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Finite-size effects in periodic coupled cluster calculations

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We provide the first rigorous study of the finite-size error in the simplest and representative coupled cluster theory, namely the coupled cluster doubles (CCD) theory, for gapped periodic systems. Assuming that the CCD equations are solved using exact Hartree-Fock orbitals and orbital energies, we prove that the convergence rate of finite-size error scales as $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$, where $N_{\mathbf{k}}$ is the number of discretization point in the Brillouin zone and characterizes the system size. Our analysis shows that the dominant error lies in the coupled cluster amplitude calculation, and the convergence of the finite-size error in energy calculations can be boosted to $\mathcal{O}(N_{\mathbf{k}}^{-1})$ with accurate amplitudes. This also provides the first proof of the scaling of the finite-size error in the third order Møller-Plesset perturbation theory (MP3) for periodic systems.

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I. INTRODUCTION

In 1990, Kenneth Wilson wrote "*ab initio* quantum chemistry is an emerging computational area that is fifty years ahead of lattice gauge theory, a principal competitor for supercomputer time, and a rich source of new ideas and new approaches to the computation of many fermion systems." [21] Coupled cluster (CC) theory is one of the most advanced *ab initio* quantum chemistry methods, and the coupled cluster singles and doubles with perturbative triples (CCSD(T)) is often referred to as the "gold standard" of molecular quantum chemistry. Compared to the success in molecular systems, applications of the CC theory to periodic systems (i.e., bulk solids and other extended systems) [1, 7, 25] have been much more limited. Nonetheless, thanks to the combined improvements of computational power and numerical algorithms in the past few years, periodic CC calculations have been increasingly performed routinely for ground state and excited state properties of condensed matter systems [5, 12, 25].

Unlike CC calculations for molecular systems, properties of periodic systems need to be evaluated in the thermodynamic limit (TDL). The TDL can be approached by employing a large supercell containing $N_{\mathbf{k}}$ unit cells, but this approach does not take advantage of the translational symmetry and is thus increasingly inefficient as the system size grows. A more efficient approach is to discretize the Brillouin zone (BZ) of one unit cell into $N_{\mathbf{k}}$ grid points, and the most widely used discretization method is a uniform grid called the Monkhorst-Pack grid [16]. The result in the TDL is given by the limit $N_{\mathbf{k}} \to \infty$. Due to the singularity caused by the long range Coulomb interaction, the convergence of the energy and other physical properties towards the TDL is often slow and follows a low-order power law. It is therefore important to understand the precise scaling of the finite-size effects in periodic CC calculations. Finite-size scaling analysis is the foundation for the power law extrapolation to calculate properties in the TDL, as well as for more advanced finite-size error correction schemes [3, 5, 11, 13].

To our knowledge, this work is the first mathematical study of finite-size errors of periodic CC theories. We interpret the finite-size error as numerical quadrature error of a trapezoidal rule applied to certain singular integrals. Thus the main body of this work is (1) to analyze the singularity structure of quantities in CC theories, and (2) to bound the trapezoidal quadrature errors associated with these singular integrands. At first glace, the task (2) seems to be a classical problem in numerical analysis. We therefore emphasize that standard quadrature error analysis for smooth integrands (see e.g., [19]) may produce an overly pessimistic upper bound of the finite-size error that does not decrease at all as $N_{\mathbf{k}}$ increases. Our quadrature error analysis adapts the Poisson summation formula in a new setting and is related to a recently developed trapezoidal rule-based quadrature analysis for certain singular integrals [10]. This provides tighter estimates and is more generally applicable than a previous quadrature analysis based on the partial Euler-Maclaurin formula for finite-size error studies [22].

To simplify the presentation and the analysis, we focus on ground-state energy calculations in three-dimensional (3D) insulating systems using the simplest and representative CC theory, i.e., the coupled cluster doubles (CCD) theory. The Brillouin zone is discretized using a Monkhorst-Pack grid of size $N_{\mathbf{k}}^{\frac{1}{3}} \times N_{\mathbf{k}}^{\frac{1}{3}} \times N_{\mathbf{k}}^{\frac{1}{3}}$. The core of CC theories is the amplitude equation, which is a nonlinear system often solved by iterative methods. In particular, when the amplitude equation is solved using a fixed point iteration with a zero initial guess, it can systematically generate a set of perturbative terms as in the Møller-Plesset perturbation theory [18] that can be represented using Feynman diagrams. In practice, the number of iterations needs to be truncated at some number n, and we refer to the resulting scheme as CCD(n). We assume that the amplitude equations are solved with exact (or in practice, sufficiently accurate) Hartree-Fock orbitals and orbital energies in the TDL (see Appendix A). The main result of this paper is that under such assumptions, the convergence rate of CCD(n) (for any fixed n) is $\mathcal{O}\left(N_{\mathbf{k}}^{-\frac{1}{3}}\right)$ (Theorem 1).

It is worth noting that previous numerical studies [5, 11] have suggested that under different assumptions, the finite-size error of the CCD energy calculation can scale as $\mathcal{O}(N_{\mathbf{k}}^{-1})$. A possible origin of this discrepancy will be discussed at the end of the manuscript in Section VII. The restriction of discussion to CCD(n) is a technical one. Under additional assumptions, the same convergence rate can be established for the converged solution of CCD with $n \to \infty$ (Corollary 3). Many finite-size error correction methods work under the assumption that the error in the CC double amplitude is small, and the finite-size error mainly comes from the evaluation of the total energy using the CC double amplitude [5]. Our analysis reveals that the opposite is true in general: most of the finite-size error is in fact in the CC double amplitude, which is responsible for the $\mathcal{O}\left(N_{\mathbf{k}}^{-\frac{1}{3}}\right)$ convergence rate. On the other hand, with accurate CC double amplitudes, the convergence rate of energy calculations could be improved to $\mathcal{O}\left(N_{\mathbf{k}}^{-1}\right)$ without any further finite-size corrections.

The finite-size error analysis of many quantum chemistry methods can be reduced to certain quadrature error analysis. This perspective has recently provided the first unified finite-size error analysis for the periodic Hartree-Fock and the second order Møller-Plesset perturbation theory (MP2) [22], and similar analysis can be carried out for more complex theories such as the random phase approximation (RPA) and the second order screened exchange (SOSEX) [24]. The commonality of these theories (beyond the Hartree-Fock level) is that they only include certain perturbative terms (referred to as the "particle-hole" Feynman diagrams, see the main text for the explanation), and the associated integrand singularities are relatively weak. As a result, for ground-state energy calculations in 3D insulating systems, the finite-size errors of MP2, RPA, SOSEX all scale as $\mathcal{O}\left(N_{\mathbf{k}}^{-1}\right)$. Starting from the third order Møller-Plesset perturbation theory (MP3), other perturbative terms (referred to as "particle-particle" and "hole-hole" diagrams) must be taken into account, and the singularities in these terms are much stronger. Our new techniques can be used to analyze the quadrature error associated with these terms. Since the MP3 diagrams form a subset of the CCD diagrams, our result also gives the first proof that the finite-size error of the MP3 energy scales as $\mathcal{O}\left(N_{\mathbf{k}}^{-\frac{1}{3}}\right)$ (Corollary 2).

II. BACKGROUND

Denote a unit cell as Ω and its Bravais lattice as $\mathbb{L}$. Denote the corresponding reciprocal Brillouin zone and lattice as Ω^* and $\mathbb{L}^*$. Consider a mean-field (Hartree-Fock) calculation with a Monkhorst-Pack mesh $\mathcal{K}$ which is a uniform mesh of size $N_{\mathbf{k}}$ discretizing Ω^*. Each molecular orbital from the calculation can be represented as

$$\psi_{n\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{N_{\mathbf{k}}}} e^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r}) = \frac{1}{|\Omega|\sqrt{N_{\mathbf{k}}}} \sum_{\mathbf{G}\in\mathbb{L}^*} \hat{u}_{n\mathbf{k}}(\mathbf{G}) e^{\mathrm{i}(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}},$$

where n is a band index and $u_{n\mathbf{k}}$ is periodic with respect to the unit cell. As is common in the chemistry literature, $n \in \{i,j\}$ refers to an occupied (or "hole") orbital and $n \in \{a,b\}$ refers to an unoccupied (or "particle") orbital. Throughout this paper, we use the following *normalized* electron repulsion integral (ERI):

$$\langle n_1\mathbf{k}_1, n_2\mathbf{k}_2 | n_3\mathbf{k}_3, n_4\mathbf{k}_4\rangle = \frac{4\pi}{|\Omega|} \sum_{\mathbf{G}\in\mathbb{L}^*}{}' \frac{1}{|\mathbf{q}+\mathbf{G}|^2} \hat{\varrho}_{n_1\mathbf{k}_1, n_3(\mathbf{k}_1+\mathbf{q})}(\mathbf{G}) \hat{\varrho}_{n_2\mathbf{k}_2, n_4(\mathbf{k}_2-\mathbf{q})}(-\mathbf{G}), \tag{1}$$

where $\mathbf{q} = \mathbf{k}_3 - \mathbf{k}_1$ is the momentum transfer vector, the crystal momentum conservation $\mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4 \in \mathbb{L}^*$ is assumed implicitly, and $\hat{\varrho}_{n'\mathbf{k}',n\mathbf{k}}(\mathbf{G}) = \langle \psi_{n'\mathbf{k}'} | e^{\mathrm{i}(\mathbf{k}'-\mathbf{k}-\mathbf{G})\cdot\mathbf{r}} | \psi_{n\mathbf{k}}\rangle$ is the pair product. This normalized ERI (and the normalized amplitude below) is used mainly for better illustration of the connection between various calculations in the finite and the TDL cases, and will introduce extra $\frac{1}{N_{\mathbf{k}}}$ factors in the energy and amplitude formulations compared to the standard ones in the literature.

In this paper, we consider insulating systems with a direct gap between occupied and virtual bands. In addition, we assume that the orbitals and orbital energies can be exactly evaluated at any $\mathbf{k}$ point and the virtual bands are truncated (i.e., only a finite number of virtual bands are included in the calculations) which corresponds to calculations using a fixed basis set. Lastly, we assume that with a proper gauge, both $\psi_{n\mathbf{k}}(\mathbf{r})$ and $\varepsilon_{n\mathbf{k}}$ are smooth and periodic with respect to $\mathbf{k} \in \Omega^*$. For systems free of topological obstructions [2, 15], these conditions may be replaced by weaker conditions using techniques based on Green's functions. However, such a treatment can introduce a considerable amount of overhead to the presentation, and therefore we adopt the stronger but simpler assumptions on the orbitals and orbital energies as stated above.

All the finite order energy diagrams in CCD share the common form

$$\begin{aligned} E_{\#}^{N_\mathbf{k}} &= \frac{1}{N_\mathbf{k}^3} \sum_{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a \in \mathcal{K}} \sum_{ijab} \left(2 \langle i\mathbf{k}_i, j\mathbf{k}_j | a\mathbf{k}_a, b\mathbf{k}_b \rangle - \langle i\mathbf{k}_i, j\mathbf{k}_j | b\mathbf{k}_b, a\mathbf{k}_a \rangle\right) T_{ijab}^{\#,N_\mathbf{k}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) \\ &= \frac{1}{N_\mathbf{k}^3} \sum_{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a \in \mathcal{K}} \sum_{ijab} W_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) T_{ijab}^{\#,N_\mathbf{k}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a), \end{aligned} \tag{2}$$

where $W_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)$ is the antisymmetrized ERI and the *normalized* double amplitude $T_{ijab}^{\#,N_\mathbf{k}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)$ is different in each term (annotated by $\#$). For example, the double amplitudes in MP2 and MP3-4h2p (reads "4 hole 2 particle", because i, j, k, l are hole indices and a, b are particle indices) energies are defined as

$$T_{ijab}^{\text{MP2},N_\mathbf{k}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) = \frac{1}{\varepsilon_{i\mathbf{k}_i, j\mathbf{k}_j}^{a\mathbf{k}_a, b\mathbf{k}_b}} \langle a\mathbf{k}_a, b\mathbf{k}_b | i\mathbf{k}_i, j\mathbf{k}_j \rangle, \tag{3}$$

$$T_{ijab}^{\text{MP3-4h2p},N_\mathbf{k}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) = \frac{1}{N_\mathbf{k}} \sum_{\mathbf{k}_k \in \mathcal{K}} \sum_{kl} \frac{1}{\varepsilon_{i\mathbf{k}_i, j\mathbf{k}_j}^{a\mathbf{k}_a, b\mathbf{k}_b}} \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j \rangle \frac{\langle a\mathbf{k}_a, b\mathbf{k}_b | k\mathbf{k}_k, l\mathbf{k}_l \rangle}{\varepsilon_{k\mathbf{k}_k, l\mathbf{k}_l}^{a\mathbf{k}_a, b\mathbf{k}_b}}. \tag{4}$$

Unlike the MP2 energy which only involves interactions between particle-hole pairs $(i\mathbf{k}_i, a\mathbf{k}_a)$ and $(j\mathbf{k}_j, b\mathbf{k}_b)$, the MP3-4h2p energy involves interactions between hole-hole pairs $(i\mathbf{k}_i, k\mathbf{k}_k)$ and $(j\mathbf{k}_j, l\mathbf{k}_l)$. It is such interaction terms involving particle-particle or hole-hole pairs in MP3 and CCD that lead to considerable difficulties in the finite-size error analysis, compared to the existing analysis for MP2.

The CCD correlation energy is also defined as Eq. (2) with the double amplitude being an infinite sum of the double amplitudes from a subset of finite order perturbation energies. Specifically, the double amplitude in CCD calculation is defined as the solution of a nonlinear amplitude equation which consists of constant, linear, and quadratic terms. The exact definition of the amplitude equation is provided in Eq. (25).

A common practice in CCD calculation is to solve the amplitude equation approximately using fixed point iterations with a zero initial guess which is equivalent to a quasi-Newton method [17]. After n iterations, we refer to the approximate amplitude as the CCD(n) amplitude and the resulting approximate energy as the CCD(n) energy. At the $(n+1)$th iteration, plugging the CCD(n) amplitude from the previous iteration into the right hand side of the amplitude equation in Eq. (25) gives the CCD$(n+1)$ amplitude. If the mean-field calculation gives a good reference wavefunction and the direct gap between occupied and virtual bands is sufficiently large, this iteration converges and CCD(n) converges to CCD [17]. The CCD(n) scheme is directly related to the Møller-Plesset perturbation theory [18]. For example, MP2 can be identified with CCD(1), and CCD(2) contains all terms in MP2 and MP3, as well as a subset of terms in MP4.

In the TDL with $\mathcal{K}$ converging to Ω^*, the correlation energy in Eq. (2) converges to an integral as

$$E_{\#}^{\text{TDL}} = \frac{1}{|\Omega^*|^3} \int_{(\Omega^*)^{\times 3}} \mathrm{d}\mathbf{k}_i \, \mathrm{d}\mathbf{k}_j \, \mathrm{d}\mathbf{k}_a \sum_{ijab} W_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) T_{ijab}^{\#,\text{TDL}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a), \tag{5}$$

where we note that the double amplitude is converged as well (indicated by its superscript "TDL"). For each finite order perturbation energy in CCD except MP2, its double amplitude also converges to an integral in the TDL. For example, the double amplitude Eq. (4) in MP3-4h2p term converges to

$$T_{ijab}^{\text{MP3-4h2p,TDL}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) = \frac{1}{|\Omega^*|}\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kl} \frac{1}{\varepsilon_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b}} \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j\rangle \frac{\langle a\mathbf{k}_a, b\mathbf{k}_b | k\mathbf{k}_k, l\mathbf{k}_l\rangle}{\varepsilon_{k\mathbf{k}_k,l\mathbf{k}_l}^{a\mathbf{k}_a,b\mathbf{k}_b}}. \quad (6)$$

Since CCD(n) consists of a finite number of perturbation terms, its double amplitude converges to a sum of many integral terms in the TDL. For more background information, we refer readers to Appendix A.

