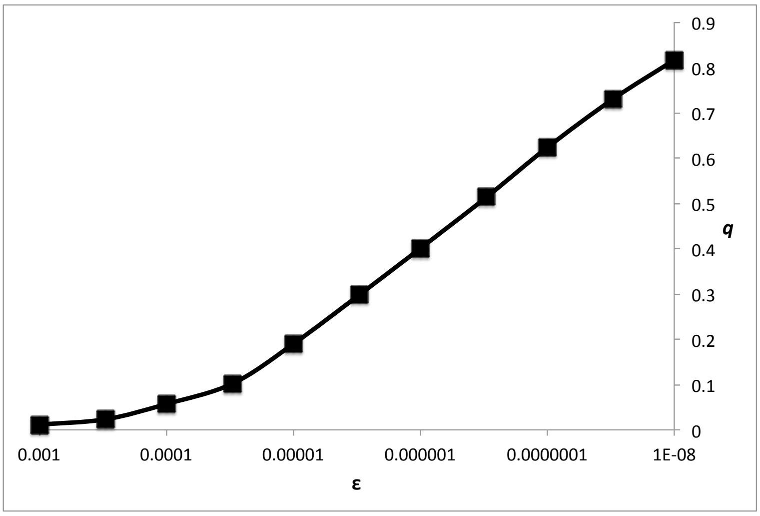

Figure 1. Population $q$ of FCI wavefunction as a function of energy truncation threshold for $C_2$

Table 2. Truncation of FCI wavefunction for $C_2$, cc-pVDZ basis set, as a function of compressive sampling power $k$, with $\epsilon = 3 \times 10^{-5}$. $\Delta E$ is the error in the energy (Hartree), and $q$ is the fraction of CI coefficients that are non-zero. The molecular orbital basis is the set of ground-state Hartree-Fock orbitals.

| $k$ | $\Delta E$ | $q$ |
|---|---|---|
| 0.50000 | 0.00007483 | 0.13202 |
| 1.00001 | 0.00011198 | 0.11333 |
| 1.50000 | 0.00004367 | 0.10419 |
| 2.00000 | 0.00003136 | 0.10216 |
| 2.50000 | 0.00003658 | 0.10172 |
| 3.00000 | 0.00003486 | 0.10156 |
| 3.50000 | 0.00003395 | 0.10148 |
| 4.00000 | 0.00003351 | 0.10143 |

eigenvalue, $\Delta E$, match the target threshold $\epsilon$ faithfully, supporting the validity of the underlying perturbative assumptions, even though in this example, the Hartree-Fock determinant is not strongly dominant. Also shown is $q$, the fraction of non-zero CI coefficients, which increases steadily as $\epsilon$ approaches zero. Figure 1 shows that in the range $10^{-4} \leq \epsilon \leq 10^{-8}$, this variation is approximately linear in $\log \epsilon$, but outside this range $q$ tends smoothly to its limiting constant values.

Table 2 shows the effect of compressive sampling for $\epsilon = 3 \times 10^{-5}$, chosen as typical for chemical applications with precision errors kept below $0.1\,\mathrm{kJ\,mol^{-1}}$. Calculations with a number of different $k$ values and $l = 1$ are shown, including $k = 2$ which is the case of no compressive sampling. It is seen that $\Delta E$ hardly changes at all with $k$, indicating that the compressive sampling is not effective in discovering exploitable sparsity.

Tables 3 and 4 show the same information for a calculation on the OH molecule at a bond length of $1.0\,\mathrm{\AA}$, using the cc-pVTZ basis set[24] with O $f$ and H $d$ functions omitted, spin-restricted Hartree-Fock theory, and the $1\sigma$ orbital frozen. The FCI energy is $-75.61667454$ Hartree. This example contains fewer electrons but more virtual orbitals, and the correlation effects are predominantly dynamic. Exactly the same behaviour is seen as for $C_2$: energy threshold truncation is effective in introducing sparsity, but penalising with $P_{k,l}$ is not.

As a final example, we consider ten hydrogen atoms arranged collinearly with

Table 3. Truncation of FCI wavefunction for OH, cc-pVTZ basis set (O $f$ and H $d$ functions omitted), as a function of energy truncation threshold $\epsilon$, with no compressive sampling penalty. $\Delta E$ is the error in the energy (Hartree), and $q$ is the fraction of CI coefficients that are non-zero. The molecular orbital basis is the set of ground-state Hartree-Fock orbitals.

