

Moments-based improved quantum computation of the electric dipole moment of molecular systems

Michael A. Jones^{1,2,*} Harish J. Vallury^{1,3} Manolo C. Per^{2,4} Harry M. Quiney¹
and Lloyd C. L. Hollenberg^{1,2}

¹*School of Physics, University of Melbourne, Parkville 3010, Australia*

²*ARC Centre of Excellence in Quantum Biotechnology, University of Melbourne, Parkville 3010, Australia*

³*CSIRO Data61, Marsfield 2122, Australia*

⁴*CSIRO Data61, Clayton 3168, Australia*

 (Received 16 October 2025; revised 15 February 2026; accepted 19 March 2026; published 1 May 2026)

With rapid progress being made in the development of platforms for quantum computation, there has been considerable interest in whether present-day and near-term devices can be used to solve problems of relevance. A commonly cited application area is the domain of quantum chemistry. While most experimental demonstrations of quantum chemical calculations on quantum devices have focused on the ground-state electronic energy of the system, other properties of the ground state, such as the electric dipole moment, are also of interest. Here, we employ the quantum-computed-moments method, based on the Lanczos cluster expansion, to estimate the dipole moment of the water molecule on an IBM Quantum superconducting quantum device. The noise-mitigated results agree with full-configuration-interaction calculations to within 0.03 ± 0.007 D ($2\% \pm 0.5\%$), compared with direct expectation-value determination (i.e., variational quantum eigensolver, VQE) with errors of the order of 0.07 D (5%), even when the VQE calculation is performed without noise. This demonstrates that moments-based energy-estimation techniques can be adapted to noise-robust evaluation of nonenergetic ground-state properties of chemical systems. We emphasize that the dipole moment is chosen as an example of such a property, but the method applies to any property determined by one- or two-body interactions.

DOI: [10.1103/y4nq-kcw1](https://doi.org/10.1103/y4nq-kcw1)

I. INTRODUCTION

The simulation of quantum chemical systems seems a natural application of interest for quantum computers; however, due to strict accuracy requirements and the need for deep circuits, practical demonstrations of quantum-computed chemistry algorithms have been limited to proof-of-principle experiments, usually employing the variational quantum eigensolver (VQE) [1] to estimate the ground-state energy of small systems. More recently, progress has been made on the implementation of more sophisticated ground-state energy-estimation methods in chemistry, such as those based on variations of the quantum phase-estimation algorithm [2], imaginary-time evolution [3,4], Hamiltonian moments [5–9], subspace

expansion [10–12], and selected configuration interaction (also known as sample-based quantum diagonalization) [13–18]. Additionally, other properties of interest have been computed, such as excited states and band-structure calculations [10,12,16,19–27], vibrational modes [28], and dipole moments [29–31]. Dipole moments and similar properties that do not depend on excited electronic states or motion of the atomic nuclei seem a straightforward extension to the ground-state energy-estimation problem. To date, however, quantum hardware computation of these properties has been primarily restricted to direct expectation-value determination rather than taking advantage of techniques to improve energy estimation such as those listed above. Nonetheless, we note that the classical description obtained through sample-based methods makes this extension straightforward, and in recent work, Oumarou *et al.* [32] proposed methods to estimate such properties through Krylov subspace diagonalization. Additionally, while specific methods have been proposed for the estimation of molecular-response properties [33–36]—i.e., the response of properties such as the dipole moment to external fields due to excitation to higher energy states—including quantum hardware computations

*Contact author: michjones@unimelb.edu.au

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

[34,36], our interest is in evaluating nonenergetic properties of the ground state in the absence of external potentials and in improving the accuracy of results when access to the exact ground state is infeasible.

Here, we show improvement over the direct estimation of ground-state observables, using the electric dipole moment of the water molecule as an example, through use of the Hamiltonian moments and the quantum computed moments (QCM) method [37,38], which does not require any additional quantum circuit depth. Based on the Lanczos-cluster-expansion framework [39–41], the noise-robust, moments-corrected estimate uses a Hellmann-Feynman approach [42–45] and was previously applied to Heisenberg models in simulation [45]. Here, we extend this by demonstrating implementation of the method using results from an IBM superconducting quantum device [46]. This provides verification that the moments-based QCM method produces superior estimates for such quantities compared with direct evaluation when applied to chemical systems. In addition, we consider a number of ways in which the Hellmann-Feynman approach can be combined with error-mitigation techniques and investigate their behavior in Appendix A.

Moments-based approaches are closely related to quantum Krylov methods [18,47–49], with moments-based techniques corresponding to a specific choice of the Krylov subspace. In particular, the Lanczos cluster expansion [39–41] correction used here essentially results in an analytic diagonalization of the subspace including approximate contributions from the remainder of the Hilbert space; however, our technique is quite different from that

proposed by Oumarou *et al.* [32]. Where we accept a higher measurement cost to minimize the quantum circuit depth, Oumarou *et al.* [32] use quantum signal-processing techniques to reduce the measurement cost at the expense of deeper circuits.

II. METHOD

A. Quantum computed moments for ground-state energy estimation

The trial-state preparation and measurement follow the methods outlined in previous work [8] and are summarized here for completeness. The water molecule (H_2O) at equilibrium geometry (HOH bond angle, 104.5° ; symmetric OH bond length, $0.96 \text{ \AA}$) is represented in the STO-3G basis by the second-quantized Hamiltonian

$$\mathcal{H} = \sum_{jk} h_{jk} a_j^\dagger a_k + \sum_{jklm} g_{jklm} a_j^\dagger a_k^\dagger a_l a_m, \quad (1)$$

where $N_s = 14$ is the number of spin orbitals, $a_j^\dagger$ (a_j) are the electronic creation (annihilation) operators and h_{jk} (g_{jklm}) are the classically computed one-body (two-body) integrals. The spatial orbitals are shown in Fig. 1(a), each corresponding to a pair of spin orbitals that account for the electronic-spin degree of freedom. Subsequently, the core ($1s$) orbitals of the oxygen atom are frozen, as they do not participate in the bonding. The resulting Hamiltonian also follows the form of Eq. (1) with $N_s = 12$ spin orbitals, but with modified values for h_{jk} and g_{jklm} , and an additional

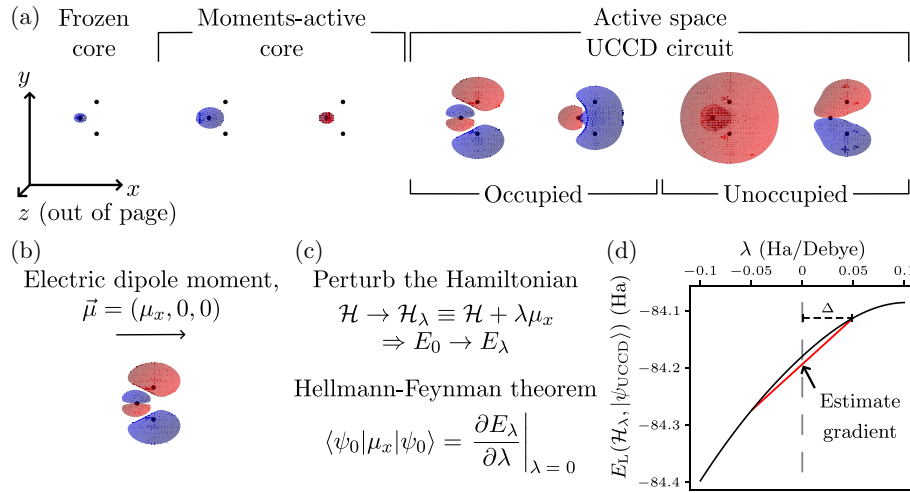


FIG. 1. Details of the application of the method to the electric dipole moment of the water molecule. (a) The STO-3G molecular orbitals used to represent the water molecule. (b) Due to symmetry considerations, the electric dipole moment must be aligned with the x axis. We use the convention that the dipole moment vector points from negative to positive charge. (c) By perturbing the Hamiltonian with the dipole operator, the Hellmann-Feynman theorem can be applied to compute the dipole moment. (d) Here, E_λ is approximated by $E_L(\mathcal{H}_\lambda, |\psi_{\text{UCCD}}\rangle)$, and the finite-difference method is employed to estimate the derivative. Note that the trial state $|\psi_{\text{trial}}\rangle = |\psi_{\text{UCCD}}\rangle$ is independent of λ , allowing for reuse of the same quantum measurements and thereby reducing the effects of stochastic error. The parameter Δ is the step size used for the finite-difference method.

constant term. The Hamiltonian moment operators, $\mathcal{H}^p$ for $p \in \{1, 2, 3, 4\}$, are determined using Wick's theorem to simplify products of creation and annihilation operators, bringing the creation and annihilation operators into normal order. Each of these moment operators is then reduced to an eight-spin-orbital operator by freezing two additional orbitals (predominantly the oxygen $2s$ and $2p_z$ orbitals, where z is the axis perpendicular to the plane of the molecule). As a result, these orbitals are not directly simulated by the quantum device but are incorporated by the moments-based correction. The remaining eight spin orbitals are each mapped to a qubit via the Jordan-Wigner encoding [50] and a selected unitary coupled cluster doubles (UCCD, [51]) trial state, $|\psi_{\text{UCCD}}\rangle$, is constructed as a variational wave function that is measured in 200 different bases (25 000 measurements each) using readout-error mitigation [52] (performed by preparing and measuring known computational basis states to calibrate the measurement statistics) and symmetry verification [53] (discarding results that do not agree with the known total spin and spin-projection values) to construct the four-body reduced density matrix (RDM). Since there are only four electrons remaining in the active space, the 4-RDM is sufficient to evaluate all relevant operators. The RDM is then rescaled [54] to enforce the correct trace. The circuit used to prepare the trial state has eight qubits, 22 two-qubit gates and a depth of 25 gates. Each measurement basis requires a different configuration of additional one- and two-qubit gates. Further details are given in Appendix B.