III. STATEMENT OF MAIN RESULTS

Comparing $E_{\#}^{N_{\mathbf{k}}}$ in Eq. (2) and $E_{\#}^{\text{TDL}}$ in Eq. (5), we could split the finite-size error of any term in CCD calculation as

$$\begin{aligned} E_{\#}^{\text{TDL}} - E_{\#}^{N_{\mathbf{k}}} &= \left(\frac{1}{|\Omega^*|^3}\int_{(\Omega^*)^{\times 3}} \mathrm{d}\mathbf{k}_i\,\mathrm{d}\mathbf{k}_j\,\mathrm{d}\mathbf{k}_a - \frac{1}{N_{\mathbf{k}}^3}\sum_{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a\in\mathcal{K}}\right)\sum_{ijab}\left(W_{ijab}T_{ijab}^{\#,\text{TDL}}\right)(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) \\ &\quad + \frac{1}{N_{\mathbf{k}}^3}\sum_{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a\in\mathcal{K}}\sum_{ijab} W_{ijab}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)\left(T_{ijab}^{\#,\text{TDL}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) - T_{ijab}^{\#,N_{\mathbf{k}}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)\right), \end{aligned} \quad (7)$$

where the two parts can be interpreted respectively as the finite-size errors in the **energy calculation using exact amplitudes** and in the **amplitude calculations**. Analyzing these two parts separately, we provide a rigorous estimate of the finite-size error in CCD(n) calculations.

Theorem 1 (Error of CCD(n))**.** *In CCD(n) calculation with any $n > 0$, the finite-size errors in energy calculation using exact amplitudes and in amplitude calculations can be estimated as*

$$\left|\left(\frac{1}{|\Omega^*|^3}\int_{(\Omega^*)^{\times 3}} \mathrm{d}\mathbf{k}_i\,\mathrm{d}\mathbf{k}_j\,\mathrm{d}\mathbf{k}_a - \frac{1}{N_{\mathbf{k}}^3}\sum_{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a\in\mathcal{K}}\right)\sum_{ijab}\left(W_{ijab}T_{ijab}^{CCD(n),TDL}\right)(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)\right| = \mathcal{O}(N_{\mathbf{k}}^{-1}),$$

$$\max_{ijab,\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a\in\mathcal{K}}\left|T_{ijab}^{CCD(n),TDL}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) - T_{ijab}^{CCD(n),N_{\mathbf{k}}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)\right| = \mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}}).$$

Combining these two estimates with Eq. (7), the overall finite-size error in CCD(n) energy calculation is

$$\left|E_{CCD(n)}^{TDL} - E_{CCD(n)}^{N_{\mathbf{k}}}\right| = \mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}}).$$

We remark that in MP2, there is no finite-size error in its amplitude calculation, i.e., $T_{ijab}^{\text{MP2,TDL}} = T_{ijab}^{\text{MP2},N_{\mathbf{k}}}$. As a result, $\left|E_{\text{MP2}}^{\text{TDL}} - E_{\text{MP2}}^{N_{\mathbf{k}}}\right| = \mathcal{O}(N_{\mathbf{k}}^{-1})$ which recovers the result in [22]. Since all terms in MP3 energy are a subset of CCD(2), the above results on CCD(n) also provide a finite-size error analysis for MP3 energy calculation.

Corollary 2. *The finite-size error in MP2 calculations is $\mathcal{O}\left(N_{\mathbf{k}}^{-1}\right)$, and the finite-size error in MP3 calculations is $\mathcal{O}\left(N_{\mathbf{k}}^{-\frac{1}{3}}\right)$.*

Theorem 1 provides the finite-size error estimates for CCD(n) calculations that consist of finite number of perturbative terms (Feynman diagrams) in CCD, but not for the converged CCD calculation. These estimates holds for any fixed number of iterations n even when the iteration does not converge as $n \to \infty$, and the prefactors in these estimates depend on n. Under additional assumptions that can control the regularities of the iterates and guarantee the convergence of the fixed point iterations in the finite and the TDL cases, we show that the convergence rate of the finite-size error for the converged CCD energy calculation matches that of the CCD(n) energy calculations.

Corollary 3 (Error of CCD)**.** *Under additional assumptions, the finite-size error in the CCD energy calculation is* $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$.

IV. PROOF FOR THEOREM 1

IV.1. Setup

In CCD(n) and all its included perturbation terms (e.g., MP2 and MP3), the double amplitudes computed in the finite case can be viewed as tensors

$$\{T_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)\}_{ijab, \mathbf{k}_i\mathbf{k}_j\mathbf{k}_a \in \mathcal{K}} \in \mathbb{C}^{n_{\text{occ}} \times n_{\text{occ}} \times n_{\text{vir}} \times n_{\text{vir}} \times N_{\mathbf{k}} \times N_{\mathbf{k}} \times N_{\mathbf{k}}},$$

where we assume n_{occ} occupied and n_{vir} virtual bands. Meanwhile, the exact double amplitudes in the TDL can be viewed as a set of functions of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \Omega^*$ indexed by band indices i, j, a, b. As shown later in Lemma 4, all these functions are in a function space $\mathbb{T}(\Omega^*)$ with special smoothness properties and the exact double amplitude can be described as

$$\{T_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)\}_{ijab} \in \mathbb{T}(\Omega^*)^{n_{\text{occ}} \times n_{\text{occ}} \times n_{\text{vir}} \times n_{\text{vir}}}.$$

In CCD(n) calculations with a finite $N_{\mathbf{k}}$, the computed amplitude approximates the exact amplitude at $\mathcal{K} \times \mathcal{K} \times \mathcal{K}$. We define a map that evaluates the exact amplitude at this discrete mesh as

$$\begin{aligned}
\mathcal{M}_{\mathcal{K}} : \mathbb{T}(\Omega^*)^{n_{\text{occ}} \times n_{\text{occ}} \times n_{\text{vir}} \times n_{\text{vir}}} &\longrightarrow \mathbb{C}^{n_{\text{occ}} \times n_{\text{occ}} \times n_{\text{vir}} \times n_{\text{vir}} \times N_{\mathbf{k}} \times N_{\mathbf{k}} \times N_{\mathbf{k}}} \\
\{T_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)\}_{ijab} &\longrightarrow \{T_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)\}_{ijab, \mathbf{k}_i\mathbf{k}_j\mathbf{k}_a \in \mathcal{K}}.
\end{aligned}$$

In the following discussions, we use T to refer to amplitude tensors in the finite case and t to refer to amplitude functions in the TDL case. We focus on estimating the error in the amplitude calculation between T and t using the (entrywise) max norm:

$$\|T - \mathcal{M}_{\mathcal{K}} t\|_\infty = \max_{ijab, \mathbf{k}_i\mathbf{k}_j\mathbf{k}_a \in \mathcal{K}} |T_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) - t_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)|. \tag{8}$$

Similarly, define $\|t\|_\infty = \max_{ijab} \|t_{ijab}\|_{L^\infty(\Omega^* \times \Omega^* \times \Omega^*)}$ and $\|\mathcal{M}_{\mathcal{K}} t\|_\infty \leqslant \|t\|_\infty$.

Define the two linear functionals that compute the correlation energy with a given double amplitude in the finite and the TDL cases, respectively, as

$$\begin{aligned}
\mathcal{G}_{N_{\mathbf{k}}}(T) &= \frac{1}{N_{\mathbf{k}}^3} \sum_{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a \in \mathcal{K}} \sum_{ijab} W_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) T_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a), \\
\mathcal{G}_{\text{TDL}}(t) &= \frac{1}{|\Omega^*|^3} \int_{(\Omega^*)^{\times 3}} \mathrm{d}\mathbf{k}_i \, \mathrm{d}\mathbf{k}_j \, \mathrm{d}\mathbf{k}_a \sum_{ijab} W_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) t_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a).
\end{aligned}$$

Furthermore, we denote the two mappings that define the fixed point iterations over the amplitude equations in the finite and the TDL cases, respectively, as

$$\begin{aligned}&\mathcal{F}_{N_{\mathbf{k}}}(T) : \mathbb{C}^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}\times N_{\mathbf{k}}\times N_{\mathbf{k}}\times N_{\mathbf{k}}} \to \mathbb{C}^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}\times N_{\mathbf{k}}\times N_{\mathbf{k}}\times N_{\mathbf{k}}},\\ &\mathcal{F}_{\text{TDL}}(t) : \mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}} \to \mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}},\end{aligned}$$

which correspond to the right hand sides of Eq. (25) and Eq. (27) with all the concerned $i, j, a, b, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$. One main technical result of this paper is to prove that the image of $\mathcal{F}_{\text{TDL}}$ is in $\mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}}$ (see Lemma 4).

Using these notations, the CCD(n) energy calculation in the finite case can be formulated as

$$E_{\text{CCD}(n)}^{N_{\mathbf{k}}} = \mathcal{G}_{N_{\mathbf{k}}}(T_n) \quad \text{with} \quad \begin{array}{ll} T_m & = \mathcal{F}_{N_{\mathbf{k}}}(T_{m-1}), m = 1, 2, \ldots \\ T_0 & = \mathbf{0} \in \mathbb{C}^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}\times N_{\mathbf{k}}\times N_{\mathbf{k}}\times N_{\mathbf{k}}} \end{array}, \tag{9}$$

and in the TDL case can be formulated as

$$E_{\text{CCD}(n)}^{\text{TDL}} = \mathcal{G}_{\text{TDL}}(t_n) \quad \text{with} \quad \begin{array}{ll} t_m & = \mathcal{F}_{\text{TDL}}(t_{m-1}), m = 1, 2, \ldots \\ t_0 & = \mathbf{0} \in \mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}} \end{array}. \tag{10}$$

To connect to the previous notations in Section III, we have

$$T_n = \{T_{ijab}^{\text{CCD}(n),N_{\mathbf{k}}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)\}_{ijab,\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a\in\mathcal{K}} \quad \text{and} \quad t_n = \{T_{ijab}^{\text{CCD}(n),\text{TDL}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)\}_{ijab}.$$

When the two fixed point iterations converge with respect to n, the corresponding CCD(n) energies converge to the CCD energies in the finite and the TDL cases, respectively.

Lastly, we introduce the notations for the trapezoidal quadrature rule that will be used in the proof. Given an n-dimensional cubic domain V, we construct a uniform mesh $\mathcal{X}$ in V by first partitioning V into subdomains uniformly and then sampling one point in each subdomain with the same offset. The (generalized) trapezoidal rule for an integrand $f(\mathbf{x})$ in V using $\mathcal{X}$ is defined as

$$\mathcal{Q}_V(f, \mathcal{X}) = \frac{|V|}{|\mathcal{X}|} \sum_{\mathbf{x}_i\in\mathcal{X}} f(\mathbf{x}_i).$$

Further denote the targeted exact integral and the corresponding quadrature error as

$$\mathcal{I}_V(f) = \int_V f(\mathbf{x})\,\mathrm{d}\mathbf{x}, \qquad \mathcal{E}_V(f, \mathcal{X}) = \mathcal{I}_V(f) - \mathcal{Q}_V(f, \mathcal{X}).$$

In the following analysis, we abuse the notation "C" to denote a generic constant that is independent of any concerned terms in the context unless otherwise specified. In other words, $f \leqslant Cg$ is equivalent to $|f| = \mathcal{O}(|g|)$.

IV.2. Proof Outline

In CCD(n) calculation, the splitting of the finite-size error shown in Eq. (7) can be written as

$$\begin{aligned}\left|E_{\text{CCD}(n)}^{\text{TDL}} - E_{\text{CCD}(n)}^{N_{\mathbf{k}}}\right| &= |\mathcal{G}_{\text{TDL}}(t_n) - \mathcal{G}_{N_{\mathbf{k}}}(T_n)| \\ &\leqslant |\mathcal{G}_{\text{TDL}}(t_n) - \mathcal{G}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}}t_n)| + |\mathcal{G}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}}t_n) - \mathcal{G}_{N_{\mathbf{k}}}(T_n)| \\ &\leqslant |\mathcal{G}_{\text{TDL}}(t_n) - \mathcal{G}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}}t_n)| + C\left\|\mathcal{M}_{\mathcal{K}}t_n - T_n\right\|_{\infty},\end{aligned} \tag{11}$$

where the last two terms can be interpreted as the error in the energy calculation using the exact CCD(n) amplitude t_n and the error in the amplitude calculation, respectively. The last inequality uses the boundedness of the linear operator $\mathcal{G}_{N_\mathbf{k}}$, i.e.,

$$
\begin{aligned}
|\mathcal{G}_{N_\mathbf{k}}(T)| &\leqslant \frac{1}{N_\mathbf{k}^3} \sum_{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a\in\mathcal{K}} \sum_{ijab} |W_{ijab}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) T_{ijab}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)| \\
&\leqslant \frac{1}{N_\mathbf{k}^3} \sum_{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a\in\mathcal{K}} \sum_{ijab} C \max_{ijab,\mathbf{k}_i\mathbf{k}_j\mathbf{k}_c\in\mathcal{K}} |T_{ijab}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)| \leqslant C\|T\|_\infty.
\end{aligned}
$$

This uses the fact that $W_{ijab}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) = \mathcal{O}(1)$.

The error in the energy calculation using exact amplitude can be interpreted as a quadrature error

$$
|\mathcal{G}_{\mathrm{TDL}}(t_n) - \mathcal{G}_{N_\mathbf{k}}(\mathcal{M}_\mathcal{K} t_n)| = \left| \frac{1}{|\Omega^*|^3} \mathcal{E}_{\Omega^*\times\Omega^*\times\Omega^*} \left(\sum_{ijab} \left(W_{ijab}[t_n]_{ijab}\right)(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a), \mathcal{K}\times\mathcal{K}\times\mathcal{K} \right) \right|. \tag{12}
$$

The defined integrand is periodic with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \Omega^*$. As a result, the dominant quadrature error is determined by the smoothness properties of the integrand. The antisymmetrized ERI, W_{ijab}, is made of two ERIs. The ERI $\langle i\mathbf{k}_i, j\mathbf{k}_j | a\mathbf{k}_a, b\mathbf{k}_b\rangle$ is singular (or slightly more accurately, nonsmooth) only at zero momentum transfer $\mathbf{q} = \mathbf{k}_a - \mathbf{k}_i = \mathbf{0}$ (and its periodic images) due to the fraction term in its definition Eq. (1),

$$
\frac{4\pi}{|\Omega|} \frac{\hat{\varrho}_{i\mathbf{k}_i,a(\mathbf{k}_i+\mathbf{q})}(\mathbf{0})\hat{\varrho}_{j\mathbf{k}_j,b(\mathbf{k}_j-\mathbf{q})}(\mathbf{0})}{|\mathbf{q}|^2}.
$$

In this term, the numerator is smooth with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}$ and the singularity solely comes from the denominator that only depends on $\mathbf{q}$. The other ERI $\langle i\mathbf{k}_i, j\mathbf{k}_j | b\mathbf{k}_b, a\mathbf{k}_a\rangle$ in W_{ijab} has similar singularity structure at its zero momentum transfer point.