| $\epsilon$ | $\Delta E$ | $q$ |
|---|---|---|
| 0.01000000 | 0.00779437 | 0.00016 |
| 0.00300000 | 0.00271044 | 0.00096 |
| 0.00100000 | 0.00093248 | 0.00398 |
| 0.00030000 | 0.00028322 | 0.01118 |
| 0.00010000 | 0.00009827 | 0.01848 |
| 0.00003000 | 0.00002924 | 0.04666 |
| 0.00001000 | 0.00001027 | 0.07688 |
| 0.00000300 | 0.00000288 | 0.16263 |
| 0.00000100 | 0.00000103 | 0.24103 |
| 0.00000030 | 0.00000030 | 0.33283 |
| 0.00000010 | 0.00000011 | 0.43326 |
| 0.00000003 | 0.00000003 | 0.53159 |
| 0.00000001 | 0.00000001 | 0.62975 |

Table 4. Truncation of FCI wavefunction for OH, cc-pVTZ basis set (O $f$ and H $d$ functions omitted), as a function of compressive sampling power $k$, with $\epsilon = 3 \times 10^{-5}$. $\Delta E$ is the error in the energy (Hartree), and $q$ is the fraction of CI coefficients that are non-zero. The molecular orbital basis is the set of ground-state Hartree-Fock orbitals.

| $k$ | $\Delta E$ | $q$ |
|---|---|---|
| 0.50000 | 0.00008773 | 0.05071 |
| 1.00001 | 0.00005247 | 0.05019 |
| 1.50000 | 0.00003360 | 0.04707 |
| 2.00000 | 0.00002924 | 0.04666 |
| 2.50000 | 0.00003213 | 0.04674 |
| 3.00000 | 0.00003232 | 0.04674 |
| 3.50000 | 0.00003236 | 0.04674 |
| 4.00000 | 0.00003237 | 0.04674 |

separations $0.8, 0.85, \ldots, 1.7\,\mathring{A}$ in the STO-3G basis set[25]. This is a tractable prototype for a system with strong static correlation and interactions that decay over distance. The results are shown in tables 5 and 6, and support the same conclusions as the previous examples. Except at aggressively large $\epsilon$ thresholds, only modest levels of sparsity are achieved in the eigenvectors.

## 4. Conclusion

The examples presented show that it is possible to use perturbation theory within the Davidson diagonalisation algorithm to mask off small coefficients in the wavefunction against a predefined target energy threshold. The resulting truncation errors in the energy are reliably close to the threshold. For a typical threshold, however, the amount of induced sparsity is relatively modest.

We have also seen how to attempt to introduce sparsity by penalty-function optimisation, with a norm-based penalty function that decreases with increasing sparsity. Whilst this is an appealing and promising approach, it appears to be ineffective in promoting wavefunction sparsity.

Table 5. Truncation of FCI wavefunction for $H_{10}$, STO-3G basis set, as a function of energy truncation threshold $\epsilon$, with no compressive sampling penalty. $\Delta E$ is the error in the energy (Hartree), and $q$ is the fraction of CI coefficients that are non-zero. The molecular orbital basis is the set of ground-state Hartree-Fock orbitals.

| $\epsilon$ | $\Delta E$ | $q$ |
|---|---|---|
| 0.01000000 | 0.01308947 | 0.00333 |
| 0.00300000 | 0.00422007 | 0.01548 |
| 0.00100000 | 0.00140513 | 0.05193 |
| 0.00030000 | 0.00057381 | 0.15079 |
| 0.00010000 | 0.00016136 | 0.25947 |
| 0.00003000 | 0.00001806 | 0.47979 |
| 0.00001000 | 0.00001272 | 0.64417 |
| 0.00000300 | 0.00000368 | 0.77700 |
| 0.00000100 | 0.00000138 | 0.87476 |
| 0.00000030 | 0.00000035 | 0.92457 |
| 0.00000010 | 0.00000012 | 0.96294 |
| 0.00000003 | 0.00000003 | 0.98559 |

Table 6. Truncation of FCI wavefunction for $H_{10}$, STO-3G basis set, as a function of compressive sampling power $k$, with $\epsilon = 3 \times 10^{-5}$. $\Delta E$ is the error in the energy (Hartree), and $q$ is the fraction of CI coefficients that are non-zero. The molecular orbital basis is the set of ground-state Hartree-Fock orbitals.

| $k$ | $\Delta E$ | $q$ |
|---|---|---|
| 0.50000 | 0.00004293 | 0.60459 |
| 1.00001 | 0.00010739 | 0.56975 |
| 1.50000 | 0.00004885 | 0.48768 |
| 2.00000 | 0.00001806 | 0.47979 |
| 2.50000 | 0.00003902 | 0.38925 |
| 3.00000 | 0.00003866 | 0.38879 |
| 3.50000 | 0.00003855 | 0.38859 |
| 4.00000 | 0.00003851 | 0.38851 |

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