In our previous work [8], the Hamiltonian moments for the water molecule were evaluated from the 4-RDM for both the trial state and the Hartree-Fock reference state, $|\psi_{\text{HF}}\rangle$, with the latter used to mitigate noise in the former by assuming a depolarizing noise model [55],

$$\text{Tr}(\rho^{\text{noisy}} \mathcal{H}^p) = (1 - q_p) \text{Tr}(\rho^{\text{exact}} \mathcal{H}^p) - q_p \text{Tr}(\rho^{\text{mixed}} \mathcal{H}^p). \quad (2)$$

Using the reference state, which has an efficiently computed classical description, $\rho_{\text{HF}}^{\text{exact}}$, the effective noise level, q_p , of the circuit can be found for each moment, and Eq. (2) can be inverted to correct the noisy trial-state result [56,57].

The moments are then combined in an analytic formula derived from the cluster expansion of the Lanczos recursion [39], which sums all Lanczos coefficients (truncated to fourth order in the moments) to obtain a corrected ground-state energy estimate, E_L [40,41],

$$E_L \equiv c_1 - \frac{c_2^2}{c_3^2 - c_2 c_4} \left(\sqrt{3c_3^2 - 2c_2 c_4} - c_3 \right), \quad (3)$$

$$c_p = \langle \mathcal{H}^p \rangle - \sum_{j=0}^{p-2} \binom{p-1}{j} c_{j+1} \langle \mathcal{H}^{p-1-j} \rangle.$$

We note that quantum computation of Hamiltonian moments has also been considered in other contexts [5], most notably in relation to the connected-moments expansion [7,58–62]; however, the combination of error-mitigation and moments-based correction described here was seen to work well, giving ground-state energy estimates within millihartree accuracy of the full-configuration interaction (FCI) result [8]. Here, we aim to reproduce this accuracy for nonenergy observables (namely the electric dipole moment).

B. Quantum computed moments for arbitrary ground-state observable estimation

To estimate ground-state observables via the Hellmann-Feynman theorem in the context of moments-based methods [42,43,45], consider the modified Hamiltonian

$$\mathcal{H}_\lambda \equiv \mathcal{H} + \lambda \mu, \quad (4)$$

where λ is a real scalar and μ is the observable of interest. For concreteness, let $\mu = \mu_x$ be the x component of the electric dipole moment operator [the only component with nonvanishing expectation value by symmetry; Fig. 1(b)],

$$\mu = \sum_{jk}^{N_s} f_{jk} a_j^\dagger a_k, \quad (5)$$

where f_{jk} are classically computed integrals. Let $|\Phi_\lambda\rangle$ be the ground state of $\mathcal{H}_\lambda$ with energy E_λ . Then, the Hellmann-Feynman theorem states

$$\frac{\partial E_\lambda}{\partial \lambda} = \langle \Phi_\lambda | \mu | \Phi_\lambda \rangle, \quad (6)$$

and evaluating at $\lambda = 0$ gives expectation values with respect to the ground state of the unmodified Hamiltonian

$$\left. \frac{\partial E_\lambda}{\partial \lambda} \right|_{\lambda=0} = \langle \Phi_0 | \mu | \Phi_0 \rangle \equiv \mu_0. \quad (7)$$

While the Hellmann-Feynman theorem also holds for states other than energy eigenstates, subject to conditions on the energy derivative with respect to trial-state parameters, here, the formulation in terms of the ground state is sufficient, as we aim to estimate nonenergetic ground-state properties [right-hand side of Eq. (7)] from approximations to the ground-state energy [left-hand side of Eq. (7)]. The applicability of various forms of the Hellmann-Feynman theorem is a much-studied topic [63,64].

To approximate the derivative, we can employ the finite-difference method [Fig. 1(d)], resulting in the approximation

$$\mu_0 \approx \frac{1}{2\Delta} (E_{\Delta} - E_{-\Delta}). \quad (8)$$

Further, using the moments-based correction to improve the evaluation of $E_{\pm\Delta}$, and therefore the estimation of the dipole moment relative to the standard variational estimate, gives

$$\mu_0 \approx \frac{1}{2\Delta} [E_L(\lambda = \Delta) - E_L(\lambda = -\Delta)]. \quad (9)$$

Note that the notation used here is that λ is a variable (with respect to which we would like to take the derivative) while $\pm\Delta$ is the value assigned to λ to compute the finite-difference approximation to the derivative.

When performing finite-difference computations on a quantum device, often, the accuracy of the difference formula is severely limited by stochastic (sampling) noise; however, in this case, the parameter with respect to which the derivative is taken, λ , appears in postprocessing rather than in the quantum computation, which allows reuse of the quantum output for both $\lambda = \pm\Delta$ values and vastly reduces the accumulation of stochastic noise. In the absence of the moments-based correction, reusing the trial state in this way effectively assumes that the ground state is independent of the change in the Hamiltonian, leading to systematic errors due to the neglect of the wave-function response terms. If the response terms are ignored completely and expectation values are taken at $\lambda = \pm\Delta$, then the estimate of the dipole moment is simply the expectation value of the dipole operator with respect to the trial state,

$$\begin{aligned} & \langle \psi_{\text{trial}} | \mu | \psi_{\text{trial}} \rangle \\ &= \frac{\langle \psi_{\text{trial}} | \mathcal{H}_{\Delta} | \psi_{\text{trial}} \rangle - \langle \psi_{\text{trial}} | \mathcal{H}_{-\Delta} | \psi_{\text{trial}} \rangle}{2\Delta}. \end{aligned} \quad (10)$$

Although Eq. (10) holds exactly (since the modified Hamiltonian is linear in λ , there is no error due to the finite-difference method), it is not practically useful, as the costs of evaluating the left- and right-hand sides are roughly equal. Additionally, the dipole-moment estimate obtained is the expectation value with respect to the trial state, rather than the ground state, so systematically approaching the ground-state expectation value requires improving the trial state, leading to additional quantum gates and therefore increased noise.

The Hellmann-Feynman theorem may be of practical use in cases where we have access to approximate ground-state energies but not to the corresponding state. This is the case for a number of ground-state energy-estimation

techniques such as the moments-based correction [42]. In this case, the estimates are able to incorporate additional information from the Hamiltonian to circumvent the limit imposed by the trial state, effectively incorporating some of the wave-function response and improving on the dipole-moment estimate. We denote the dipole-moment estimate computed this way using the moments-based correction, E_L , as $\mu_L(\Delta, |\psi\rangle)$, so the final expression used for the dipole-moment estimation in this work is

$$\begin{aligned} \mu_0 &\approx \mu_L(\Delta, |\psi_{\text{trial}}\rangle) \\ &\equiv \frac{1}{2\Delta} [E_L(\Delta, |\psi_{\text{trial}}\rangle) - E_L(-\Delta, |\psi_{\text{trial}}\rangle)]. \end{aligned} \quad (11)$$

Figure 2 outlines three methods for calculating the reference-state calibrated moments from the reduced density matrix. The first of these (method A) we term *de novo* moments, as each time Δ is adjusted (e.g., to test for convergence to the $\Delta \rightarrow 0$ limit), the classical computation of the moments must begin anew, though the quantum computation need not be repeated. To minimize the required classical computation, we can instead express $\mathcal{H}_{\lambda}^p$ analytically as a polynomial in λ and compute the operator-valued coefficients (methods B and C in Fig. 2). This allows evaluation of the moments-corrected ground-state energy estimate for many values of Δ with minimal additional processing; however, as indicated in Fig. 2, we apply the reference-state calibration to the modified moments, $\langle \mathcal{H}_{\lambda}^p \rangle$. We term this method “postmitigated moments,” as the reference-state error mitigation is performed after construction of the modified moments (method B). In contrast to applying the reference-state calibration to the coefficients, $\langle \mathcal{H} \rangle$, $\langle \mu \rangle$ etc. (method C), we find this to be more stable to noise. We refer to method C as the “premitigated moments” method because reference-state error mitigation is performed before construction of the modified moments. Further investigation of the adverse error propagation is given in Appendix A. Based on the noise analysis, Appendix A also introduces methods D and E, which we refer to as “truncated postmitigated moments” and “truncated premitigated moments,” respectively, and which correspond to discarding higher-order dependencies on Δ .

We note that there are many improvements and adaptations that can be made to VQE to improve its accuracy and/or reduce the quantum computational cost, and we expect many of these to be compatible with the moments-based methods for energy and nonenergetic property estimation. Such techniques could include explicit correlation methods [65–70], Hamiltonian factorization and downfolding schemes [71–80], specific basis choices or the use of basis-set-free methods [81–83], and/or perturbative techniques [84,85].