We characterize the singularity structure of CC amplitudes and ERIs in terms of the algebraic singularity of certain orders (see Section V.1). Our first main technical result is that the singularity structure of all the exact CCD(n) amplitude entries, $[t_n]_{ijab}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)$, is the same as that of the ERI $\langle i\mathbf{k}_i, j\mathbf{k}_j | a\mathbf{k}_a, b\mathbf{k}_b\rangle$ (or equivalently the MP2/CCD(1) amplitude entries).

Lemma 4 (Singularity structure of the amplitude)**.** *In CCD(n) calculation with $n > 0$, each entry of the exact double amplitude $t_n = \{T_{ijab}^{CCD(n),TDL}\}_{ijab}$ belongs to the following function space*

$$
\begin{aligned}
\mathbb{T}(\Omega^*) = \big\{ f(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) : & \ f \textit{ is periodic with respect to } \mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a \in \Omega^*, \\
& f \textit{ is smooth everywhere except at } \mathbf{k}_a = \mathbf{k}_i \textit{ with algebraic singularity of order } 0, \\
& f \textit{ is smooth with respect to } \mathbf{k}_i,\mathbf{k}_j \textit{ at nonsmooth point } \mathbf{k}_a = \mathbf{k}_i \big\}.
\end{aligned}
$$

Our second technical result is the estimate of the quadrature error in the energy calculation using an arbitrary amplitude t whose each entry lies in $\mathbb{T}(\Omega^*)$ (which covers the exact CCD(n) amplitude).

Lemma 5 (Energy error with exact amplitude)**.** *For an arbitrary amplitude $t \in \mathbb{T}(\Omega^*)^{n_{occ}\times n_{occ}\times n_{vir}\times n_{vir}}$, the finite-size error in energy calculation using t can be estimated as*

$$
|\mathcal{G}_{TDL}(t) - \mathcal{G}_{N_\mathbf{k}}(\mathcal{M}_\mathcal{K} t)| \leqslant C N_\mathbf{k}^{-1}.
$$

The above two lemmas together prove that the finite-size error in the energy calculation using the exact CCD(n) amplitude in Eq. (12) scales as $\mathcal{O}(N_{\mathbf{k}}^{-1})$. As will be seen later, this can be much faster than the convergence rate using the numerically computed amplitudes.

Similar to the finite-size error splitting in Eq. (11), the error in amplitude calculation can be split into two terms using the recursive definitions of the CCD(n) amplitudes in the finite and the TDL cases

$$\begin{aligned} \|\mathcal{M}_{\mathcal{K}} t_n - T_n\|_\infty &= \|\mathcal{M}_{\mathcal{K}} \mathcal{F}_{\text{TDL}}(t_{n-1}) - \mathcal{F}_{N_{\mathbf{k}}}(T_{n-1})\|_\infty \\ &\leqslant \|\mathcal{M}_{\mathcal{K}} \mathcal{F}_{\text{TDL}}(t_{n-1}) - \mathcal{F}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}} t_{n-1})\|_\infty + \|\mathcal{F}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}} t_{n-1}) - \mathcal{F}_{N_{\mathbf{k}}}(T_{n-1})\|_\infty . \end{aligned} \tag{13}$$

The first term is the error between the exact CCD(n) amplitude and the one computed using the exact CCD($n-1$) amplitude $\mathcal{M}_{\mathcal{K}} t_{n-1}$. The second term is the error between the amplitude computed using the exact CCD($n-1$) amplitude, $\mathcal{M}_{\mathcal{K}} t_{n-1}$, and the one using the amplitude CCD($n-1$) amplitude, T_{n-1}. The latter can be interpreted as the error accumulation from the CCD($n-1$) amplitude calculation.

To estimate the first error term in the Eq. (13), the error between each exact and approximate amplitude entry using t_{n-1}, i.e.,

$$[\mathcal{M}_{\mathcal{K}} \mathcal{F}_{\text{TDL}}(t_{n-1}) - \mathcal{F}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}} t_{n-1})]_{ijab,\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a}, \quad \forall i,j,a,b,\ \forall \mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a \in \mathcal{K},$$

can be decomposed into the summation of a series of quadrature errors that are associated with the calculation of different linear and quadratic terms in the amplitude equation.

One third technical result is the estimate of these quadrature errors, and it suggests that this first error term is of scale $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$.

Lemma 6 (Amplitude error in a single iteration)**.** *For an arbitrary amplitude $t \in \mathbb{T}(\Omega^*)^{n_{occ}\times n_{occ}\times n_{vir}\times n_{vir}}$, the finite-size error in the next iteration amplitude calculation when using t can be estimated as*

$$\|\mathcal{M}_{\mathcal{K}} \mathcal{F}_{TDL}(t) - \mathcal{F}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}} t)\|_\infty \leqslant C N_{\mathbf{k}}^{-\frac{1}{3}}. \tag{14}$$

To address the second error term in Eq. (13), the goal is to show that the application of $\mathcal{F}_{N_{\mathbf{k}}}$ propagates the error in the CCD($n-1$) amplitude calculation in a controlled way. Specifically, noting that $\mathcal{F}_{N_{\mathbf{k}}}(T)$ consists of constant, linear and quadratic terms of T (see Eq. (25) for details), we can use its explicit definition to show that the second error term can be bounded by the error in the CCD($n-1$) amplitude calculation.

Lemma 7 (Lipschitz continuity of the finite CCD iteration mapping)**.** *For two arbitrary amplitude tensors $T, S \in \mathbb{C}^{n_{occ}\times n_{occ}\times n_{vir}\times n_{vir}\times N_{\mathbf{k}}\times N_{\mathbf{k}}\times N_{\mathbf{k}}}$, the iteration map $\mathcal{F}_{N_{\mathbf{k}}}$ in the finite case satisfies*

$$\|\mathcal{F}_{N_{\mathbf{k}}}(T) - \mathcal{F}_{N_{\mathbf{k}}}(S)\|_\infty \leqslant C\left(1 + \|T\|_\infty + \|S\|_\infty\right)\|T - S\|_\infty. \tag{15}$$

Substitute $t = t_{n-1}$ in Eq. (14) and $T = T_n, S = \mathcal{M}_{\mathcal{K}} t_{n-1}$ in Eq. (15), the error splitting in Eq. (13) can then be further estimated as

$$\|\mathcal{M}_{\mathcal{K}} t_n - T_n\|_\infty \leqslant C\left(1 + \|T_{n-1}\|_\infty + \|t_{n-1}\|_\infty\right)\|\mathcal{M}_{\mathcal{K}} t_{n-1} - T_{n-1}\|_\infty + C N_{\mathbf{k}}^{-\frac{1}{3}}, \tag{16}$$

where the second constant C depends on the exact CCD($n-1$) amplitude t_{n-1}. Combining this recursive relation and the fact that $\mathcal{M}_{\mathcal{K}} t_0 = T_0$, the finite-size error in the amplitude calculation can be estimated as

$$\|\mathcal{M}_{\mathcal{K}} t_n - T_n\|_\infty \leqslant C N_{\mathbf{k}}^{-\frac{1}{3}}, \qquad \forall n > 0,$$

where constant C depends on t_m with $m = 0, 1, 2, \ldots, n-1$. Lastly, we finish our proof of Theorem 1 by combining this estimate, Lemma 5 with $t = t_{n-1}$, and the error splitting in Eq. (11).

V. MAIN TECHNICAL TOOLS

Our main idea is to interpret the finite size errors in the CCD(n) energy and amplitude calculations as numerical quadrature errors of trapezoidal rules over certain singular integrals. Specifically, all the averaged summations over $\mathcal{K}$ in $\mathcal{G}_{N_\mathbf{k}}$ and $\mathcal{F}_{N_\mathbf{k}}$ are trapezoidal rules that approximate corresponding integrations over Ω^* in $\mathcal{G}_{\text{TDL}}$ and $\mathcal{F}_{\text{TDL}}$. The problem is thus reduced to estimating the quadrature errors of trapezoidal rules over the integrands defined in $\mathcal{G}_{\text{TDL}}$ and $\mathcal{F}_{\text{TDL}}$ which consist of ERIs and exact amplitudes.

In general, the asymptotic error of a trapezoidal rule depends on boundary condition and smoothness property of the integrand. In the ideal case when an integrand is periodic and smooth, the quadrature error decays super-algebraically, i.e., faster than $N_\mathbf{k}^{-l}$ with any $l > 0$, according to the standard Euler-Maclaurin formula. (See Lemma 12 with a simple Fourier analysis explanation.) All the integrands defined in $\mathcal{G}_{\text{TDL}}$ and $\mathcal{F}_{\text{TDL}}$ turn out be periodic but have point singularities. Therefore it is important to characterize the singularity structure of ERIs and exact amplitudes that constitute all these concerned integrands.

The proof of Theorem 1 involves two main technical challenges: describing the singularity structure of exact amplitudes in Lemma 4, and the quadrature error estimates for integrands defined in energy and amplitude calculations using exact amplitudes in Lemma 5 and Lemma 6. For the first challenge, we define a class of functions with algebraic singularity of certain orders in Section V.1. For the second challenge, we summarize five general integral forms from the energy and amplitude calculations in Section V.2 and provide tight quadrature error estimates based on Poisson summation formula.

V.1. Algebraic singularity

Consider an ERI $\langle n_1\mathbf{k}_1, n_2\mathbf{k}_2|n_3\mathbf{k}_3, n_4\mathbf{k}_4\rangle$ as a periodic function of $\mathbf{k}_1, \mathbf{k}_2, \mathbf{q} = \mathbf{k}_3 - \mathbf{k}_1$ over Ω^* while $\mathbf{k}_4 = \mathbf{k}_2 - \mathbf{q}$ using the crystal momentum conservation. By its definition, this ERI can be split as

$$\langle n_1\mathbf{k}_1, n_2\mathbf{k}_2|n_3\mathbf{k}_3, n_4\mathbf{k}_4\rangle = \frac{4\pi}{|\Omega|}\frac{\hat{\varrho}_{n_1\mathbf{k}_1,n_3(\mathbf{k}_1+\mathbf{q})}(\mathbf{0})\hat{\varrho}_{n_2\mathbf{k}_2,n_4(\mathbf{k}_2-\mathbf{q})}(\mathbf{0})}{|\mathbf{q}|^2} + \frac{4\pi}{|\Omega|}\sum_{\mathbf{G}\in\mathbb{L}^*\setminus\{\mathbf{0}\}}\frac{\cdots}{|\mathbf{q}+\mathbf{G}|^2}, \quad (17)$$

where all the terms with $\mathbf{G} \neq \mathbf{0}$ are smooth with respect to $\mathbf{k}_1, \mathbf{k}_2, \mathbf{q} \in \Omega^*$ and the singularity of the ERI only comes from the first fraction term at $\mathbf{q} = \mathbf{0}$. The numerator of this fraction is smooth with respect to $\mathbf{k}_1, \mathbf{k}_2, \mathbf{q} \in \Omega^*$ (note the assumption that $\psi_{n\mathbf{k}}(\mathbf{r})$ is smooth with respect to $\mathbf{k}$) and is of scale $\mathcal{O}(|\mathbf{q}|^a)$ with $a \in \{0, 1, 2\}$ near $\mathbf{q} = \mathbf{0}$ using orbital orthogonality. The exact value of a depends on the relation between band indices (n_1, n_2) and (n_3, n_4). As can be verified by direct calculation, this fraction and its derivatives over $\mathbf{q}$ with any fixed $\mathbf{k}_1, \mathbf{k}_2$ satisfy the following characterization.

Definition 8 (Algebraic singularity for univariate functions)**.** *A function $f(\mathbf{x})$ has* ***algebraic singularity of order*** *$\gamma \in \mathbb{R}$ at $\mathbf{x}_0 \in \mathbb{R}^d$ if there exists $\delta > 0$ such that*

$$\left|\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}^{\boldsymbol{\alpha}}} f(\mathbf{x})\right| \leqslant C_{\boldsymbol{\alpha}}|\mathbf{x}-\mathbf{x}_0|^{\gamma-|\boldsymbol{\alpha}|}, \qquad \forall 0 < |\mathbf{x}-\mathbf{x}_0| < \delta,\ \forall \boldsymbol{\alpha} \geqslant 0,$$

where constant $C_{\boldsymbol{\alpha}}$ depends on δ and the non-negative d-dimensional derivative multi-index $\boldsymbol{\alpha}$. For brevity, f is also said to be singular (or nonsmooth) at $\mathbf{x}_0$ with order γ.

Remark 9. *In integral equations, algebraic singularity is commonly used to describe the asymptotic behavior of a kernel function near a singular point. The above definition slightly generalizes this concept to additionally include the asymptotic behaviors of all the derivatives. Note that when $\gamma > 0$, $f(\mathbf{x})$ is continuous but nonsmooth at $\mathbf{x}_0$ since its derivatives can be singular at this point. In this case, we slightly abuse the name and still refer to $\mathbf{x}_0$ as a point of algebraic singularity.*

A simple example of such nonsmooth functions is $p(\mathbf{x})/|\mathbf{x}|^2$ where $p(\mathbf{x})$ is smooth and of scale $\mathcal{O}(|\mathbf{x}|^{\gamma+2})$ near $\mathbf{x} = \mathbf{0}$. Using this concept, the leading fraction term in Eq. (17) is nonsmooth at $\mathbf{q} = \mathbf{0}$ with order $\gamma \in \{-2, -1, 0\}$. Since the smooth terms in Eq. (17) do not change the inequalities in Definition 8 qualitatively, the ERI example is also nonsmooth at $\mathbf{q} = \mathbf{0}$ with order γ. In addition, to connect the algebraic singularities of the ERI example with varying $\mathbf{k}_1, \mathbf{k}_2 \in \Omega^*$, we further introduce the algebraic singularity with respect to one variable for a multivariate function.

Definition 10 (Algebraic singularity for multivariate functions)**.** *A function $f(\mathbf{x}, \mathbf{y})$ is smooth with respect to $\mathbf{y} \in V_Y \subset \mathbb{R}^{d_y}$ for any fixed $\mathbf{x}$ and has algebraic singularity of order γ with respect to $\mathbf{x}$ at $\mathbf{x}_0 \in \mathbb{R}^{d_x}$ if there exists $\delta > 0$ such that*

$$\left|\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}^{\boldsymbol{\alpha}}}\left(\frac{\partial^{\boldsymbol{\beta}}}{\partial \mathbf{y}^{\boldsymbol{\beta}}} f(\mathbf{x}, \mathbf{y})\right)\right| \leqslant C_{\boldsymbol{\alpha},\boldsymbol{\beta}} |\mathbf{x} - \mathbf{x}_0|^{\gamma - |\boldsymbol{\alpha}|}, \quad \forall 0 < |\mathbf{x} - \mathbf{x}_0| < \delta, \forall \mathbf{y} \in V_Y, \forall \boldsymbol{\alpha}, \boldsymbol{\beta} \geqslant 0,$$

where constant $C_{\boldsymbol{\alpha},\boldsymbol{\beta}}$ depends on δ, $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$. Compared to the univariate case in Definition 8, the key additions are the shared algebraic singularity of partial derivatives over $\mathbf{y}$ at $\mathbf{x} = \mathbf{x}_0$ with order γ and the independence of $C_{\boldsymbol{\alpha},\boldsymbol{\beta}}$ on $\mathbf{y} \in V_Y$.