We also note that the major computational bottlenecks of the method presented here are the required number of

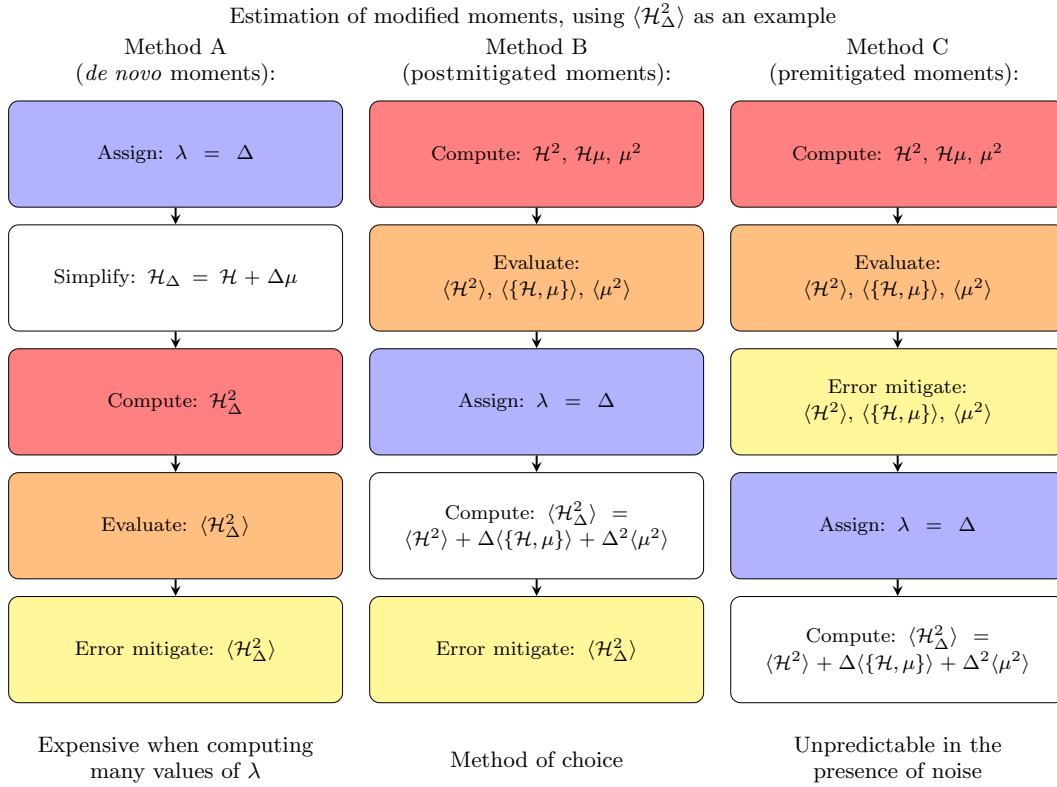


FIG. 2. Three methods for evaluating the second moment, $\langle \mathcal{H}_\Delta^2 \rangle$. The computational bottleneck is the implementation of Wick's theorem to multiply the fermionic operators (red boxes). Performing these steps in a way that is independent of Δ (methods B and C) allows for much more efficient evaluation of $\langle \mathcal{H}_\Delta^2 \rangle$ at many values of λ because steps prior to assigning a value of λ (above the blue boxes) do not need to be repeated; however, for certain values of λ , method C was seen to be unstable to errors from the quantum computation (see Appendix A). Note that $\langle \{\mathcal{H}, \mu\} \rangle \equiv \langle \mathcal{H}\mu + \mu\mathcal{H} \rangle = 2\text{Re}\langle \mathcal{H}\mu \rangle$. The methods may be generalized to evaluate $\langle \mathcal{H}_\Delta^p \rangle$.

quantum measurements and the necessary classical pre-computation. In computing the p th-order moment operator $\mathcal{H}^p$, there are N_s^{4p} products of excitations that need to be considered for N_s spin orbitals, given that there are N_s spin orbitals and therefore N_s^4 terms in the electronic Hamiltonian. While this is clearly challenging for large systems, it can be circumvented by methods that encode the Hamiltonian directly into the quantum circuit [86–89], as opposed to the term-by-term evaluation used here, to allow implementation on current noisy hardware. These circuit-based methods may also be able to reduce the shot requirement; however, they require circuit depths that are beyond current quantum hardware and so are better suited to the fault-tolerant or early-fault-tolerant regimes. Additionally, the Hamiltonian cannot be varied in postprocessing, as it is encoded directly into the quantum circuit; this would likely result in a loss of robustness in the finite-differencing step used to estimate nonenergetic properties, as the stochastic (sampling) errors on each side of the finite difference would be independent. Whether there exists a “best of both” solution that reduces the preprocessing and the number of shots while avoiding amplification of stochastic noise in the finite difference step remains an open question.

III. RESULTS AND DISCUSSION

The results of the dipole-moment calculation are shown in Fig. 3. With the moments-based correction, the UCCD trial-state results from *ibm_torino* (blue dash-dotted line) reproduce statevector simulation results (green dash-dotted line) to within 1 standard deviation (shaded area, determined from 100 bootstrapping samples) for small values of Δ , i.e., in the region of interest. On the other hand, direct evaluation of the expectation value (blue dotted line) fails to reproduce the statevector simulation results (green dotted line) [90]. In addition to the moments-corrected μ_L being closer to the target FCI value, its variance is also notably smaller than that of the expectation value, $\langle \mu \rangle$, demonstrating that the observed error-robustness property [38] of the moments-based energy correction transfers to the evaluation of nonenergetic properties.

We note that the expectation values, $\langle \mu \rangle$, appear to be completely independent of Δ , as expected, since the finite-difference error vanishes when the function is linear as in Eq. (10). In truth, there is a very slight Δ dependence in the error-mitigated (blue dotted) results, since the postmitigated moments method assigns $\lambda = \Delta$ prior to reference-state calibration, leading to an approximation

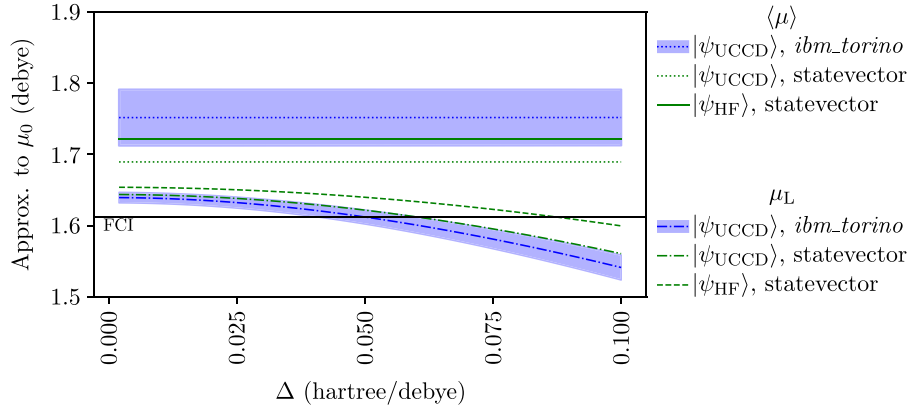


FIG. 3. Results of evaluating the dipole moment using statevector simulation (green) and quantum hardware (blue) compared with the FCI result (black). The dash-dotted and dotted lines indicate the results from the UCCD state with and without the moments-based correction, respectively. The shaded regions represent 1 standard deviation (determined by 100 bootstrapping samples). Note that as Δ varies, the approximation is calculated from the same data from the quantum device (or simulator), postprocessed using different values of Δ . Dashed and solid lines show the results for the Hartree-Fock reference state with and without the moments correction, respectively. Since the Hartree-Fock state is used to correct the quantum hardware results, the hardware results for the Hartree-Fock state exactly match the statevector simulation and are not shown. The finite-difference error vanishes in the limit $\Delta \rightarrow 0$; the results for larger Δ values are plotted to demonstrate the convergence of μ_L .

to the truly linear behavior of the statevector simulation; however, the difference between $\Delta = 0$ and $\Delta \rightarrow \infty$ is only 3.7 mD. On the other hand, $E_L(\mathcal{H}_\lambda, |\psi\rangle)$ is highly nonlinear, leading to a Δ dependence in its approximate derivative μ_L .

While the percentage error in μ_L (1.7%) is smaller than that of $\langle\mu\rangle$ (8.7%), these are both much larger than the percentage error in the energy estimation (0.05% and 1.2% for E_L and $\langle\mathcal{H}\rangle$, respectively). This is to be expected because for small rotations, $d\theta$, away from the true ground state, the changes in the observables are

$$\begin{aligned} d\langle\mathcal{H}\rangle_\theta &= \mathcal{O}(d\theta^2), \\ d\langle\mu\rangle_\theta &= \mathcal{O}(d\theta), \end{aligned} \quad (12)$$

where the $\mathcal{O}(d\theta)$ term in the energy change vanishes since the trial state is near a minimum. The same arguments hold for the changes in the moments $d\langle\mathcal{H}^p\rangle = \mathcal{O}(d\theta^2)$ and modified moments $d\langle\mathcal{H}_\lambda^p\rangle = \mathcal{O}(d\theta)$.

It is also possible that the reason the percentage error in the dipole operator is larger than that of the energy is that, despite having fewer terms in Eq. (5) compared with Eq. (1), the Hamiltonian is dominated by diagonal terms, in particular the one-body terms, g_{jj} ; in contrast, for the dipole, there is also a significant contribution from the off-diagonal terms, f_{jk} . Since the off-diagonal elements of the reference-state RDM and the mixed-state RDM are the same,

$$\text{Tr}(\rho_{\text{HF}}^{\text{exact}} a_j^\dagger a_k) = \text{Tr}(\rho^{\text{mixed}} a_j^\dagger a_k) = 0, \quad j \neq k, \quad (13)$$

they cannot be corrected directly using the reference-state error mitigation because Eq. (2) does not have a unique

solution for q . For this reason, the noise calibration is applied to $\langle\mathcal{H}_\Delta^p\rangle$ instead of to the individual RDM elements, which allows errors in the off-diagonal elements to be corrected indirectly, essentially by assuming that the correct q value is related to the error level in the diagonal elements; however, a more effective method to correct these off-diagonal elements might be to use reference states that have nonzero off-diagonal terms.

IV. CONCLUSION

While ground-state energies are important in the application of quantum algorithms to quantum chemistry, it is also important to be able to evaluate other chemical properties in an accurate, noise-robust way. In contrast to previous results that evaluate expectation values, we have demonstrated that moments-based ground-state energy corrections can be adapted to evaluate such properties (using the electric dipole moment of the water molecule as an example) using the Hellmann-Feynman theorem, and that such a method transfers the inherent accuracy and noise-robustness of the corrected energy value to the quantity of interest. These observations broaden the potential range of applications in chemistry to which quantum devices could provide a potential advantage over classical computation.

ACKNOWLEDGMENTS

The research was supported by the University of Melbourne through the establishment of the IBM Quantum Network Hub at the University. The research was also supported by the Australian Research Council Centre of

Excellence in Quantum Biotechnology through Project No. CE230100021. M.A.J. and H.J.V. were supported by the Australian Commonwealth Government through Research Training Program Scholarships. This work was further supported by resources provided by the Pawsey Supercomputing Research Centre with funding from the Australian Government and the Government of Western Australia, and by The University of Melbourne's Research Computing Services and the Petascale Campus Initiative.

M.A.J. and L.C.L.H. conceived the project. M.A.J. set up the computational framework and performed the calculations and data analysis, with input from all authors.

DATA AVAILABILITY

The data that support the findings of this article are openly available [91].

APPENDIX A: ESTIMATING MODIFIED MOMENTS IN THE PRESENCE OF NOISE

1. Noise analysis of the premitigated-moments method

In Fig. 2, we asserted that the premitigated-moments method (method C) was unsuitable due to its unpredictable behavior in mitigating quantum noise. Here, we will investigate this behavior in more detail. To do this, we will begin at the point where the pre- and postmitigated methods diverge, that is, with the error mitigation of the coefficients, $\langle \mathcal{H} \rangle$, $\langle \mu \rangle$, $\langle \mathcal{H}\mu \rangle$, etc. Figure 4 shows the application of the reference-state error-mitigation technique to these

coefficients. Notably, the coefficients that are quadratic in the dipole operator, μ , are mitigated poorly. In fact, for quadratic and quartic operators, the noisy measurements drift *away* from the maximally mixed state compared with their noiseless values. This suggests that the white-noise model used in the mitigation does not accurately characterize the behavior of the device, though it works reasonably well for the unmodified moments. We note that the quartic term is still mitigated reasonably well since the Hartree-Fock (HF) and UCCD expectation values are close to each other, relative to the operator norm.

While the fractional errors in Fig. 4 appear to depend only on the order in μ , there is small variation between terms that is not visible on the scale presented. The final error levels for the UCCD state after mitigation are approximately $-0.1\% \pm 0.3\%$ for terms independent of μ , $-2.0\% \pm 1.5\%$ for linear terms, $20\% - 30\% \pm 4\%$ for the quadratic terms, $-1.3\% \pm 1.7\%$ for cubic terms and $2.6\% \pm 2.9\%$ for the quartic term. The error in the final moments-corrected dipole-moment estimate (where the statevector simulation and mixed state are defined as 0% and 100% error, respectively) is about $0.1\% - 0.3\% \pm 0.2 - 0.3\%$, depending on the method.

Noting that the error mitigation works well for terms that are constant or linear in μ , and that the dependency on Δ and μ matches, the dipole-moment estimation should be reasonable in the region $\Delta^2 \ll \Delta$. This is observed in Fig. 5, which presents the same results as Fig. 3 in addition to the equivalent results from method C and another two methods (D and E) outlined below.

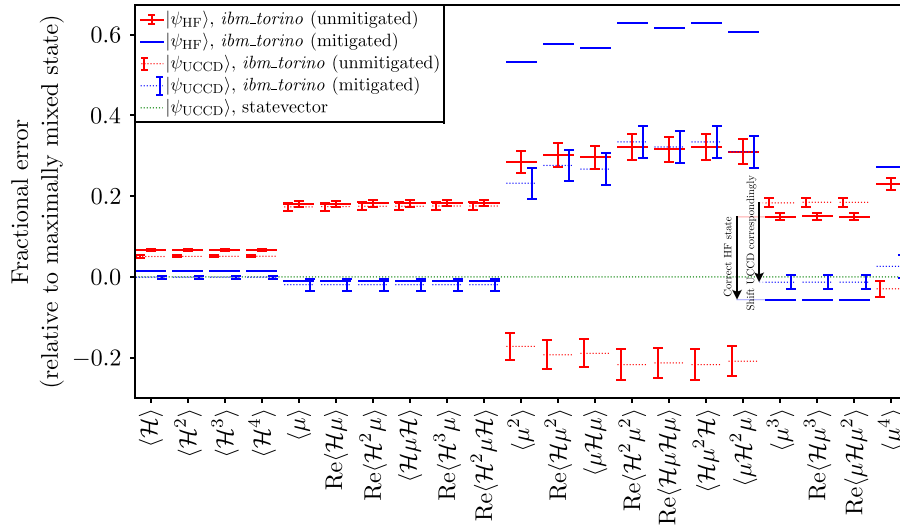


FIG. 4. Effectiveness of reference-state error mitigation for the premitigated-moments method. The known correction required for the Hartree-Fock state (solid lines) is used to correct the UCCD-state results (dotted lines), in both cases, the red and blue lines indicate unmitigated and mitigated results, respectively. There is no uncertainty in the mitigated Hartree-Fock-state values, as these are the classically efficient baseline used for the calibration. The errors have been normalized so that the maximally mixed state gives an error of 1. While most of the coefficients have errors of the order of a few percent, the terms quadratic in μ all have errors of at least 20% due to the breakdown of the noise model. Note that here, “unmitigated” means no reference-state error mitigation; other error-mitigation techniques have been applied.

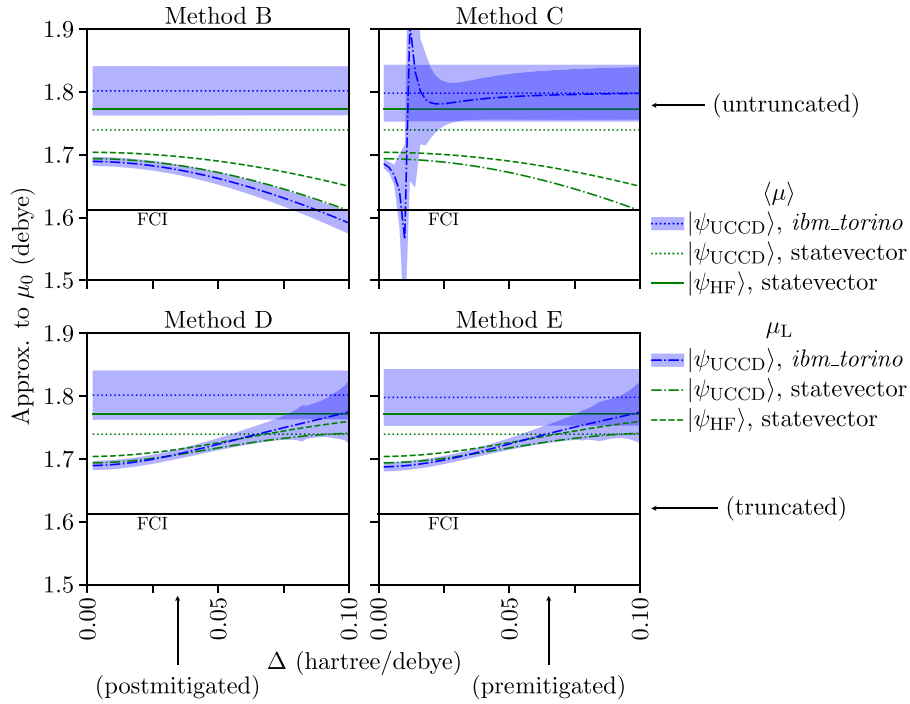


FIG. 5. Final estimates of μ_0 using methods B–E. In each case, the moments-based estimate (blue dash-dotted line) converges (in the $\Delta \rightarrow 0$) limit to the statevector result (green dash-dotted line). The blue and green dotted lines indicate the application of the Hellmann-Feynman theorem without the moments correction for mitigated data and statevector simulation, respectively. Results from the Hartree-Fock state are shown in dashed (solid) green with (without) the moments correction. Note that dotted lines indicate UCCD results and dashes represent the moments correction, so dash-dotted lines correspond to both. The colors indicate the backend (hardware or simulator). The left (right) column plots correspond to postmitigated (premitigated) methods and the top (bottom) row to untruncated (truncated) methods.