A simple example of such nonsmooth functions is $p(\mathbf{x}, \mathbf{y})/|\mathbf{x}|^2$ where $p(\mathbf{x}, \mathbf{y})$ is smooth and of scale $\mathcal{O}(|\mathbf{x}|^{\gamma+2})$ near $\mathbf{x} = \mathbf{0}$. The first fraction term in Eq. (17) is of this form with $\mathbf{x} = \mathbf{q}$, $\mathbf{y} = (\mathbf{k}_i, \mathbf{k}_j)$, and $\gamma \in \{-2, -1, 0\}$. Therefore the ERI example is smooth everywhere with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q} \in \Omega^*$ except at $\mathbf{q} = \mathbf{0}$ with order $\gamma \in \{-2, -1, 0\}$. If treating the ERI as a function of $\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3$, then the ERI is smooth everywhere except at $\mathbf{k}_3 = \mathbf{k}_1$ with order γ. This defines the algebraic singularity used in $\mathbb{T}(\Omega^*)$ in Lemma 4.

Lemma 4 states that all entries of the exact CCD(n) amplitude, $[t_n]_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)$, are smooth everywhere in $\Omega^* \times \Omega^* \times \Omega^*$ except at $\mathbf{q} = \mathbf{k}_a - \mathbf{k}_i = \mathbf{0}$ with order 0 (as specified in $\mathbb{T}(\Omega^*)$). Due to similar singularity structures between CC amplitudes and ERIs, we also refer to $\mathbf{q}$ as the *momentum transfer of the amplitude*. Recall that the exact CCD(1) amplitude $t_1 = \left\{\langle a\mathbf{k}_a, b\mathbf{k}_b | i\mathbf{k}_i, j\mathbf{k}_j\rangle / \varepsilon_{i\mathbf{k}_i, j\mathbf{k}_j}^{a\mathbf{k}_a, b\mathbf{k}_b}\right\}_{ijab}$ has entries in $\mathbb{T}(\Omega^*)$ and t_n is defined by recursively applying $\mathcal{F}_{\text{TDL}}$ to t_1. It is thus sufficient to prove that

$$\mathcal{F}_{\text{TDL}}(t) \in \mathbb{T}(\Omega^*)^{n_{\text{occ}} \times n_{\text{occ}} \times n_{\text{vir}} \times n_{\text{vir}}}, \quad \forall t \in \mathbb{T}(\Omega^*)^{n_{\text{occ}} \times n_{\text{occ}} \times n_{\text{vir}} \times n_{\text{vir}}}.$$

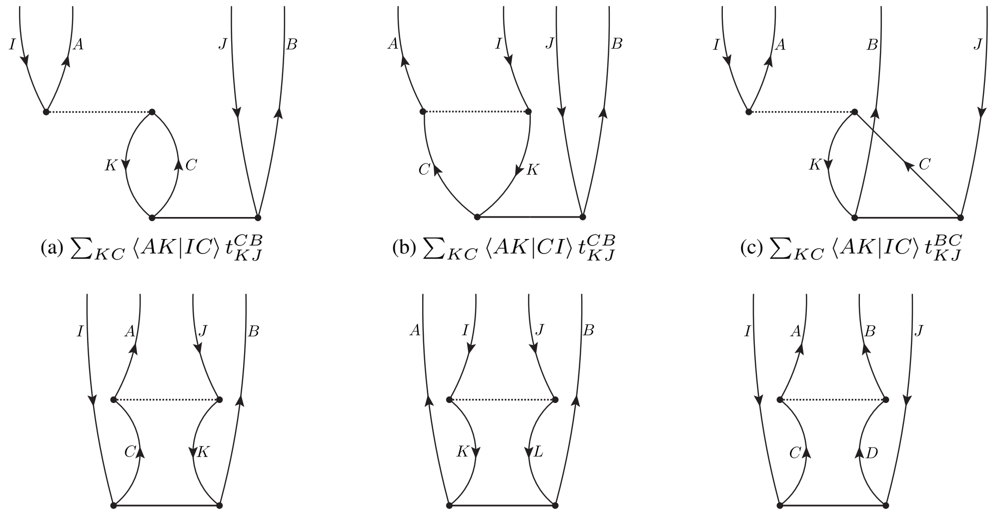
For each set of (i, j, a, b), $[\mathcal{F}_{\text{TDL}}(t)]_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)$ consists of constant, linear, and quadratic terms of t (see Eq. (27)). It turns out that each of these terms as a function of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q} = \mathbf{k}_a - \mathbf{k}_i$ is smooth everywhere except at $\mathbf{q} = \mathbf{0}$ with order 0. The constant term is an MP2 amplitude entry $[t_1]_{ijab}$ and can be verified directly. All the linear terms are of integral form with integrands being the product of an ERI and an amplitude entry, and can be categorized into three classes with representative examples (ignoring orbital energy fraction and constant prefactor) as

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} \langle a\mathbf{k}_a, k\mathbf{k}_k | i\mathbf{k}_i, c\mathbf{k}_c\rangle\, t_{kjcb}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_c), \tag{18}$$

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} \langle a\mathbf{k}_a, k\mathbf{k}_k | i\mathbf{k}_i, c\mathbf{k}_c\rangle\, t_{kjbc}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_b), \tag{19}$$

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kl} \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j\rangle\, t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a), \tag{20}$$

The difference among the three classes is the singularity structure of the integrand with respect to the integration variable, e.g., $\mathbf{k}_k$ in the above examples. For any fixed $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q} \neq \mathbf{0}$, the singular points of integrands



(a) $\sum_{KC} \langle AK|IC\rangle\, t_{KJ}^{CB}$ (b) $\sum_{KC} \langle AK|CI\rangle\, t_{KJ}^{CB}$ (c) $\sum_{KC} \langle AK|IC\rangle\, t_{KJ}^{BC}$

(d) $\sum_{KC} \langle AK|CJ\rangle\, t_{IK}^{CB}$ (e) $\sum_{KL} \langle KL|IJ\rangle\, t_{KL}^{AB}$ (f) $\sum_{CD} \langle AB|CD\rangle\, t_{IJ}^{CD}$

Figure V.1. Diagrams of all the linear terms in the amplitude calculation. The dashed horizontal line denotes the ERI, and the solid horizontal line denotes the t amplitude. The permuted amplitudes from (a), (b), (c), and (d) are not plotted. A capital letter P refers to an orbital index $(p, \mathbf{k}_p)$. The amplitude calculation in (f) can be formulated as an integral over momentum vector $\mathbf{k}_c$, and in all other subplots as integrals over momentum vector $\mathbf{k}_k$.

in the three examples above with respect to $\mathbf{k}_k$ are: (1) none, (2) $\mathbf{k}_k = \mathbf{k}_b$, (3) $\mathbf{k}_k = \mathbf{k}_i$ and $\mathbf{k}_k = \mathbf{k}_a$. Hence the three integrands are nonsmooth with respect to $\mathbf{k}_k$ at zero, one, and two points, respectively. The goal is to show that derivatives of these integrals with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q} \in \Omega^*$ exist except at $\mathbf{q} = \mathbf{0}$ and satisfy the algebraic singularity condition in Definition 10 with order $\gamma = 0$. The first two classes can be addressed using the Leibniz integral rule for the differentiation under integral sign and direct derivative estimates after the interchange of differentiation and integration operations. The third class requires more involved analysis based on an additional technical lemma in Appendix E. Figure V.1 plots diagrams of all the linear amplitude terms where the first class contains (a), the second contains (b) and (c), and the third contains (d), (e) and (f).

All the quadratic terms are also of integral form with integrands being the product of an ERI and two amplitude entries, and the analysis of their smoothness property can be decomposed into two subproblems similar to those for the linear terms above. For example, the 4h2p quadratic term is defined as

$$\frac{1}{|\Omega^*|^2} \int_{\Omega^*} \mathrm{d}\mathbf{k}_k \left(\int_{\Omega^*} \mathrm{d}\mathbf{k}_c \sum_{klcd} \langle k\mathbf{k}_k, l\mathbf{k}_l | c\mathbf{k}_c, d\mathbf{k}_d \rangle\, t_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) \right) t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a). \tag{21}$$

The term in the parenthesis is a linear term as a function of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}, \mathbf{k}_k$ and is singular at $\mathbf{k}_k = \mathbf{k}_i$ with order 0. In other words, its singularity structure is the same as the ERI $\langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j \rangle$ in the 4h2p linear term in Eq. (20). The overall 4h2p quadratic term thus resembles the 4h2p linear term, simply with the ERI replaced by the term in the parenthesis above, and the analysis for linear terms still can be applied to prove its algebraic singularity of order 0 at $\mathbf{q} = \mathbf{0}$.

V.2. Quadrature error for periodic functions with algebraic singularity

Two quadrature error estimate problems need to be addressed for Theorem 1: the error in the energy calculation using exact amplitude in Lemma 5 and the error in the single iteration amplitude calculation using exact amplitude in Lemma 6. Integrands in these two problems are all products of orbital energy fractions, ERIs, and exact amplitudes, and are periodic with respect to involved momentum vectors over Ω^*. Therefore, their smoothness properties determine the dominant quadrature errors with a trapezoidal rule. Specifically, the integrand nonsmoothness only comes from the algebraic singularity of ERIs and amplitudes at their zero momentum transfer points (and their periodic images). The dominant quadrature errors thus come from the numerical quadrature over integration variables dependent on the momentum transfer vectors of the included ERIs and amplitudes.

By proper change of variables and splitting of error terms, all the quadrature error estimates in Lemma 5 and Lemma 6 can be summarized as those for the five types of integrals described in Table V.1. The quadrature errors for these five integrals are studied separately in Lemmas 12, 14, 16, 18 and 20 in Appendix D. In our finite-size error analysis, we have $V = \Omega^*$, $d = 3$, and $m = N_{\mathbf{k}}^{-\frac{1}{3}}$.

Table V.1. Five types of integrals in the quadrature error estimate problems that appear in CCD energy and amplitude calculations. All functions are assumed to be periodic with respect to $V \subset \mathbb{R}^d$ and are smooth everywhere except at the singular points. Parameter d denotes the domain dimension and m denotes the number of points along each dimension in the uniform mesh used for trapezoidal quadrature rule (thus the mesh has m^d points). The "Estimate" column gives the quadrature error estimates for the trapezoidal rule using an m^d-sized mesh, which are obtained in the lemma specified in the "Lemma" column.

Description	Singular points and order	Estimate	Lemma
$\int_V \mathrm{d}\mathbf{x} f(\mathbf{x})$	None	Super-Algebraic	Lemma 12
$\int_V \mathrm{d}\mathbf{x} f(\mathbf{x})$	$\mathbf{x} = \mathbf{0}$ of order γ	$m^{-(d+\gamma)}$	Lemma 14
$\int_V \mathrm{d}\mathbf{x} f_1(\mathbf{x}) f_2(\mathbf{x})$	$f_1(\mathbf{x})$: $\mathbf{x} = \mathbf{0}$ of order γ; $f_2(\mathbf{x})$: $\mathbf{x} = \mathbf{z}$ of order 0	$m^{-(d+\gamma)}$	Lemma 16
$\int_{V\times V} \mathrm{d}\mathbf{x}_1 \, \mathrm{d}\mathbf{x}_2 f_1(\mathbf{x}_1,\mathbf{x}_2) f_2(\mathbf{x}_1,\mathbf{x}_2)$	$f_i(\mathbf{x}_1,\mathbf{x}_2)$: $\mathbf{x}_i = \mathbf{0}$ of order γ_i, $i = 1, 2$	$m^{-(d+\min_i \gamma_i)}$	Lemma 18
$\int_{V\times V} \mathrm{d}\mathbf{x}_1 \, \mathrm{d}\mathbf{x}_2 f_1(\mathbf{x}_1,\mathbf{x}_2) f_2(\mathbf{x}_1,\mathbf{x}_2) f_3(\mathbf{x}_1,\mathbf{x}_2 \pm \mathbf{x}_1)$	$f_i(\mathbf{x}_1,\mathbf{x}_2)$: $\mathbf{x}_i = \mathbf{0}$ of order γ_i, $i = 1, 2$; $f_3(\mathbf{x}_1,\mathbf{z})$: $\mathbf{z} = \mathbf{0}$ of order 0	$m^{-(d+\min_i \gamma_i)}$	Lemma 20

To demonstrate the classification in Table V.1, we provide three examples.

Example 1: The 4h2p linear terms in the amplitude calculation with any fixed i, j, a, b and $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ using an exact amplitude $t \in \mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}}$ is of the form

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kl} \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j \rangle \, t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a). \tag{22}$$

The ERI and the amplitude have momentum transfers $\mathbf{q}_1 = \mathbf{k}_i - \mathbf{k}_k$ and $\mathbf{q}_2 = \mathbf{k}_a - \mathbf{k}_k$, respectively. By the change of variable $\mathbf{k}_k \to \mathbf{k}_i - \mathbf{q}_1$, the integral with each k, l over $\mathbf{q}_1 \in \Omega^*$ is a product of two functions that have algebraic singularity at $\mathbf{q}_1 = \mathbf{0}$ and $\mathbf{q}_2 = \mathbf{q}_1 + (\mathbf{k}_a - \mathbf{k}_i) = \mathbf{0}$ and thus belongs to type 3 in Table V.1. Since the ERI with $k = i, l = j$ is nonsmooth at $\mathbf{q}_1 = \mathbf{0}$ with order -2, the overall quadrature error in the calculation of this term using $\mathcal{K}$ is of scale $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$ according to the estimate.

Example 2: The correlation energy exchange term using an exact amplitude t is of form

$$\int_{\Omega^* \times \Omega^* \times \Omega^*} \mathrm{d}\mathbf{k}_i \, \mathrm{d}\mathbf{k}_j \, \mathrm{d}\mathbf{k}_a \sum_{ijab} \langle i\mathbf{k}_i, j\mathbf{k}_j | b\mathbf{k}_b, a\mathbf{k}_a \rangle \, t_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a).$$

The ERI and the amplitude have momentum transfers $\mathbf{q}_1 = \mathbf{k}_b - \mathbf{k}_i$ and $\mathbf{q}_2 = \mathbf{k}_a - \mathbf{k}_i$, respectively. By the change of variables $\mathbf{k}_j \to \mathbf{k}_a + \mathbf{q}_1$ and $\mathbf{k}_i \to \mathbf{k}_a - \mathbf{q}_2$, the integrand is defined over $\mathbf{k}_a, \mathbf{q}_1, \mathbf{q}_2 \in \Omega^*$ and is smooth with respect to $\mathbf{k}_a$. As a result, the partial integral over $\mathbf{k}_a$ belongs to type 1 and for each fixed $\mathbf{k}_a$ the partial integral over $\mathbf{q}_1, \mathbf{q}_2$ belongs to type 4. Since the included ERIs and amplitudes all have algebraic singularities of order 0, the quadrature error in calculating this term scales as $\mathcal{O}(N_{\mathbf{k}}^{-1})$.