In method B, the error mitigation is not applied directly to $\langle \mu^2 \rangle$ and is instead applied to $\langle \mathcal{H}_\Delta^2 \rangle = \langle \mathcal{H}^2 + \Delta \{ \mathcal{H}, \mu \} + \Delta^2 \mu^2 \rangle$, where the correction is dominated by the lower-order terms in Δ , so the effects of the noise model failing are less pronounced. If we consider the action of the reference-state error mitigation to be some nonlinear function f , then the different results between methods B and C are simply a manifestation of this nonlinearity:

$$f(\langle \mathcal{H} \rangle + \Delta \langle \mu \rangle) \neq f(\langle \mathcal{H} \rangle) + \Delta f(\langle \mu \rangle). \quad (\text{A1})$$

It is also curious that in the large- Δ limit, the estimate from premitigation converges to $\langle \mu \rangle$. Examining this, we find that the component of Eq. (3) inside the square root becomes negative [92] for $|\Delta| \gtrsim 0.01$. Since the imaginary part is discarded, the resulting correction term will have the form $-c_3/(c_3^2 - c_2 c_4)$. In this case, we also see that due to noise, c_4 is an order of magnitude larger than c_2 or c_3 for $|\Delta| \gtrsim 0.04$, causing the correction term to vanish and the estimates to converge to the naive expectation-value estimate. Additionally, discarding the imaginary component results in a discontinuity in the gradient of E_L . This forms a pair of sharp peaks in Fig. 6, and using points

near these in the finite-difference approximation leads to the spike observed in Fig. 5.

2. Truncation of higher-order terms

Since the error is predominantly introduced via the quadratic and, to a lesser extent, higher-order terms, we could also consider simply discarding these terms. Because the region of interest is the limit as $\Delta \rightarrow 0$, this should not affect the final estimate too much. Running two methods, methods D and E, corresponding to truncation of methods B and C, respectively, we find that these converge to within 0.1 mD of the untruncated versions at $\Delta = 0.02$ and expect that the remaining discrepancy will vanish as Δ approaches zero. Table I summarizes the differences between methods A–E.

We note in Fig. 5 that the truncation in methods D and E affects not only the quantum hardware results but also the classical simulation values. Due to the truncation of the noisy higher-order terms, we find that method E performs much better than method C, while method D offers only a minimal reduction in variance over method B due to method B's much more stable behavior in comparison with method C. We note that if it was the linear terms that had mitigated poorly, then the truncated methods would

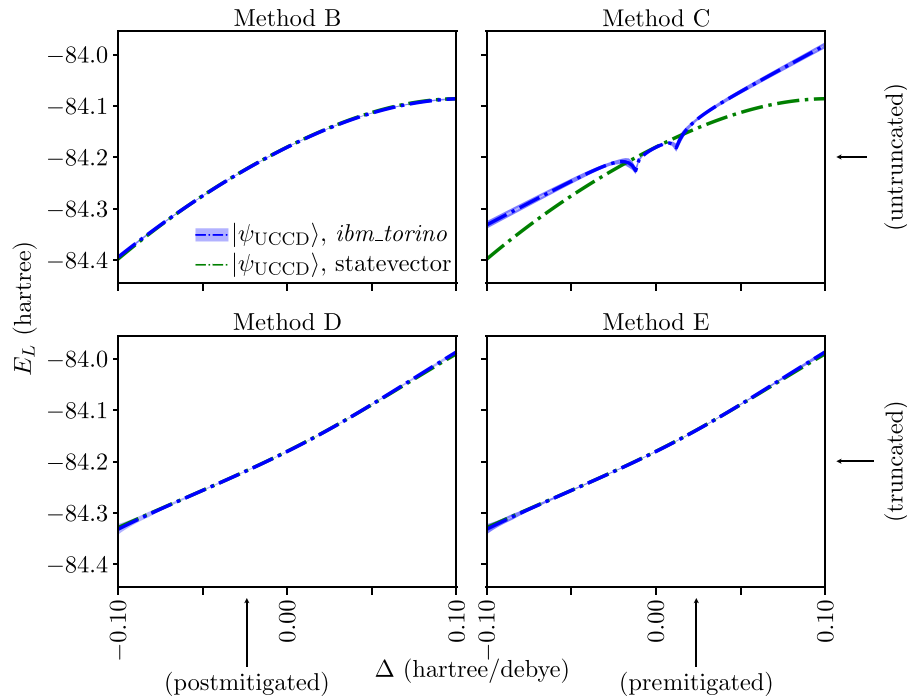


FIG. 6. Estimation of E_L as a function of Δ through methods B–E. The error-mitigated results from *ibm_torino* (blue) match the statevector results (green) closely for methods B, D, and E, but method C is too sensitive to noise in the higher-order terms in Δ . The left (right) column plots correspond to postmitigated (premitigated) methods and the top (bottom) row to untruncated (truncated) methods.

not be likely to grant any improvement in noise resilience, though method B may still be able to succeed, provided that the constant terms are not too noisy.

presented in prior work [8]. To keep this work self-contained, they are summarized below.

1. Trial-state design

To limit the quantum circuit depth, the trial circuit involves only the four double-excitation operators with the largest contribution to the true ground state. The qubit-to-spin-orbital ordering is selected in such a way that the number of operations required to implement these excitations is minimized and single excitations are excluded

APPENDIX B: QUANTUM COMPUTATIONAL DETAILS

The quantum computational details—i.e., the trial-state design, the measurement scheme, and the method for extracting operators from reduced density matrices—were

TABLE I. Brief summary and comparison of moments-corrected dipole-moment estimation methods through the Hellmann-Feynman theorem. The key difference between methods A–C is at what stage the finite difference step-size Δ is converted from a symbolic parameter to a numeric one. Note that the approximations introduced by methods D and E converge in the same limit in which the finite-difference approximation to the Hellmann-Feynman theorem (present in all methods) does.

Method	Description	Features
A	Δ is numeric throughout	Faster for a single choice of Δ but need to start over each time Δ is changed
B	Δ is symbolic for moment-operator computation and numeric for error calibration	Changing Δ requires the (classical) error calibration to be repeated, but not the moment-operator computation
C	Δ is symbolic throughout	Changing Δ is trivial, simple substitution into a formula; more prone to errors
D	Analogous to B, but moment operators are approximated	Slightly lower cost and very slightly lower variance for small Δ
E	Analogous to C, but moment operators are approximated	Slightly lower cost and much lower variance for small Δ (relative to C)

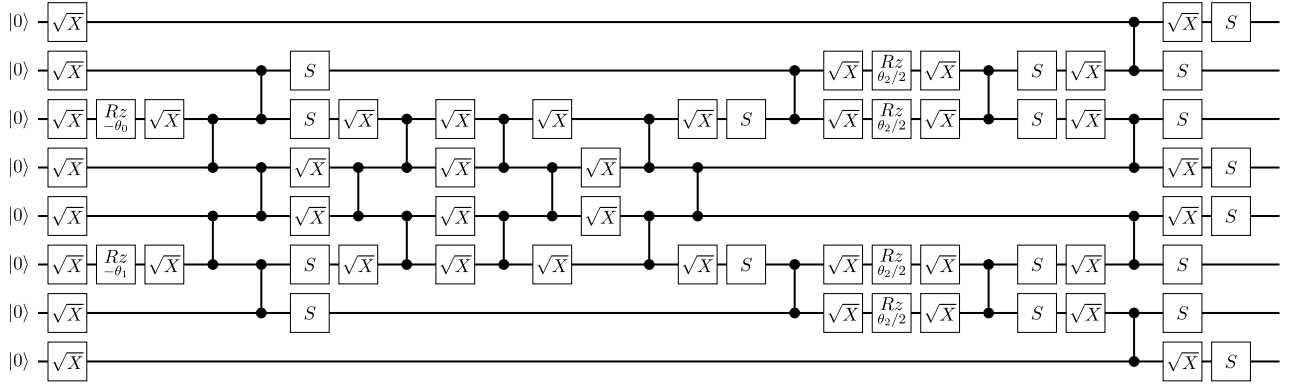


FIG. 7. Structure of the UCCD circuit used to prepare the trial state. The optimized parameters found were $(\theta_0, \theta_1, \theta_2) = (-0.051\pi, -0.029\pi, 0.032\pi)$, and the corresponding Hamiltonian is available in the supporting data [91]. The state preparation is followed by basis-rotation circuits to allow measurement of the required operators as described in Sec. B 2.

because they contribute less than the double excitations for the specific Hamiltonian in question. This results in a trial circuit with three variational parameters that are optimized using the simultaneous perturbation stochastic approximation minimizer and classical statevector simulation. The stochastic optimization was run five times and found to converge to the same minimum after about 50–60 iterations in each case. The trial-circuit structure is given in Fig. 7.

2. Measurement of reduced density matrices

Instead of converting the fermionic operators to be measured into Paulis using the Jordan-Wigner transform, the operators are decomposed into sums of products of the real and imaginary components of single-excitation operators,

e.g., for real-valued spin orbitals,

$$\begin{aligned} \langle a_j^\dagger a_k^\dagger a_l a_m \rangle &= -\langle \text{Re}(a_j^\dagger a_l) \text{Re}(a_k^\dagger a_m) \rangle \\ &\quad + \langle \text{Im}(a_j^\dagger a_l) \text{Im}(a_k^\dagger a_m) \rangle. \end{aligned} \quad (\text{B1})$$

Each real or imaginary component can then be measured by the circuits given in Fig. 8; the resulting eigenvalues for each factor in the product are multiplied and averaged over many shots. Each term in the summation is averaged separately, and the averages are summed. One advantage of this measurement method is that it can allow symmetry verification based on the known charge and spin of the molecule. Additionally, the number of qubits involved in each measurement scales with the order of the excitation instead of

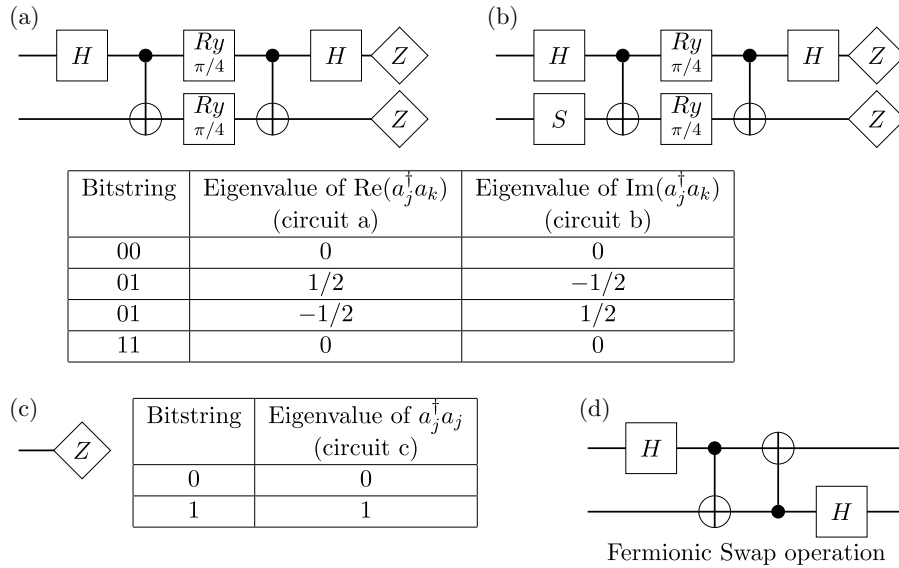


FIG. 8. Circuits used to measure the (a) real and (b) imaginary parts of a single excitation operator $a_j^\dagger a_k$ and (c) the number operator $a_j^\dagger a_j$. (d) The fermionic swap operator is used to reorder qubits when $|j - k| > 1$.

with the separation between the orbitals; i.e., it avoids the long Z -Pauli strings present in the straightforward Jordan-Wigner transform. This can potentially reduce the effect of measurement errors and allows for more trivially commuting bases that can be measured simultaneously. It is noted, however, that the fermionic swap network required to reorder the measurements introduces a linear overhead in additional circuit depth. More details can be found in Ref. [8].

3. Computation of operators from reduced density matrices

To compute the 4-RDM, all relevant (i.e., spin-preserving) four-body excitations of the form

$$R_{j_1 j_2 j_3 j_4}^{k_1 k_2 k_3 k_4} = \langle a_{j_1}^\dagger a_{j_2}^\dagger a_{j_3}^\dagger a_{j_4}^\dagger a_{k_1} a_{k_2} a_{k_3} a_{k_4} \rangle \quad (\text{B2})$$

are measured using the method described above. Lower-order excitations can then be computed from the RDMs using

$$R_{j_1 \dots j_p}^{k_1 \dots k_p} = \frac{1}{N_e - p} \sum_l R_{j_1 \dots j_p l}^{k_1 \dots k_p l}. \quad (\text{B3})$$

-
- [1] A. Peruzzo, J. McClean, P. Shadbolt, M.-H. Yung, X.-Q. Zhou, P. J. Love, A. Aspuru-Guzik, and J. L. O'Brien, A variational eigenvalue solver on a photonic quantum processor, *Nat. Commun.* **5**, 4213 (2014).
 - [2] Z. Li, X. Liu, H. Wang, S. Ashhab, J. Cui, H. Chen, X. Peng, and J. Du, Quantum simulation of resonant transitions for solving the eigenproblem of an effective water Hamiltonian, *Phys. Rev. Lett.* **122**, 090504 (2019).
 - [3] K. Yeter-Aydeniz, R. C. Pooser, and G. Siopsis, Practical quantum computation of chemical and nuclear energy levels using quantum imaginary time evolution and Lanczos algorithms, *npj Quantum Inf.* **6**, 63 (2020).
 - [4] Z. Zong, S. Huai, T. Cai, W. Jin, Z. Zhan, Z. Zhang, K. Bu, L. Sui, Y. Fei, Y. Zheng, S. Zhang, J. Wu, and Y. Yin, Determination of molecular energies via variational-based quantum imaginary time evolution in a superconducting qubit system, *Sci. China Phys. Mech. Astron.* **67**, 240311 (2024).
 - [5] P. Suchsland, F. Tacchino, M. H. Fischer, T. Neupert, P. K. Barkoutsos, and I. Tavernelli, Algorithmic error mitigation scheme for current quantum processors, *Quantum* **5**, 492 (2021).
 - [6] M. A. Jones, H. J. Vallury, C. D. Hill, and L. C. L. Hollenberg, Chemistry beyond the Hartree-Fock energy via quantum computed moments, *Sci. Rep.* **12**, 8985 (2022).
 - [7] S. Guo *et al.*, Experimental quantum computational chemistry with optimized unitary coupled cluster ansatz, *Nat. Phys.* **20**, 1240 (2024).
 - [8] M. A. Jones, H. J. Vallury, and L. C. L. Hollenberg, Ground-state-energy calculation for the water molecule on a superconducting quantum processor, *Phys. Rev. Appl.* **21**, 064017 (2024).
 - [9] M. C. Per, N. Rhodes, M. Srikumar, and J. W. Dai, Chemically-accurate prediction of the ionisation potential of helium using a quantum processor, [arXiv:2502.02023 \[quant-ph\]](#).
 - [10] J. I. Colless, V. V. Ramasesh, D. Dahlen, M. S. Blok, M. E. Kimchi-Schwartz, J. R. McClean, J. Carter, W. A. de Jong, and I. Siddiqi, Computation of molecular spectra on a quantum processor with an error-resilient algorithm, *Phys. Rev. X* **8**, 011021 (2018).
 - [11] M. Motta, W. Kirby, I. Liepuoniute, K. J. Sung, J. Cohn, A. Mezzacapo, K. Klymko, N. Nguyen, N. Yoshioka, and J. E. Rice, Subspace methods for electronic structure simulations on quantum computers, *Electron. Struct.* **6**, 013001 (2024).
 - [12] M. A. Castellanos, M. Motta, and J. E. Rice, Quantum computation of $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ excited states of aromatic heterocycles, *Mol. Phys.* **122**, e2282736 (2024).
 - [13] K. Kanno, M. Kohda, R. Imai, S. Koh, K. Mitarai, W. Mizukami, and Y. O. Nakagawa, Quantum-selected configuration interaction: classical diagonalization of hamiltonians in subspaces selected by quantum computers, [arXiv:2302.11320 \[quant-ph\]](#).
 - [14] J. Robledo-Moreno, M. Motta, H. Haas, A. Javadi-Abhari, P. Jurcevic, W. Kirby, S. Martiel, K. Sharma, S. Sharma, T. Shirakawa, I. Sitdikov, R.-Y. Sun, K. J. Sung, M. Takita, M. C. Tran, S. Yunoki, and A. Mezzacapo, Chemistry beyond exact solutions on a quantum-centric supercomputer, *Sci. Adv.* **11**, eadu9991 (2025).
 - [15] D. Kaliakin, A. Shajan, J. R. Moreno, Z. Li, A. Mitra, M. Motta, C. Johnson, A. A. Saki, S. Das, I. Sitdikov, A. Mezzacapo, and K. M. Merz, Jr., Accurate quantum-centric simulations of supramolecular interactions, *Commun. Phys.* **8**, 396 (2025).
 - [16] I. Liepuoniute, K. D. Doney, J. Robledo-Moreno, J. A. Job, W. S. Friend, and G. O. Jones, Quantum-centric study of methylene singlet and triplet states, *J. Chem. Theory Comput.* **21**, 5062 (2025).
 - [17] A. Pellow-Jarman, S. McFarthing, D. H. Kang, P. Yoo, E. E. Elala, R. Pellow-Jarman, P. M. Nakliang, J. Kim, and J.-K. K. Rhee, HIVQE: Handover iterative variational quantum eigensolver for efficient quantum chemistry calculations, [arXiv:2503.06292 \[quant-ph\]](#).
 - [18] J. Yu, J. R. Moreno, J. T. Iosue, L. Bertels, D. Claudino, B. Fuller, P. Groszkowski, T. S. Humble, P. Jurcevic, W. Kirby, T. A. Maier, M. Motta, B. Pokharel, A. Seif, A. Shehata, K. J. Sung, M. C. Tran, V. Tripathi, A. Mezzacapo, and K. Sharma, Quantum-centric algorithm for sample-based Krylov diagonalization, [arXiv:2501.09702 \[quant-ph\]](#).
 - [19] Q. Gao, G. O. Jones, M. Motta, M. Sugawara, H. C. Watanabe, T. Kobayashi, E. Watanabe, Y.-y. Ohnishi, H. Nakamura, and N. Yamamoto, Applications of quantum computing for investigations of electronic transitions in phenylsulfonyl-carbazole TADF emitters, *npj Comput. Mater.* **7**, 70 (2021).
 - [20] S. Gocho, H. Nakamura, S. Kanno, Q. Gao, T. Kobayashi, T. Inagaki, and M. Hatanaka, Excited state calculations using variational quantum eigensolver with spin-restricted ansätze and automatically-adjusted constraints, *npj Comput. Mater.* **9**, 13 (2023).
 - [21] P. J. Ollitrault, A. Kandala, C.-F. Chen, P. K. Barkoutsos, A. Mezzacapo, M. Pistoia, S. Sheldon, S. Woerner, J. M.