Example 3: The 4h2p quadratic term in the amplitude calculation is of form

$$\int_{\Omega^* \times \Omega^*} \mathrm{d}\mathbf{k}_k \, \mathrm{d}\mathbf{k}_c \sum_{klcd} \langle k\mathbf{k}_k, l\mathbf{k}_l | c\mathbf{k}_c, d\mathbf{k}_d \rangle \, t_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a),$$

where $\mathbf{k}_l = \mathbf{k}_i + \mathbf{k}_j - \mathbf{k}_k$ and $\mathbf{k}_d = \mathbf{k}_i + \mathbf{k}_j - \mathbf{k}_c$ by crystal momentum conservation. The three momentum transfers are $\mathbf{q}_1 = \mathbf{k}_c - \mathbf{k}_k$, $\mathbf{q}_2 = \mathbf{k}_c - \mathbf{k}_i$, and $\mathbf{q}_3 = \mathbf{k}_a - \mathbf{k}_k$. By the change of variables $\mathbf{k}_c \to \mathbf{k}_i + \mathbf{q}_2$ and $\mathbf{k}_k \to \mathbf{k}_i + \mathbf{q}_2 - \mathbf{q}_1$, we have $\mathbf{q}_3 = \mathbf{q}_1 - \mathbf{q}_2 + (\mathbf{k}_a - \mathbf{k}_i)$ and the integral over $\mathbf{q}_1, \mathbf{q}_2$ belongs to type 5. Since the included ERIs and amplitudes all have algebraic singularities of order 0, the quadrature error in calculating this term scales as $\mathcal{O}(N_{\mathbf{k}}^{-1})$.

Table V.2 summarizes the quadrature error estimates for all the CCD amplitude and energy calculations in the proofs of Lemma 5 and Lemma 6. Note that all entries of the exact CCD(n) amplitude t_n are smooth everywhere except at $\mathbf{k}_a = \mathbf{k}_i$ with order 0. The order of singularity of an ERI $\langle n_1\mathbf{k}_1, n_2\mathbf{k}_2 | n_3\mathbf{k}_3, n_4\mathbf{k}_4 \rangle$ at $\mathbf{k}_3 - \mathbf{k}_1 = \mathbf{0}$ equals to -2, -1, and 0 respectively when its band indices match (i.e., $n_1 = n_3, n_2 = n_4$), partially match (i.e., $n_1 = n_3, n_2 \neq n_4$ or $n_1 \neq n_3, n_2 = n_4$), and do not match (i.e., $n_1 \neq n_3, n_2 \neq n_4$). Excluding the special terms with super-algebraically decaying errors, the rule of thumb for these quadrature error estimates is that one calculation can have $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$, $\mathcal{O}(N_{\mathbf{k}}^{-\frac{2}{3}})$, and $\mathcal{O}(N_{\mathbf{k}}^{-1})$ errors respectively when it contains ERIs with matching, partially matching, and no-matching band indices. For example, the 4h2p linear term in Eq. (22) consists of ERI-amplitude products with varying band indices k, l. The products with (1) $k = i, l = j$, (2) $k = i, l \neq j$ or $k \neq i, l = j$, and (3) $k \neq i, l \neq j$ have respectively $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$, $\mathcal{O}(N_{\mathbf{k}}^{-\frac{2}{3}})$, and $\mathcal{O}(N_{\mathbf{k}}^{-1})$ quadrature errors due to the ERI $\langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j \rangle$. The overall quadrature error in the 4h2p linear term is thus $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$, dominated by the product term with $k = i, l = j$. In CCD amplitude calculations, ERIs with matching or partially matching band indices only appear in terms whose diagrams have interaction vertices with ladder structure, see Fig. V.1 for the four linear terms in Table V.2 with $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$ error.

VI. NUMERICAL EXAMPLES

To demonstrate the finite-size errors in the energy and amplitude calculations with large $\mathbf{k}$-point meshes, we consider a model system with an effective potential field. Using a fixed effective potential field, we can obtain orbitals and orbital energies in the TDL at any $\mathbf{k}$ point, which satisfies the assumptions in Theorem 1. This simplified setup also enables us to perform large calculations using up to $N_{\mathbf{k}} = 16^3 = 4096$ $\mathbf{k}$ points, which can be interpreted as a supercell with 4096 atoms in total.

Table V.2. Error estimate for each individual term that appear in CCD energy (using exact amplitudes) as well as amplitude calculations. All amplitude terms assume fixed I, J, A, B and the integral over momentum vector and the summation over band indices are over intermediate orbitals, e.g., K, L, C, D. Each of the 3h3p linear terms permuted by $\mathcal{P}$ in Eq. (27) has the same error estimates as the unpermuted one and is not listed here.

	Type	Terms	Error Estimate
Energy		$\sum_{IJAB} \langle IJ\|AB\rangle\, t_{IJ}^{AB}$, $\sum_{IJAB} \langle IJ\|BA\rangle\, t_{IJ}^{AB}$	$N_{\mathbf{k}}^{-1}$
Amplitude	constant	$\langle AB\|IJ\rangle$	0
	linear	$\langle KL\|IJ\rangle\, t_{KL}^{AB}$, $\langle AB\|CD\rangle\, t_{IJ}^{CD}$, $\langle AK\|CI\rangle\, t_{KJ}^{CB}$, $\langle AK\|CJ\rangle\, t_{KI}^{BC}$	$N_{\mathbf{k}}^{-\frac{1}{3}}$
		$\langle AK\|IC\rangle\, t_{KJ}^{BC}$	$N_{\mathbf{k}}^{-1}$
		$\langle AK\|IC\rangle\, t_{KJ}^{CB}$	Super-Algebraic
	quadratic	$\langle LK\|DC\rangle\, t_{IL}^{AD} t_{KJ}^{CB}$	Super-Algebraic
		all other terms	$N_{\mathbf{k}}^{-1}$

Let the unit cell be $\Omega = [0,1]^3$, we use 16 planewave basis functions along each direction to discretize operators and functions in this unit cell (i.e., the number of $\mathbf{G}$ points is $16^3 = 4096$ and this is independent of $N_{\mathbf{k}}$). At each momentum vector $\mathbf{k} \in \Omega^*$, we solve the effective Kohn-Sham equation to obtain $n_{\text{occ}} = 1$ occupied orbital and $n_{\text{vir}} = 1$ virtual orbital where the Gaussian effective potential is defined as

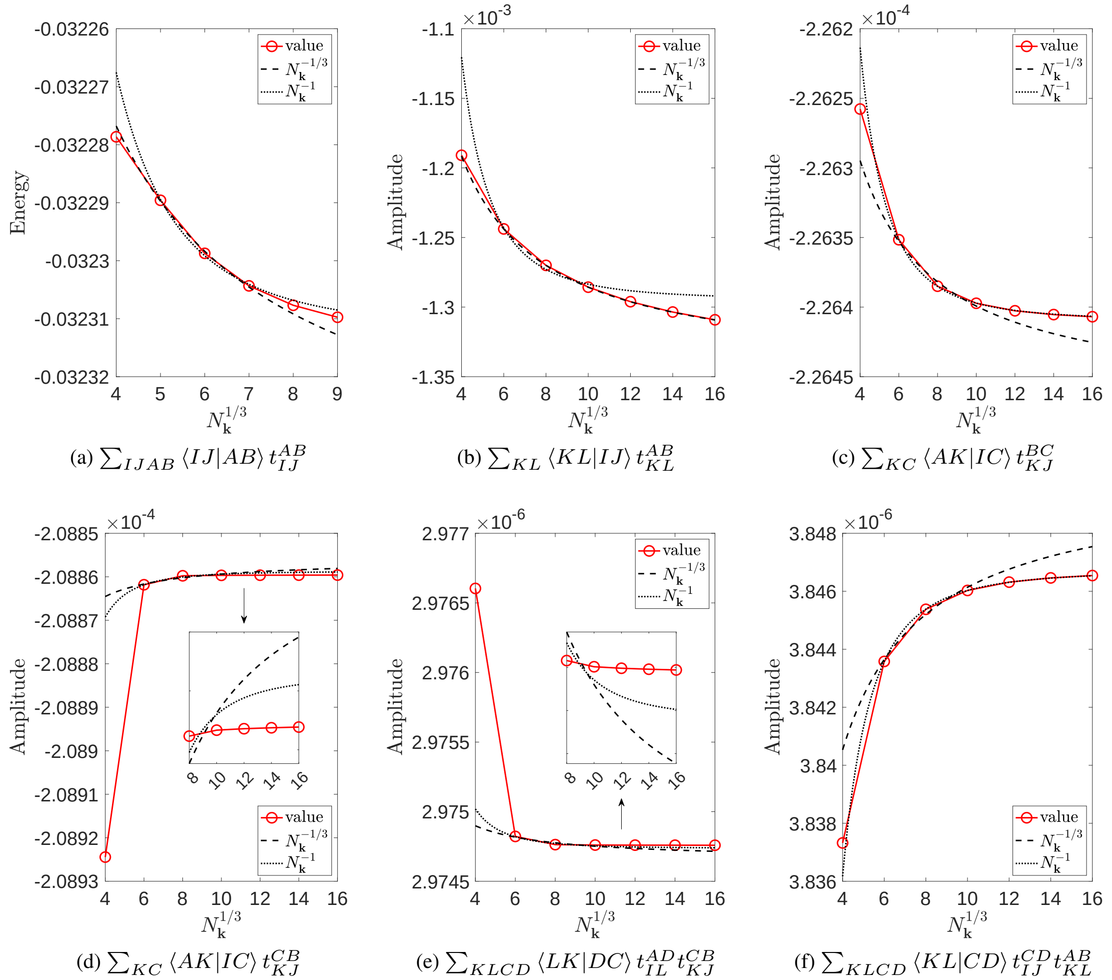
$$V(\mathbf{r}) = \sum_{\mathbf{R}\in\mathbb{L}} C \exp\left(-\frac{1}{2}(\mathbf{r}+\mathbf{R}-\mathbf{r}_0)^\top \Sigma^{-1}(\mathbf{r}+\mathbf{R}-\mathbf{r}_0)\right),$$

with $\mathbf{r}_0 = (0.5, 0.5, 0.5)$, $\Sigma = \operatorname{diag}(0.1^2, 0.2^2, 0.3^2)$, and $C = -200$. This model problem has a direct gap of size around 30.4 between the occupied and virtual bands.

Figure VI.1 plots six calculations that are representative of the error estimates summarized in Table V.2 using the exact CCD(1) amplitude. To identify the asymptotic error scaling without reference values, in each plot, we use the three data points at small $N_{\mathbf{k}}$ to construct the power-law extrapolations (i.e., the curve fitting in the form $C_0 + C_1 N_{\mathbf{k}}^{-s}$) and the discrepancy between the extrapolation and the actual values at larger $N_{\mathbf{k}}$ can then be used to measure the fitting quality. As can be observed, the convergence rates of the tested energy and amplitude calculations are consistent with the theoretical estimates in Table V.2. These results thus justify the finite-size error estimates in Lemma 5 and Lemma 6, and the series of general quadrature error estimates for periodic functions with algebraic singularity obtained in Lemmas 12, 14, 16, 18 and 20. These numerical evidences demonstrate that the estimate of the finite-size error in Theorem 1 is sharp.

VII. DISCUSSION

We have investigated the convergence rate of the periodic coupled cluster theory calculations towards the thermodynamic limit. The analysis in this paper focuses on the simplest and representative CC theory, i.e., the coupled cluster doubles (CCD) theory. Since CCD consists of many finite order perturbation energy terms from Møller-Plesset perturbation theory, this also provides the first finite-size error analysis of these included perturbation energy terms, e.g., MP3. We interpret the finite-size error as numerical quadrature error. The key steps include: (1) analyze the singularity structure, i.e., the *algebraic singularity* of the integrand; (2) bound the quadrature error of the (univariate or multivariate) trapezoidal rule of singular integrands with certain algebraic singularity. Our quadrature analysis based on the Poisson summation formula for certain "punctured" trapezoidal rules and may be of independent interest in other contexts.



(a) $\sum_{IJAB} \langle IJ|AB\rangle t_{IJ}^{AB}$ (b) $\sum_{KL} \langle KL|IJ\rangle t_{KL}^{AB}$ (c) $\sum_{KC} \langle AK|IC\rangle t_{KJ}^{BC}$

(d) $\sum_{KC} \langle AK|IC\rangle t_{KJ}^{CB}$ (e) $\sum_{KLCD} \langle LK|DC\rangle t_{IL}^{AD} t_{KJ}^{CB}$ (f) $\sum_{KLCD} \langle KL|CD\rangle t_{IJ}^{CD} t_{KL}^{AB}$

Figure VI.1. Energy and amplitude calculations using exact CCD(1) amplitude. All the amplitudes are evaluated at $\mathbf{k}_i = \mathbf{k}_j = (0,0,0), \mathbf{k}_a = (0,0,\pi)$ and $(i,j,a,b) = (1,1,2,2)$. The power-law extrapolations use the three data points at $N_\mathbf{k} = 5^3, 6^3, 7^3$ in (a) and $N_\mathbf{k} = 6^3, 8^3, 10^3$ in the remaining subplots. These calculations are estimated theoretically in Table V.2 to have quadrature errors decay asymptotically in the rate of $N_\mathbf{k}^{-1}$, $N_\mathbf{k}^{-\frac{1}{3}}$, $N_\mathbf{k}^{-1}$, super-algebraically, super-algebraically, and $N_\mathbf{k}^{-1}$, respectively. In subplots (d) and (e), the actual data curve converges faster than the power-law extrapolations using $N_\mathbf{k}^{-1}$ and $N_\mathbf{k}^{-\frac{1}{3}}$. This can be seen as evidence that the quadrature errors decay super-algebraically.

Our main result in Theorem 1 studied the finite-size error in CCD(n) calculation with any $n > 0$ number of fixed point iterations over the amplitude equation. However, for gapless and small-gap systems, it has been observed in practice that this fixed point iteration might not converge or the amplitude equation might have multiple solutions. In the case of divergence, the perturbative interpretation of CCD is not valid any more and our analysis over CCD(n) can not be exploited to study the finite size error in the CCD energy calculation. In the case of multiple solutions, CCD itself is not well defined and nor is the problem of its finite-size error analysis.

Our finite-size error analysis is not applicable to theories that do not directly rely on Hartree-Fock orbitals and orbital energies, such as tensor network methods or quantum Monte Carlo methods. The precise analysis of these methods may require a more detailed understanding of the behavior of structure factors [4, 9]. For gapless systems (e.g., metals), additional singularities are introduced by the orbital energy differences of the form $(\varepsilon_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b})^{-1}$ and the occupation number near the Fermi surface. Quadrature error analysis in this case needs to take into account of these additional singularity structures, and the finite-size scaling in metals can also be qualitatively different from that in gapped systems [5, 8, 14].

Our statement that the finite-size error in CCD energy calculation scales as $\mathcal{O}(N_{\mathbf{k}}^{-\frac{1}{3}})$ may seem pessimistic compared to numerical results in the literature [5, 11], which find the finite-size error of certain CC calculations can scale as $\mathcal{O}(N_{\mathbf{k}}^{-1})$ when the nonlinear amplitude equation is solved self-consistently. Our analysis is sharp for any constant number of iteration steps in the CCD(n) scheme, under the assumption of the Hartree-Fock orbitals and orbital energies can be evaluated exactly at any given $\mathbf{k}$ point. The orbital energies are needed in setting up the CC iterations (Eq. (25)), and the correction of finite-size errors in the occupied orbital energies are important for the accurate evaluation of the Fock exchange energy. However, since the abstract form of the CC amplitude equation (Eq. (24)) can be statement without explicitly referring to orbital energies, there may be a fortuitous error cancellation when the iteration scheme reaches self-consistency. Specifically, if CCD calculation uses inexact Hartree-Fock orbital energies without any correction, the special structure of CCD amplitude equations implies that this simple scheme can be equivalent to a more complex one, which simultaneously applies the Madelung constant correction to the Hartree-Fock orbital energies [23], and the shifted Ewald kernel [4] correction to the ERIs. In other words, the finite-size error correction to the orbital energy alone may be detrimental in CCD theories. The analysis of this more complex method is beyond the scope of this work. A viable path may be combining the quadrature error analysis with the singularity subtraction method [6, 20, 23] for simultaneous correction of the orbital energies and ERIs.