- Gambetta, and I. Tavernelli, Quantum equation of motion for computing molecular excitation energies on a noisy quantum processor, *Phys. Rev. Res.* **2**, 043140 (2020).
- [22] M. Ganzhorn, D. J. Egger, P. Barkoutsos, P. Ollitrault, G. Salis, N. Moll, M. Roth, A. Fuhrer, P. Mueller, S. Woerner, I. Tavernelli, and S. Filipp, Gate-efficient simulation of molecular eigenstates on a quantum computer, *Phys. Rev. Appl.* **11**, 044092 (2019).
- [23] F. T. Cerasoli, K. Sherbert, J. Sławińska, and M. Buongiorno Nardelli, Quantum computation of silicon electronic band structure, *Phys. Chem. Chem. Phys.* **22**, 21816 (2020).
- [24] T. Ohgoe, H. Iwakiri, M. Kohda, K. Ichikawa, Y. O. Nakagawa, H. O. Valencia, and S. Koh, Demonstrating quantum computation for quasiparticle band structures, *Phys. Rev. Res.* **6**, L022022 (2024).
- [25] D. Singh, S. Mehendale, Arvind, and K. Dorai, Ground and excited state energy calculations of the H₂ molecule using a variational quantum eigensolver algorithm on an NMR quantum simulator, [arXiv:2407.01000](https://arxiv.org/abs/2407.01000) [quant-ph].
- [26] S. H. Sureshbabu, M. Sajjan, S. Oh, and S. Kais, Implementation of quantum machine learning for electronic structure calculations of periodic systems on quantum computing devices, *J. Chem. Inf. Model.* **61**, 2667 (2021).
- [27] S. Zhang, A. Karim, H. M. Quiney, and M. Usman, Full band structure calculation of semiconducting materials on a noisy quantum processor, *Phys. Rev. A* **110**, 062415 (2024).
- [28] S. T. Stober, S. M. Harwood, D. Tenev, P. K. Barkoutsos, T. P. Gujarati, and S. Mostame, Considerations for evaluating thermodynamic properties with hybrid quantum-classical computing work flows, *Phys. Rev. A* **105**, 012425 (2022).
- [29] J. E. Rice, T. P. Gujarati, M. Motta, T. Y. Takeshita, E. Lee, J. A. Latone, and J. M. Garcia, Quantum computation of dominant products in lithium–sulfur batteries, *J. Chem. Phys.* **154**, 134115 (2021).
- [30] P. Chawla, Shweta, K. R. Swain, T. Patel, R. Bala, D. Shetty, K. Sugisaki, S. B. Mandal, J. Riu, J. Nogué, V. S. Prasanna, and B. P. Das, Relativistic variational-quantum-eigensolver calculations of molecular electric dipole moments on quantum hardware, *Phys. Rev. A* **111**, 022817 (2025).
- [31] A. M. de Lima, E. S. Teixeira, E. D. R. de Aguiar Silva, and R. L. Longo, Effects of one-particle reduced density matrix optimization in variational quantum eigensolvers (VQE), *J. Comput. Chem.* **47**, e70289 (2026).
- [32] O. Oumarou, P. J. Ollitrault, C. L. Cortes, M. Scheurer, R. M. Parrish, and C. Gogolin, Molecular properties from quantum Krylov subspace diagonalization, *J. Chem. Theory Comput.* **21**, 4543 (2025).
- [33] X. Cai, W.-H. Fang, H. Fan, and Z. Li, Quantum computation of molecular response properties, *Phys. Rev. Res.* **2**, 033324 (2020).
- [34] K. Huang, X. Cai, H. Li, Z.-Y. Ge, R. Hou, H. Li, T. Liu, Y. Shi, C. Chen, D. Zheng, K. Xu, Z.-B. Liu, Z. Li, H. Fan, and W.-H. Fang, Variational quantum computation of molecular linear response properties on a superconducting quantum processor, *J. Phys. Chem. Lett.* **13**, 9114 (2022).
- [35] A. Kumar, A. Asthana, V. Abraham, T. D. Crawford, N. J. Mayhall, Y. Zhang, L. Cincio, S. Tretiak, and P. A. Dub, Quantum simulation of molecular response properties in the NISQ era, *J. Chem. Theory Comput.* **19**, 9136 (2023).
- [36] S.-N. Sun, B. Marinelli, J. M. Koh, Y. Kim, L. B. Nguyen, L. Chen, J. M. Kreikebaum, D. I. Santiago, I. Siddiqi, and A. J. Minnich, Quantum computation of frequency-domain molecular response properties using a three-qubit iToffoli gate, *npj Quantum Inf.* **10**, 55 (2024).
- [37] H. J. Vallury, M. A. Jones, C. D. Hill, and L. C. L. Hollenberg, Quantum computed moments correction to variational estimates, *Quantum* **4**, 373 (2020).
- [38] H. J. Vallury, M. A. Jones, G. A. L. White, F. M. Creevey, C. D. Hill, and L. C. L. Hollenberg, Noise-robust ground state energy estimates from deep quantum circuits, *Quantum* **7**, 1109 (2023).
- [39] L. C. L. Hollenberg, Plaquette expansion in lattice Hamiltonian models, *Phys. Rev. D* **47**, 1640 (1993).
- [40] L. C. L. Hollenberg and N. S. Witte, General nonperturbative estimate of the energy density of lattice Hamiltonians, *Phys. Rev. D* **50**, 3382 (1994).
- [41] L. C. L. Hollenberg and N. S. Witte, Analytic solution for the ground-state energy of the extensive many-body problem, *Phys. Rev. B* **54**, 16309 (1996).
- [42] L. C. L. Hollenberg and M. J. Tomlinson, Staggered magnetisation in the Heisenberg antiferromagnet, *Aust. J. Phys.* **47**, 137 (1994).
- [43] N. S. Witte, L. C. L. Hollenberg, and Z. Weihong, Two-dimensional XXZ model ground-state properties using an analytic Lanczos expansion, *Phys. Rev. B* **55**, 10412 (1997).
- [44] I. Kassal and A. Aspuru-Guzik, Quantum algorithm for molecular properties and geometry optimization, *J. Chem. Phys.* **131**, 224102 (2009).
- [45] H. J. Vallury and L. C. L. Hollenberg, in *2023 IEEE International Conference on Quantum Computing and Engineering (QCE)* (IEEE Publishing, Bellevue, WA, USA, 2023), Vol. 01, pp. 295–299.
- [46] A. Javadi-Abhari, M. Treinish, K. Krsulich, C. J. Wood, J. Lishman, J. Gacon, S. Martiel, P. D. Nation, L. S. Bishop, A. W. Cross, B. R. Johnson, and J. M. Gambetta, Quantum computing with Qiskit, [arXiv:2405.08810](https://arxiv.org/abs/2405.08810) [quant-ph].
- [47] R. M. Parrish and P. L. McMahon, Quantum filter diagonalization: Quantum eigendecomposition without full quantum phase estimation, [arXiv:1909.08925](https://arxiv.org/abs/1909.08925).
- [48] N. H. Stair, R. Huang, and F. A. Evangelista, A multireference quantum Krylov algorithm for strongly correlated electrons, *J. Chem. Theory Comput.* **16**, 2236 (2020).
- [49] N. Yoshioka, M. Amico, W. Kirby, P. Jurcevic, A. Dutt, B. Fuller, S. Garion, H. Haas, I. Hamamura, A. Ivrii, R. Majumdar, Z. Mineev, M. Motta, B. Pokharel, P. Rivero, K. Sharma, C. J. Wood, A. Javadi-Abhari, and A. Mezzacapo, Krylov diagonalization of large many-body Hamiltonians on a quantum processor, *Nat. Commun.* **16**, 5014 (2025).
- [50] P. Jordan and E. Wigner, Über das Paulische Äquivalenzverbot, *Zeitsch. Phys.* **47**, 631 (1928).
- [51] A. G. Taube and R. J. Bartlett, New perspectives on unitary coupled-cluster theory, *Int. J. Quantum Chem.* **106**, 3393 (2006).
- [52] P. D. Nation, H. Kang, N. Sundaresan, and J. M. Gambetta, Scalable mitigation of measurement errors on quantum computers, *PRX Quantum* **2**, 040326 (2021).

- [53] X. Bonet-Monroig, R. Sagastizabal, M. Singh, and T. E. O'Brien, Low-cost error mitigation by symmetry verification, *Phys. Rev. A* **98**, 062339 (2018).
- [54] J. Tilly, P. V. Sriluckshmy, A. Patel, E. Fontana, I. Rungger, E. Grant, R. Anderson, J. Tennyson, and G. H. Booth, Reduced density matrix sampling: Self-consistent embedding and multiscale electronic structure on current generation quantum computers, *Phys. Rev. Res.* **3**, 033230 (2021).
- [55] A. M. Dalzell, N. Hunter-Jones, and F. G. S. L. Brandão, Random quantum circuits transform local noise into global white noise, *Commun. Math. Phys.* **405**, 78 (2024).
- [56] P. Czarnik, A. Arrasmith, P. J. Coles, and L. Cincio, Error mitigation with Clifford quantum-circuit data, *Quantum* **5**, 592 (2021).
- [57] P. Lolur, M. Skogh, W. Dobrautz, C. Warren, J. Biznárová, A. Osman, G. Tancredi, G. Wendin, J. Bylander, and M. Rahm, Reference-state error mitigation: A strategy for high accuracy quantum computation of chemistry, *J. Chem. Theory Comput.* **19**, 783 (2023).
- [58] J. Cioslowski, Connected moments expansion: A new tool for quantum many-body theory, *Phys. Rev. Lett.* **58**, 83 (1987).
- [59] K. Kowalski and B. Peng, Quantum simulations employing connected moments expansions, *J. Chem. Phys.* **153**, 201102 (2020).
- [60] B. Peng and K. Kowalski, Variational quantum solver employing the PDS energy functional, *Quantum* **5**, 473 (2021).
- [61] D. Claudino, B. Peng, N. P. Bauman, K. Kowalski, and T. S. Humble, Improving the accuracy and efficiency of quantum connected moments expansions, *Quantum Sci. Technol.* **6**, 034012 (2021).
- [62] B. Ganoe and M. Head-Gordon, Doubles connected moments expansion: A tractable approximate Horn–Weinstein approach for quantum chemistry, *J. Chem. Theory Comput.* **19**, 9187 (2023).
- [63] S. T. Epstein, A. C. Hurley, R. E. Wyatt, and R. G. Parr, Integrated and integral Hellmann–Feynman formulas, *J. Chem. Phys.* **47**, 1275 (1967).
- [64] R. McWeeny and B. Sutcliffe, *Methods of Molecular Quantum Mechanics*, Pure and Applied Mathematics (Academic Press, 1969).
- [65] S. McArdle and D. P. Tew, Improving the accuracy of quantum computational chemistry using the transcorrelated method, [arXiv:2006.11181](https://arxiv.org/abs/2006.11181) [quant-ph].
- [66] M. Motta, T. P. Gujarati, J. E. Rice, A. Kumar, C. Masteran, J. A. Latone, E. Lee, E. F. Valeev, and T. Y. Takeshita, Quantum simulation of electronic structure with a transcorrelated Hamiltonian: improved accuracy with a smaller footprint on the quantum computer, *Phys. Chem. Chem. Phys.* **22**, 24270 (2020).
- [67] A. Kumar, A. Asthana, C. Masteran, E. F. Valeev, Y. Zhang, L. Cincio, S. Tretiak, and P. A. Dub, Quantum simulation of molecular electronic states with a transcorrelated Hamiltonian: Higher accuracy with fewer qubits, *J. Chem. Theory Comput.* **18**, 5312 (2022).
- [68] P. Schleich, J. S. Kottmann, and A. Aspuru-Guzik, Improving the accuracy of the variational quantum eigensolver for molecular systems by the explicitly-correlated perturbative [2]R12-correction, *Phys. Chem. Chem. Phys.* **24**, 13550 (2022).
- [69] I. O. Sokolov, W. Dobrautz, H. Luo, A. Alavi, and I. Tavernelli, Orders of magnitude increased accuracy for quantum many-body problems on quantum computers via an exact transcorrelated method, *Phys. Rev. Res.* **5**, 023174 (2023).
- [70] W. Dobrautz, I. O. Sokolov, K. Liao, P. L. Ríos, M. Rahm, A. Alavi, and I. Tavernelli, Toward real chemical accuracy on current quantum hardware through the transcorrelated method, *J. Chem. Theory Comput.* **20**, 4146 (2024).
- [71] D. Poulin, M. B. Hastings, D. Wecker, N. Wiebe, A. C. Doherty, and M. Troyer, The Trotter step size required for accurate quantum simulation of quantum chemistry, *Quant. Inf. Comput.* **15**, 0361 (2015).
- [72] J. Lee, D. W. Berry, C. Gidney, W. J. Huggins, J. R. McClean, N. Wiebe, and R. Babbush, Even more efficient quantum computations of chemistry through tensor hypercontraction, *PRX Quantum* **2**, 030305 (2021).
- [73] M. Motta, E. Ye, J. R. McClean, Z. Li, A. J. Minnich, R. Babbush, and G. K.-L. Chan, Low rank representations for quantum simulation of electronic structure, *npj Quantum Inf.* **7**, 83 (2021).
- [74] O. Oumarou, M. Scheurer, R. M. Parrish, E. G. Hohenstein, and C. Gogolin, Accelerating quantum computations of chemistry through regularized compressed double factorization, *Quantum* **8**, 1371 (2024).
- [75] M. Metcalf, N. P. Bauman, K. Kowalski, and W. A. de Jong, Resource-efficient chemistry on quantum computers with the variational quantum eigensolver and the double unitary coupled-cluster approach, *J. Chem. Theory Comput.* **16**, 6165 (2020).
- [76] N. P. Bauman, E. J. Bylaska, S. Krishnamoorthy, G. H. Low, N. Wiebe, C. E. Granade, M. Roetteler, M. Troyer, and K. Kowalski, Downfolding of many-body Hamiltonians using active-space models: Extension of the sub-system embedding sub-algebras approach to unitary coupled cluster formalisms, *J. Chem. Phys.* **151**, 014107 (2019).
- [77] N. P. Bauman, G. H. Low, and K. Kowalski, Quantum simulations of excited states with active-space downfolded Hamiltonians, *J. Chem. Phys.* **151**, 234114 (2019).
- [78] N. P. Bauman, C. Jaroslav, V. Libor, P. Jiří, and K. Kowalski, Variational quantum eigensolver for approximate diagonalization of downfolded Hamiltonians using generalized unitary coupled cluster ansatz, *Quantum Sci. Technol.* **6**, 034008 (2021).
- [79] N. P. Bauman and K. Kowalski, Coupled cluster downfolding theory: towards universal many-body algorithms for dimensionality reduction of composite quantum systems in chemistry and materials science, *Mater. Theory* **6**, 17 (2022).
- [80] R. Huang, C. Li, and F. A. Evangelista, Leveraging small-scale quantum computers with unitarily downfolded Hamiltonians, *PRX Quantum* **4**, 020313 (2023).
- [81] R. Babbush, N. Wiebe, J. McClean, J. McClain, H. Neven, and G. K.-L. Chan, Low-depth quantum simulation of materials, *Phys. Rev. X* **8**, 011044 (2018).
- [82] C.-L. Hong, T. Tsai, J.-P. Chou, P.-J. Chen, P.-K. Tsai, Y.-C. Chen, E.-J. Kuo, D. Srolovitz, A. Hu, Y.-C. Cheng, and H.-S. Goan, Accurate and efficient quantum computations

- of molecular properties using Daubechies wavelet molecular orbitals: A benchmark study against experimental data, *PRX Quantum* **3**, 020360 (2022).
- [83] J. S. Kottmann, P. Schleich, T. Tamayo-Mendoza, and A. Aspuru-Guzik, Reducing qubit requirements while maintaining numerical precision for the variational quantum eigensolver: A basis-set-free approach, *J. Phys. Chem. Lett.* **12**, 663 (2021).
- [84] I. G. Ryabinkin, A. F. Izmaylov, and S. N. Genin, A posteriori corrections to the iterative qubit coupled cluster method to minimize the use of quantum resources in large-scale calculations, *Quantum Sci. Technol.* **6**, 024012 (2021).
- [85] A. Tammaro, D. E. Galli, J. E. Rice, and M. Motta, N-electron valence perturbation theory with reference wave functions from quantum computing: Application to the relative stability of hydroxide anion and hydroxyl radical, *J. Phys. Chem. A* **127**, 817 (2023).
- [86] A. Duan, Matrix product states in quantum information processing, Master's thesis, University of Melbourne, Australia, 2015.
- [87] K. Seki and S. Yunoki, Quantum power method by a superposition of time-evolved states, *PRX Quantum* **2**, 010333 (2021).
- [88] A. Fiusco, Quantum-computed moments via time-evolved states, Master's thesis, University of Melbourne, Australia, 2024.
- [89] M. Jones, Quantum computation, quantum chemistry and Hamiltonian moments, Ph.D. thesis, University of Melbourne, Australia, 2025.
- [90] A guide on reading plotted results: line patterns indicate whether the moments correction was used (dashes) and the trial state (dotted for UCCD). For example, a dash-dotted line indicates moments-corrected results and the UCCD state, while a solid line (no dashes and no dots) represents no moments and the HF state. Colors correspond to backends: green lines for statevector simulation and blue for error-mitigated *ibm_torino* results. Red (Appendix only) indicates *ibm_torino* results without reference-state error mitigation.
- [91] M. Jones, Supporting data for "Moments-based quantum computation of the electric dipole moment of molecular systems" (2025), <https://doi.org/10.26188/30370351>
- [92] This only happens in the presence of significant noise (note the amount by which the mitigation fails in Fig. 4) and can serve as an indicator that the noise is too strong.