While we have focused on the finite-size error of the ground state energy, we think our quadrature based analysis includes some of the essential ingredients in analyzing the finite-size errors for a wide range of diagrammatic methods in quantum physics and quantum chemistry, such as n-th order Møller-Plesset perturbation theory (MPn), GW, CCSD, CCSD(T), and equation of motion coupled cluster (EOM-CC) theories.

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Appendix A BRIEF INTRODUCTION OF MP3 AND CCD

The double amplitude $T_{ijab}^{\#,N_{\mathbf{k}}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)$ is commonly denoted in the literature as $t_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b}$ which assumes implicitly the crystal momentum conservation,

$$\mathbf{k}_i+\mathbf{k}_j-\mathbf{k}_a-\mathbf{k}_b\in\mathbb{L}^*,$$

and a set of $\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a$ can determine a unique $\mathbf{k}_b\in\Omega^*$ accordingly. This explains our notation of the double amplitude as a function of $\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a$ with discrete indices i,j,a,b. For brevity, we use a capital letter P to denote an orbital index $(p,\mathbf{k}_p)$, and use $P\in\{I,J,K,L\}$ to refer to occupied orbitals (a.k.a., holes) and $P\in\{A,B,C,D\}$ to refer to unoccupied orbitals (a.k.a., particles). Any summation $\sum_P$ refers to summing over all occupied or virtual band indices p and all momentum vectors $\mathbf{k}_p\in\mathcal{K}$ while the crystal momentum conservation is enforced according to the summand.

A.1 Amplitude in the finite case

With a finite $\mathbf{k}$-point mesh $\mathcal{K}$ of size $N_{\mathbf{k}}$, the normalized MP3 amplitude $T_{ijab}^{\text{MP3},N_{\mathbf{k}}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)=t_{IJ}^{AB}$ with $\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a\in\mathcal{K}$ is defined as

$$\begin{aligned} t_{IJ}^{AB} &= \frac{1}{\varepsilon_{IJ}^{AB}}\langle AB|IJ\rangle + \frac{1}{\varepsilon_{IJ}^{AB}}\Bigg[\frac{1}{N_{\mathbf{k}}}\sum_{KL}\langle KL|IJ\rangle\, s_{KL}^{AB} + \frac{1}{N_{\mathbf{k}}}\sum_{CD}\langle AB|CD\rangle\, s_{IJ}^{CD} \\ &\quad + \mathcal{P}\left(\frac{1}{N_{\mathbf{k}}}\sum_{KC}(2\langle AK|IC\rangle - \langle AK|CI\rangle)s_{KJ}^{CB} - \langle AK|IC\rangle\, s_{KJ}^{BC} - \langle AK|CJ\rangle\, s_{KI}^{BC}\right)\Bigg], \end{aligned} \tag{23}$$

where $\varepsilon_{IJ}^{AB}=\varepsilon_I+\varepsilon_J-\varepsilon_A-\varepsilon_B$, $s_{IJ}^{AB}=\langle AB|IJ\rangle/\varepsilon_{IJ}^{AB}$ is the normalized MP2 double amplitude, and $\mathcal{P}$ is a permutation operator defined as $\mathcal{P}(\cdots)_{IJ}^{AB}=(\cdots)_{IJ}^{AB}+(\cdots)_{JI}^{BA}$. The three included summations are referred to as the 4-hole-2-particle (4h2p), 2-hole-4-particle (2h4p), and 3-hole-3-particle (3h3p) terms in MP3 according to the number of dummy occupied and virtual orbitals involved. We note that the summation over each P implicitly enforces the crystal momentum conservation. For example, the MP3-4h2p amplitude is explicitly written as

$$\frac{1}{\varepsilon_{IJ}^{AB}}\frac{1}{N_{\mathbf{k}}}\sum_{KL}\langle KL|IJ\rangle\, s_{KL}^{AB} = \frac{1}{\varepsilon_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b}}\frac{1}{N_{\mathbf{k}}}\sum_{\mathbf{k}_k\in\mathcal{K}}\sum_{kl}\langle k\mathbf{k}_k,l\mathbf{k}_l|i\mathbf{k}_i,j\mathbf{k}_j\rangle\, s_{k\mathbf{k}_k,l\mathbf{k}_l}^{a\mathbf{k}_a,b\mathbf{k}_b},$$

where $\mathbf{k}_l\in\mathcal{K}$ is uniquely determined by $\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_k$.

In CCD theory with a finite mesh $\mathcal{K}$, the wavefunction is represented in an exponential ansatz as

$$|\Psi\rangle = e^{\mathcal{T}}|\Phi\rangle := \exp\left(\frac{1}{N_{\mathbf{k}}}\sum_{IJAB} t_{IJ}^{AB} a_A^\dagger a_B^\dagger a_J a_I\right)|\Phi\rangle,$$

where $a_P^\dagger$ and a_P are creation and annihilation operators, $|\Phi\rangle$ is the reference Hartree-Fock determinant, and $t_{IJ}^{AB}=T_{ijab}^{\text{CCD},N_{\mathbf{k}}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)$ is the *normalized* CCD double amplitude. The double amplitude satisfies the amplitude equation (which is derived from the Galerkin projection) as

$$\langle\Phi_{IJ}^{AB}, e^{-\mathcal{T}}\mathcal{H}_{\mathcal{K}}e^{\mathcal{T}}\Phi\rangle = 0, \qquad \forall I,J,A,B, \tag{24}$$

where $|\Phi_{IJ}^{AB}\rangle = a_A^\dagger a_B^\dagger a_J a_I |\Phi\rangle$ is an excited single Slater determinant and $\mathcal{H}_{\mathcal{K}}$ is the model Hamiltonian with $\mathbf{k}$-point mesh $\mathcal{K}$. In practice, this nonlinear amplitude equation can be solved using a quasi-Newton method [17], which can be equivalently written in the form of a fixed point iteration as

$$\begin{aligned} t_{IJ}^{AB} = \frac{1}{\varepsilon_{IJ}^{AB}} \langle AB|IJ\rangle + \frac{1}{\varepsilon_{IJ}^{AB}}\mathcal{P}\left(\sum_C \kappa_C^A t_{IJ}^{CB} - \sum_K \kappa_I^K t_{KJ}^{AB}\right) + \frac{1}{\varepsilon_{IJ}^{AB}}\Bigg[\frac{1}{N_\mathbf{k}}\sum_{KL}\chi_{IJ}^{KL} t_{KL}^{AB} \\ + \frac{1}{N_\mathbf{k}}\sum_{CD}\chi_{CD}^{AB} t_{IJ}^{CD} + \mathcal{P}\left(\frac{1}{N_\mathbf{k}}\sum_{KC}(2\chi_{IC}^{AK} - \chi_{CI}^{AK})t_{KJ}^{CB} - \chi_{IC}^{AK} t_{KJ}^{BC} - \chi_{CJ}^{AK} t_{KI}^{BC}\right)\Bigg]. \end{aligned} \tag{25}$$

This reformulation of CCD amplitude equation can also be derived from the CCSD amplitude equation in [7] by removing all the terms related to single amplitudes and normalizing the involved ERIs and amplitudes (which gives the extra $1/N_\mathbf{k}$ factor in the equation and the intermediate blocks). The intermediate blocks in the equation are defined as

$$\begin{aligned} \kappa_C^A &= -\frac{1}{N_\mathbf{k}^2}\sum_{KLD}\left(2\langle KL|CD\rangle - \langle KL|DC\rangle\right) t_{KL}^{AD}, \\ \kappa_I^K &= \frac{1}{N_\mathbf{k}^2}\sum_{LCD}\left(2\langle KL|CD\rangle - \langle KL|DC\rangle\right) t_{IL}^{CD}, \\ \chi_{IJ}^{KL} &= \langle KL|IJ\rangle + \frac{1}{N_\mathbf{k}}\sum_{CD}\langle KL|CD\rangle t_{IJ}^{CD}, \\ \chi_{CD}^{AB} &= \langle AB|CD\rangle, \\ \chi_{IC}^{AK} &= \langle AK|IC\rangle + \frac{1}{2N_\mathbf{k}}\sum_{LD}\left(2\langle LK|DC\rangle - \langle LK|CD\rangle\right) t_{IL}^{AD} - \langle LK|DC\rangle t_{IL}^{DA}, \\ \chi_{CI}^{AK} &= \langle AK|CI\rangle - \frac{1}{2N_\mathbf{k}}\sum_{LD}\langle LK|CD\rangle t_{IL}^{DA}, \end{aligned}$$

and their momentum vector indices also assume the crystal momentum conservation

$$\begin{aligned} \kappa_P^Q &\to \mathbf{k}_p - \mathbf{k}_q \in \mathbb{L}^*, \\ \chi_{PQ}^{RS} &\to \mathbf{k}_p + \mathbf{k}_q - \mathbf{k}_r - \mathbf{k}_s \in \mathbb{L}^*. \end{aligned}$$

A.2 Amplitude in the TDL

In the TDL with $\mathcal{K}$ converging to Ω^*, all the averaged summation $N_\mathbf{k}^{-1}\sum_{\mathbf{k}\in\mathcal{K}}$ converge to integration $|\Omega^*|^{-1}\int_{\Omega*} \mathrm{d}\mathbf{k}$ in MP3 and CCD. It is worth noting that the double amplitude is computed approximately on $\mathcal{K}\times\mathcal{K}\times\mathcal{K}$ as a tensor in the finite case and it converges to a function of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ defined in $\Omega^*\times\Omega^*\times\Omega^*$ in the TDL. In MP3, the exact amplitude $T_{ijab}^{\text{MP3,TDL}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) = t_{IJ}^{AB}$ with any $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \Omega^*$ can be formulated according to Eq. (23) as

$$\begin{aligned} t_{IJ}^{AB} = \frac{1}{\varepsilon_{IJ}^{AB}}\langle AB|IJ\rangle + \frac{1}{\varepsilon_{IJ}^{AB}}\Bigg[\frac{1}{|\Omega^*|}\int_{\Omega^*}\mathrm{d}\mathbf{k}_k\sum_{kl}\langle KL|IJ\rangle s_{KL}^{AB} + \frac{1}{|\Omega^*|}\int_{\Omega^*}\mathrm{d}\mathbf{k}_c\sum_{cd}\langle AB|CD\rangle s_{IJ}^{CD} \\ + \mathcal{P}\left(\frac{1}{|\Omega^*|}\int_{\Omega^*}\mathrm{d}\mathbf{k}_k\sum_{kc}(2\langle AK|IC\rangle - \langle AK|CI\rangle)s_{KJ}^{CB} - \langle AK|IC\rangle s_{KJ}^{BC} - \langle AK|CJ\rangle s_{KI}^{BC}\right)\Bigg]. \end{aligned} \tag{26}$$

Similarly in CCD, the amplitude equation in the TDL for the exact amplitudes as functions of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \Omega^*$ can be formulated according to Eq. (25) as

$$t_{IJ}^{AB} = \frac{1}{\varepsilon_{IJ}^{AB}} \langle AB|IJ\rangle + \frac{1}{\varepsilon_{IJ}^{AB}} \mathcal{P}\left(\sum_{C} \kappa_C^A t_{IJ}^{CB} - \sum_{K} \kappa_I^K t_{KJ}^{AB}\right) + \frac{1}{\varepsilon_{IJ}^{AB}} \left[\frac{1}{|\Omega^*|}\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kl} \chi_{IJ}^{KL} t_{KL}^{AB} \right.$$
$$\left. + \frac{1}{|\Omega^*|}\int_{\Omega^*} \mathrm{d}\mathbf{k}_c \sum_{cd} \chi_{CD}^{AB} t_{IJ}^{CD} + \mathcal{P}\left(\frac{1}{|\Omega^*|}\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} (2\chi_{IC}^{AK} - \chi_{CI}^{AK}) t_{KJ}^{CB} - \chi_{IC}^{AK} t_{KJ}^{BC} - \chi_{CJ}^{AK} t_{KI}^{BC}\right)\right], \tag{27}$$

where the intermediate blocks in the TDL are defined as

$$\begin{aligned}
\kappa_C^A &= -\frac{1}{|\Omega^*|^2} \int_{\Omega^*\times\Omega^*} \mathrm{d}\mathbf{k}_k \, \mathrm{d}\mathbf{k}_l \sum_{kld} \left(2\langle KL|CD\rangle - \langle KL|DC\rangle\right) t_{KL}^{AD}, \\
\kappa_I^K &= \frac{1}{|\Omega^*|^2} \int_{\Omega^*\times\Omega^*} \mathrm{d}\mathbf{k}_c \, \mathrm{d}\mathbf{k}_d \sum_{lcd} \left(2\langle KL|CD\rangle - \langle KL|DC\rangle\right) t_{IL}^{CD}, \\
\chi_{IJ}^{KL} &= \langle KL|IJ\rangle + \frac{1}{|\Omega^*|} \int_{\Omega^*} \mathrm{d}\mathbf{k}_c \sum_{cd} \langle KL|CD\rangle \, t_{IJ}^{CD}, \\
\chi_{CD}^{AB} &= \langle AB|CD\rangle, \\
\chi_{IC}^{AK} &= \langle AK|IC\rangle + \frac{1}{2|\Omega^*|} \int_{\Omega^*} \mathrm{d}\mathbf{k}_l \sum_{ld} \left(2\langle LK|DC\rangle - \langle LK|CD\rangle\right) t_{IL}^{AD} - \langle LK|DC\rangle \, t_{IL}^{DA}, \\
\chi_{CI}^{AK} &= \langle AK|CI\rangle - \frac{1}{2|\Omega^*|} \int_{\Omega^*} \mathrm{d}\mathbf{k}_l \sum_{ld} \langle LK|CD\rangle \, t_{IL}^{DA}.
\end{aligned}$$

A.3 Amplitude in CCD(n)

In this paper, we use CCD(n) to refer to solving the CCD amplitude approximately by applying n fixed point iterations with a zero initial guess to the amplitude equation and then using the obtained amplitude to compute an approximate CCD energy. In the finite case, the initial amplitude for CCD(0) is set as zero and when $n = 1$ we have

$$T_{ijab}^{\mathrm{CCD}(1),N_\mathbf{k}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) = \frac{1}{\varepsilon_{IJ}^{AB}} \langle AB|IJ\rangle .$$

Therefore, CCD(1) can be identified with MP2. In CCD(2) calculation, the CCD(1) amplitude is plugged into the right hand side of Eq. (25) where the constant term plus all the linear terms exactly gives the MP3 amplitude in Eq. (23) and all the quadratic terms belong to the MP4 amplitude. Therefore, CCD(2) contains all terms in MP2 and MP3, as well as a subset of MP4.

At the nth iteration, we plug the CCD($n-1$) amplitude from the ($n-1$)th iteration into the right hand side of the amplitude equation in Eq. (25) and the left hand side gives the CCD(n) amplitude, i.e.,

$$T_{ijab}^{\mathrm{CCD}(n),N_\mathbf{k}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) = \frac{1}{\varepsilon_{IJ}^{AB}} \langle AB|IJ\rangle + \frac{1}{\varepsilon_{IJ}^{AB}} \mathcal{P}\left(\sum_{C} \kappa_C^A T_{ijcb}^{\mathrm{CCD}(n-1),N_\mathbf{k}}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) \right.$$
$$\left. - \sum_{K} \kappa_I^K T_{kjab}^{\mathrm{CCD}(n-1),N_\mathbf{k}}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_a)\right) + \frac{1}{\varepsilon_{IJ}^{AB}} \frac{1}{N_\mathbf{k}} \sum_{KL} \chi_{IJ}^{KL} T_{klab}^{\mathrm{CCD}(n-1),N_\mathbf{k}}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) + \cdots$$

where all the involved intermediate blocks are now computed using CCD$(n-1)$ amplitude, e.g.,

$$\kappa_C^A = \frac{1}{N_{\mathbf{k}}^2}\sum_{KLD}\left(2\langle KL|CD\rangle - \langle KL|DC\rangle\right) T_{klad}^{\text{CCD}(n-1),N_{\mathbf{k}}}(\mathbf{k}_k,\mathbf{k}_l,\mathbf{k}_c),$$
$$\chi_{IJ}^{KL} = \langle KL|IJ\rangle + \frac{1}{N_{\mathbf{k}}}\sum_{CD}\langle KL|CD\rangle T_{ijcd}^{\text{CCD}(n-1),N_{\mathbf{k}}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_c).$$

In the finite case, by unfolding the fixed point iteration, the CCD(n) amplitude consists of many averaged summations of products of ERIs and orbital energy fractions which correspond to the double amplitudes of certain perturbation terms in Møller-Plesset perturbation theory [18] with order up to 2^n. Each averaged summation in the unfolded CCD(n) amplitude converges in the TDL to an integral over involved intermediate momentum vectors in Ω^*. As a result, the CCD(n) amplitude in the TDL could be explicitly formulated as a summation of many integrals which are respectively approximated by trapezoidal rules in the finite case.

On the other hand, the CCD(n) amplitude in the TDL can also be defined recursively by applying n fixed point iterations to the amplitude equation in the TDL in Eq. (27), i.e.,

$$\begin{aligned}T_{ijab}^{\text{CCD}(n),\text{TDL}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) &= \frac{1}{\varepsilon_{IJ}^{AB}}\langle AB|IJ\rangle + \frac{1}{\varepsilon_{IJ}^{AB}}\mathcal{P}\left(\sum_c \kappa_{c\mathbf{k}_a}^{a\mathbf{k}_a} T_{ijcb}^{\text{CCD}(n-1),\text{TDL}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)\right.\\ &\left.-\sum_k \kappa_{i\mathbf{k}_i}^{k\mathbf{k}_i} T_{kjab}^{\text{CCD}(n-1),\text{TDL}}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a)\right) + \frac{1}{\varepsilon_{IJ}^{AB}}\frac{1}{|\Omega^*|}\int_{\Omega^*}\mathrm{d}\mathbf{k}_k \sum_{kl}\chi_{IJ}^{KL} T_{klab}^{\text{CCD}(n-1),\text{TDL}}(\mathbf{k}_k,\mathbf{k}_l,\mathbf{k}_a) + \cdots\end{aligned} \tag{28}$$

where all the involved intermediate blocks are computed using exact CCD$(n-1)$ amplitude. Unfolding this fixed point iteration, it can be verified that this recursive definition of the amplitude in the TDL is consistent with the above definition obtained by taking the thermodynamic limit of each individual averaged summation term in the double amplitude in the finite case.

Appendix B PROOF OF THEOREM 1

B.1 Proof of Lemma 4: Singularity structure of the exact CCD(n) amplitude

Based on the smoothness property of ERIs in Section V.1, it can be verified that each CCD(1) amplitude entry, i.e., the MP2 amplitude with any (i,j,a,b), lies in $\mathbb{T}(\Omega^*)$ and thus satisfies the statement in the lemma. Since the exact CCD(n) amplitude with $n > 1$ is defined by recursively applying $\mathcal{F}_{\text{TDL}}$ in Eq. (10) to the CCD(1) amplitude, it is sufficient to prove that $\mathcal{F}_{\text{TDL}}(t) \in \mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}}, \forall t \in \mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}}$.

Consider an arbitrary $t \in \mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}}$. Fixing a set of (i,j,a,b), we focus on analyzing the constant, linear, and quadratic terms included in the entry $[\mathcal{F}_{\text{TDL}}(t)]_{ijab}$

$$\begin{aligned}\left[\mathcal{F}_{\text{TDL}}(t)\right]_{ijab}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a) &= \frac{1}{\varepsilon_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b}}\langle a\mathbf{k}_a, b\mathbf{k}_b|i\mathbf{k}_i, j\mathbf{k}_j\rangle\\ &+\frac{1}{\varepsilon_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b}}\frac{1}{|\Omega^*|}\int_{\Omega^*}\mathrm{d}\mathbf{k}_k\sum_{kl}\langle k\mathbf{k}_k, l\mathbf{k}_l|i\mathbf{k}_i, j\mathbf{k}_j\rangle t_{klab}(\mathbf{k}_k,\mathbf{k}_l,\mathbf{k}_a)\\ &+\frac{1}{\varepsilon_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b}}\frac{1}{|\Omega^*|^2}\int_{\Omega^*}\mathrm{d}\mathbf{k}_k\int_{\Omega^*}\mathrm{d}\mathbf{k}_c\sum_{klcd}\langle k\mathbf{k}_k, l\mathbf{k}_l|c\mathbf{k}_c, d\mathbf{k}_d\rangle t_{ijcd}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_c)t_{klab}(\mathbf{k}_k,\mathbf{k}_l,\mathbf{k}_a)\\ &+\cdots,\end{aligned} \tag{29}$$

where the listed linear and quadratic terms come from the 4h2p term $\chi_{IJ}^{KL} t_{KL}^{AB}$ in Eq. (27) and the neglected terms include all the remaining linear and quadratic terms. Our goal is to prove that each of these terms as a function of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ is in $\mathbb{T}(\Omega^*)$. It can be verified directly that these terms satisfy the periodicity condition described in $\mathbb{T}(\Omega^*)$. Therefore we focus on showing that these terms are smooth everywhere except at $\mathbf{q} = \mathbf{k}_a - \mathbf{k}_i = \mathbf{0}$ with order 0 and is smooth with respect to $\mathbf{k}_i, \mathbf{k}_j \in \Omega^*$ when $\mathbf{q} = \mathbf{0}$. We recall the algebraic singularity for multivariate functions in Definition 10 that a periodic function $f(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)$ is smooth everywhere in except at $\mathbf{k}_a = \mathbf{k}_i$ with order γ if with the change of variable $\mathbf{k}_a \to \mathbf{k}_i + \mathbf{q}$, there exists constants $\{C_{\boldsymbol{\alpha},\boldsymbol{\beta}}\}$ satisfying

$$\left|\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{q}^{\boldsymbol{\alpha}}}\left(\frac{\partial^{\boldsymbol{\beta}}}{\partial(\mathbf{k}_i, \mathbf{k}_j)^{\boldsymbol{\beta}}} f(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q})\right)\right| \leqslant C_{\boldsymbol{\alpha},\boldsymbol{\beta}} |\mathbf{q}|^{\gamma - |\boldsymbol{\alpha}|}, \quad \forall \mathbf{q} \in \Omega^* \setminus \{\mathbf{0}\}, \mathbf{k}_i, \mathbf{k}_j \in \Omega^*, \forall \boldsymbol{\alpha}, \boldsymbol{\beta} \geqslant 0, \tag{30}$$

where the inequality is extended to all $\mathbf{q} \in \Omega^* \setminus \{\mathbf{0}\}$ by using the function smoothness.

The constant term is exactly an MP2/CCD(1) amplitude entry and lies in $\mathbb{T}(\Omega^*)$.

All the linear terms takes the form of an integral over an intermediate momentum vector in Ω^*, and the integrand is products of one ERI and one amplitude entry (see Eq. (29) for an example). These linear terms can be categorized into three classes according to the number of singular points of the integrand with respect to the intermediate momentum vector, see Table B.1. The analysis of smoothness properties with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ is similar for terms in the same class. Below we illustrate the analysis for one example in each class.

Table B.1. Classification of linear amplitude terms by the number of nonsmooth points with respect to the intermediate momentum vector. The 3h3p terms permuted by $\mathcal{P}$ are of the same class as the unpermuted ones and thus not listed. The summations over intermediate orbitals, e.g., K, L, C, D, and the prefactor $1/\varepsilon_{IJ}^{AB}$ are omitted for brevity.

Number of singular points	Linear terms
0	$\langle AK\|IC\rangle\, t_{KJ}^{CB}$
1	$\langle AK\|CI\rangle\, t_{KJ}^{CB}$, $\langle AK\|IC\rangle\, t_{KJ}^{BC}$
2	$\langle KL\|IJ\rangle\, t_{KL}^{AB}$, $\langle AB\|CD\rangle\, t_{IJ}^{CD}$, $\langle AK\|CJ\rangle\, t_{IK}^{CB}$

For linear terms with no singular point, we consider the 3h3p term $\langle AK|IC\rangle\, t_{KJ}^{CB}$ detailed as (ignoring the prefactor and orbital energy fraction),

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} \langle a\mathbf{k}_a, k\mathbf{k}_k | i\mathbf{k}_i, c\mathbf{k}_c\rangle\, t_{kjcb}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_c),$$

where $\mathbf{k}_c = \mathbf{k}_a + \mathbf{k}_k - \mathbf{k}_i = \mathbf{k}_k + \mathbf{q}$. The ERIs and the amplitudes have momentum transfers $\mathbf{k}_i - \mathbf{k}_a = -\mathbf{q}$ and $\mathbf{k}_c - \mathbf{k}_k = \mathbf{q}$, respectively, which are both independent of $\mathbf{k}_k$. As a result, for any $(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}) \in (\Omega^*)^{\times 3}$ with $\mathbf{q} \neq \mathbf{0}$, there exists an open domain containing this point where the above integrand is smooth with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}$ and $\mathbf{k}_k \in \Omega^*$. This meets the condition of the Leibniz integral rule which can then be used to prove that this integral is smooth at all points $(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}) \in (\Omega^*)^{\times 3}$ with $\mathbf{q} \neq \mathbf{0}$ and any derivative of this integral equals to the integral of the corresponding integrand derivatives. The algebraic singularity condition in Eq. (30) for this term at any $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q} \in \Omega^*$ with $\mathbf{q} \neq \mathbf{0}$ can then be verified as

$$\begin{aligned}
\left|\partial_{\mathbf{q}}^{\boldsymbol{\alpha}} \partial_{\mathbf{k}_i\mathbf{k}_j}^{\boldsymbol{\beta}} \int_{\Omega^*} \mathrm{d}\mathbf{k}_k \cdots \right| &= \left| \int_{\Omega^*} \mathrm{d}\mathbf{k}_k \partial_{\mathbf{q}}^{\boldsymbol{\alpha}} \partial_{\mathbf{k}_i\mathbf{k}_j}^{\boldsymbol{\beta}} \cdots \right| \\
&\leqslant C \int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} \left| \sum_{\substack{\boldsymbol{\alpha}_0+\boldsymbol{\alpha}_1=\boldsymbol{\alpha} \\ \boldsymbol{\beta}_0+\boldsymbol{\beta}_1=\boldsymbol{\beta}}} \partial_{\mathbf{q}}^{\boldsymbol{\alpha}_0} \partial_{\mathbf{k}_i\mathbf{k}_j}^{\boldsymbol{\beta}_0} \langle a\mathbf{k}_a, k\mathbf{k}_k | i\mathbf{k}_i, c\mathbf{k}_c\rangle \, \partial_{\mathbf{q}}^{\boldsymbol{\alpha}_1} \partial_{\mathbf{k}_i\mathbf{k}_j}^{\boldsymbol{\beta}_1} t_{kjcb}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_c) \right| \\
&\leqslant C \int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} \left| \sum_{\substack{\boldsymbol{\alpha}_0+\boldsymbol{\alpha}_1=\boldsymbol{\alpha} \\ \boldsymbol{\beta}_0+\boldsymbol{\beta}_1=\boldsymbol{\beta}}} C_{\boldsymbol{\alpha},\boldsymbol{\beta}} |\mathbf{q}|^{0-|\boldsymbol{\alpha}_0|} |\mathbf{q}|^{0-|\boldsymbol{\alpha}_1|} \right| \\
&\leqslant C_{\boldsymbol{\alpha},\boldsymbol{\beta}} |\mathbf{q}|^{-|\boldsymbol{\alpha}|},
\end{aligned} \tag{31}$$

where $C_{\boldsymbol{\alpha},\boldsymbol{\beta}}$ denotes a generic constant depending on $\boldsymbol{\alpha}, \boldsymbol{\beta}$ and the third inequality uses the algebraic singularity of the ERIs and the amplitudes at $\mathbf{q} = \mathbf{0}$ with order 0. Lastly, the ERI terms at $\mathbf{q} = \mathbf{0}$ are smooth with respect to $\mathbf{k}_i, \mathbf{k}_j$ (see Eq. (1)) and so are the amplitudes by the assumption $t_{kjcb} \in \mathbb{T}(\Omega^*)$. We thus can use the Leibniz integral rule to prove that the integral at $\mathbf{q} = \mathbf{0}$ is smooth with respect to $\mathbf{k}_i, \mathbf{k}_j$. The above discussion then shows this integral to be in $\mathbb{T}(\Omega^*)$.

For linear terms with one nonsmooth point, we consider the 3h3p term $\langle AK|CI\rangle \, t_{KJ}^{CB}$ detailed as

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} \langle a\mathbf{k}_a, k\mathbf{k}_k | c\mathbf{k}_c, i\mathbf{k}_i\rangle \, t_{kjcb}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_c),$$

where the ERIs and the amplitudes have momentum transfers $\mathbf{k}_k - \mathbf{k}_i$ and $\mathbf{k}_c - \mathbf{k}_k = \mathbf{q}$, respectively. By the change of variable $\mathbf{k}_k \to \mathbf{k}_i + \mathbf{q}_1$ and the integrand periodicity, this term can be reformulated as

$$\int_{\Omega^*} \mathrm{d}\mathbf{q}_1 \sum_{kc} \langle a\mathbf{k}_a, k(\mathbf{k}_i + \mathbf{q}_1) | c(\mathbf{k}_a + \mathbf{q}_1), i\mathbf{k}_i\rangle \, t_{kjcb}(\mathbf{k}_i + \mathbf{q}_1, \mathbf{k}_j, \mathbf{k}_a + \mathbf{q}_1),$$

where the integrand is nonsmooth at $\mathbf{q}_1 = \mathbf{0}$ due to the ERIs and is asymptotically of scale $\mathcal{O}(1/|\mathbf{q}_1|^2)$ near $\mathbf{q}_1 = \mathbf{0}$. As a result, for any $(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}) \in (\Omega^*)^{\times 3}$ with $\mathbf{q} \neq \mathbf{0}$, there exists an open domain containing this point where the concerned integrand is smooth with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}$ and $\mathbf{q}_1 \in \Omega^* \setminus \{\mathbf{0}\}$ and its absolute value is bounded by $C/|\mathbf{q}_1|^2$ from above which is integrable in Ω^*. This still meets the condition of the Leibniz integral rule which can then be used to prove that this integral is smooth at all points $(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}) \in (\Omega^*)^{\times 3}$ with $\mathbf{q} \neq \mathbf{0}$. The algebraic singularity of the integral at $\mathbf{q} = \mathbf{0}$ can be similarly proved as in Eq. (31), except now that the ERI derivatives are estimated as

$$\left| \partial_{\mathbf{q}}^{\boldsymbol{\alpha}_0} \partial_{\mathbf{k}_i\mathbf{k}_j}^{\boldsymbol{\beta}_0} \langle a\mathbf{k}_a, k(\mathbf{k}_i + \mathbf{q}_1) | c\mathbf{k}_c, i\mathbf{k}_i\rangle \right| \leqslant C_{\boldsymbol{\alpha},\boldsymbol{\beta}}/|\mathbf{q}_1|^2,$$

by noting that the ERI here has momentum transfer $\mathbf{q}_1$ and is smooth with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}$.

For linear terms with two nonsmooth points, we consider the 4h2p linear term $\langle KL|IJ\rangle \, t_{KL}^{AB}$ as detailed in Eq. (29). We first denote the ERI and the amplitude with band indices (k, l) as

$$\begin{aligned}
F_1^{kl}(\mathbf{q}_1, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) &= \langle k(\mathbf{k}_i - \mathbf{q}_1), l(\mathbf{k}_j + \mathbf{q}_1) | i\mathbf{k}_i, j\mathbf{k}_j\rangle \,, \\
F_2^{kl}(\mathbf{q}_2, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) &= t_{klab}(\mathbf{k}_a - \mathbf{q}_2, \mathbf{k}_i + \mathbf{k}_j - \mathbf{k}_a + \mathbf{q}_2, \mathbf{k}_a),
\end{aligned}$$

where $\mathbf{q}_1$ and $\mathbf{q}_2$ are the respective momentum transfers of the two terms. Note that F_1^{kl} does not depend on $\mathbf{k}_a$ which is included as a variable for general cases. Both the ERI and the amplitude depend on $\mathbf{k}_k$ and $\mathbf{k}_l$ and these dependencies will be converted to that of $\mathbf{q}_1, \mathbf{q}_2$ using change of variables and the crystal momentum conservation. For example, we have $t_{klab}(\mathbf{k}_k, \mathbf{k}_i + \mathbf{k}_j - \mathbf{k}_k, \mathbf{k}_a) = t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) = t_{klab}(\mathbf{k}_a - \mathbf{q}_2, \mathbf{k}_i + \mathbf{k}_j - \mathbf{k}_a + \mathbf{q}_2, \mathbf{k}_a)$.

Note that F_1^{kl} can be verified to be periodic and smooth everywhere with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a, \mathbf{q}_1 \in \Omega^*$ except at $\mathbf{q}_1 = \mathbf{0}$ with order $0, -1$, or -2 depending on the relation between (k, l) and (i, j). Similarly, F_2^{kl} is periodic and smooth everywhere with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a, \mathbf{q}_2 \in \Omega^*$ except at $\mathbf{q}_2 = \mathbf{0}$ with order 0 by the assumption $t_{klab} \in \mathbb{T}(\Omega^*)$. Using this notation, the 4h2p linear term can be reformulated as

$$\begin{aligned}&\frac{1}{|\Omega^*|}\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kl} F_1^{kl}(\mathbf{k}_i - \mathbf{k}_k, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) F_2^{kl}(\mathbf{k}_a - \mathbf{k}_k, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) \\ &\quad = \frac{1}{|\Omega^*|}\int_{\Omega^*} \mathrm{d}\mathbf{q}_1 \sum_{kl} F_1^{kl}(\mathbf{q}_1, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) F_2^{kl}(\mathbf{q}_1 - (\mathbf{k}_i - \mathbf{k}_a), \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a),\end{aligned}$$

where the second equation applies $\mathbf{k}_k \to \mathbf{k}_i - \mathbf{q}_1$ and uses the integrand periodicity with respect to $\mathbf{k}_k \in \Omega^*$. Due to the integrand being nonsmooth at $\mathbf{q}_1 = \mathbf{k}_i - \mathbf{k}_a$ and $\mathbf{q}_1 = \mathbf{0}$, the smoothness property of the integral with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ cannot be obtained using the Leibniz integral rule as in the previous two cases. We provide a technical lemma analyzing the singularity structure of such a function in integral form.

Lemma 11. *Let $f(\mathbf{x}_1, \mathbf{x}_2, \mathbf{z})$ be defined in $V \times V \times V_Z$ with $V = [-\frac{1}{2}, \frac{1}{2}]^d$ and $V_Z \subset \mathbb{R}^{d_z}$ of arbitrary dimension d_z. Assume $f(\mathbf{x}_1, \mathbf{x}_2, \mathbf{z})$ is periodic with respect to $\mathbf{x}_1, \mathbf{x}_2 \in V$ and smooth everywhere except at $\mathbf{x}_1 = \mathbf{0}$ or $\mathbf{x}_2 = \mathbf{0}$ where the nonsmooth behavior can be characterized as*

$$\left|\frac{\partial^{\boldsymbol{\alpha}_1}}{\partial \mathbf{x}_1^{\boldsymbol{\alpha}_1}}\frac{\partial^{\boldsymbol{\alpha}_2}}{\partial \mathbf{x}_2^{\boldsymbol{\alpha}_2}}\left(\frac{\partial^{\boldsymbol{\beta}}}{\partial \mathbf{z}^{\boldsymbol{\beta}}} f(\mathbf{x}_1, \mathbf{x}_2, \mathbf{z})\right)\right| \leqslant C_{\boldsymbol{\alpha}_1, \boldsymbol{\alpha}_2, \boldsymbol{\beta}} |\mathbf{x}_1|^{\gamma_1 - |\boldsymbol{\alpha}_1|} |\mathbf{x}_2|^{\gamma_2 - |\boldsymbol{\alpha}_2|}, \forall \mathbf{x}_1, \mathbf{x}_2 \in V \setminus \{\mathbf{0}\}, \mathbf{z} \in V_Z, \quad (32)$$

with any derivative orders $\boldsymbol{\alpha}_1, \boldsymbol{\alpha}_2, \boldsymbol{\beta} \geqslant 0$.

Assuming $\min_i \gamma_i \geqslant -d + 1$, the partially integrated function,

$$F(\mathbf{y}, \mathbf{z}) = \int_V \mathrm{d}\mathbf{x} f(\mathbf{x}, \mathbf{x} - \mathbf{y}, \mathbf{z}),$$

is smooth everywhere in $V \times V_Z$ except at $\mathbf{y} = \mathbf{0}$ with order $\max(\gamma_1, \gamma_2)$.

Proof. See Appendix E. □

To convert to the condition in Lemma 11, we reformulate the integrand for the 4h2p linear term as

$$f(\mathbf{q}_1, \mathbf{q}_2; \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) = \sum_{kl} F_1^{kl}(\mathbf{q}_1, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a) F_2^{kl}(\mathbf{q}_2, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a),$$

which satisfies Eq. (32) with $\mathbf{x}_1 = \mathbf{q}_1$, $\mathbf{x}_2 = \mathbf{q}_2$, $\mathbf{z} = (\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a)$, $\gamma_1 = -2$, and $\gamma_2 = 0$. Lemma 11 shows that the partially integrated function,

$$\frac{1}{|\Omega^*|}\int_{\Omega^*} \mathrm{d}\mathbf{q}_1 f(\mathbf{q}_1, \mathbf{q}_1 - \mathbf{y}; \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a),$$

is periodic and smooth everywhere with respect to $\mathbf{y}, \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \Omega^*$ except at $\mathbf{y} = \mathbf{0}$ with order 0. Since the 4h2p linear term equals this function with $\mathbf{y} = \mathbf{k}_i - \mathbf{k}_a$, we can follow Definition 10 to show that it is periodic and smooth everywhere except at $\mathbf{k}_a = \mathbf{k}_i$ with order 0. This proves the 4h2p linear term to be in $\mathbb{T}(\Omega^*)$.

All the quadratic terms are also of the same integral form where the integration is over two intermediate momentum vectors in Ω^* and the integrand is products of an ERI and two amplitude entries (see Eq. (29) for an example). Smoothness property analysis for these quadratic terms can be decomposed into two subproblems that can be addressed by the earlier analysis for linear terms. We use the 4h2p quadratic term in Eq. (29) to demonstrate the analysis.

In the 4h2p quadratic term, the momentum vectors $\mathbf{k}_l = \mathbf{k}_i + \mathbf{k}_j - \mathbf{k}_k$ and $\mathbf{k}_d = \mathbf{k}_i + \mathbf{k}_j - \mathbf{k}_c$, and the integrand is a function of $\mathbf{k}_k$ and $\mathbf{k}_c$ for any fixed $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$. We first consider the partial integration over $\mathbf{k}_c$ as

$$H_{ijkl}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_k) = \int_{\Omega^*} \mathrm{d}\mathbf{k}_c \sum_{cd} \langle k\mathbf{k}_k, l\mathbf{k}_l | c\mathbf{k}_c, d\mathbf{k}_d \rangle\, t_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c).$$

The integrand here as a function of $\mathbf{k}_c$ is nonsmooth at $\mathbf{k}_c = \mathbf{k}_k$ and $\mathbf{k}_c = \mathbf{k}_i$ both with order 0, due to the ERI and the amplitude. It is thus of the same form as the linear term with two nonsmooth points studied earlier, and we can use the same analysis to show that this intermediate function $H_{ijkl}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_k)$ is smooth everywhere except at $\mathbf{k}_i = \mathbf{k}_k$ with order 0. The overall quadratic term can then be written as

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kl} H_{ijkl}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_k) t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a).$$

The integrand here is of similar form to the 4h2p linear term, simply with the ERI term $\langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j \rangle$ replaced by H_{ijkl} which has the same nonsmooth behavior at $\mathbf{k}_k = \mathbf{k}_i$ with order 0. Using the same analysis for linear terms based on Lemma 11 can then show that this term as a function of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ is smooth everywhere except at $\mathbf{k}_i = \mathbf{k}_a$ with order 0. This proves the 4h2p quadratic term to be in $\mathbb{T}(\Omega^*)$. All the other quadratic terms can be similarly analyzed using the analysis for the three types of linear terms above.

With all the constant, linear, and quadratic terms in $[\mathcal{F}_{\text{TDL}}(t)]_{ijab}$ shown to be in $\mathbb{T}(\Omega^*)$, we finish the proof of Lemma 4.

B.2 Proof of Lemma 5: Error in energy calculation using exact amplitude

Consider an arbitrary amplitude $t \in \mathbb{T}(\Omega^*)^{n_{\text{occ}} \times n_{\text{occ}} \times n_{\text{vir}} \times n_{\text{vir}}}$. By expanding the antisymmetrized ERI, the finite-size error in the energy calculation using t can be decomposed into the errors in the direct and the exchange term calculations as

$$\begin{aligned}\mathcal{G}_{\text{TDL}}(t) - \mathcal{G}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}} t) = {} & \frac{2}{|\Omega^*|^3} \mathcal{E}_{\Omega^* \times \Omega^* \times \Omega^*} \left(\sum_{ijab} \langle i\mathbf{k}_i, j\mathbf{k}_j | a\mathbf{k}_a, b\mathbf{k}_b \rangle\, t_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a), \mathcal{K} \times \mathcal{K} \times \mathcal{K} \right) \\ & - \frac{1}{|\Omega^*|^3} \mathcal{E}_{\Omega^* \times \Omega^* \times \Omega^*} \left(\sum_{ijab} \langle i\mathbf{k}_i, j\mathbf{k}_j | b\mathbf{k}_b, a\mathbf{k}_a \rangle\, t_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a), \mathcal{K} \times \mathcal{K} \times \mathcal{K} \right).\end{aligned}$$

For each set of band indices i, j, a, b, we denote the integrand for the direct term calculation, i.e., the first term above, with the change of variable $\mathbf{k}_a \to \mathbf{k}_i + \mathbf{q}$ as

$$F_{\text{d}}^{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}) = \langle i\mathbf{k}_i, j\mathbf{k}_j | a(\mathbf{k}_i + \mathbf{q}), b(\mathbf{k}_j - \mathbf{q}) \rangle\, t_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_i + \mathbf{q}).$$

The momentum transfers of the ERI and the amplitude entry are both equal to $\mathbf{q}$. Expanding the ERI near $\mathbf{q} = \mathbf{0}$ and using the assumption $t_{ijab} \in \mathbb{T}(\Omega^*)$, we can show that F_{d}^{ijab} is periodic and smooth everywhere with respect to $\mathbf{k}_i, \mathbf{k}_j, \mathbf{q} \in \Omega^*$ except at $\mathbf{q} = \mathbf{0}$ with order 0. Since $\mathbf{k}_i$ and $\mathbf{k}_a$ are sampled on the same mesh $\mathcal{K}$, the induced mesh for $\mathbf{q} = \mathbf{k}_a - \mathbf{k}_i$ (map each $\mathbf{q}$ to $\mathbf{q} + \mathbf{G} \in \Omega^*$ with some $\mathbf{G} \in \mathbb{L}^*$ using the integrand periodicity with respect to Ω^*) is of the same size as $\mathcal{K}$ and always contains $\mathbf{q} = \mathbf{0}$. Denote this induced mesh as $\mathcal{K}_{\mathbf{q}}$. The quadrature error in the direct term calculation with each set of i, j, a, b can then be formulated and split as

$$\begin{aligned}&\mathcal{E}_{\Omega^*\times\Omega^*\times\Omega^*}\left(F_{\mathrm{d}}^{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}), \mathcal{K}\times\mathcal{K}\times\mathcal{K}_{\mathbf{q}}\right)\\ =&\mathcal{E}_{\Omega^*\times\Omega^*}\left(\int_{\Omega^*}\mathrm{d}\mathbf{q} F_{\mathrm{d}}^{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}), \mathcal{K}\times\mathcal{K}\right) + \frac{|\Omega^*|^2}{N_{\mathbf{k}}^2}\sum_{\mathbf{k}_i,\mathbf{k}_j\in\mathcal{K}}\mathcal{E}_{\Omega^*}\left(F_{\mathrm{d}}^{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}), \mathcal{K}_{\mathbf{q}}\right).\end{aligned} \tag{33}$$

Fixing $\mathbf{k}_i, \mathbf{k}_j \in \mathcal{K}$, $F_{\mathrm{d}}^{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q})$ as a function of $\mathbf{q}$ is periodic and smooth everywhere in Ω^* except at $\mathbf{q} = \mathbf{0}$ with order 0 from the analysis above. Lemma 14 provides quadrature error estimates for such periodic functions with a single point of algebraic singularity, and specifically in this case we have

$$\left|\mathcal{E}_{\Omega^*}\left(F_{\mathrm{d}}^{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}), \mathcal{K}_{\mathbf{q}}\right)\right| \leqslant C N_{\mathbf{k}}^{-1}, \quad \forall \mathbf{k}_i, \mathbf{k}_j \in \mathcal{K},$$

where constant C is independent of $\mathbf{k}_i, \mathbf{k}_j$ by using the algebraic singularity characterization of F_{d}^{ijab} at $\mathbf{q} = \mathbf{0}$ and the prefactor estimate in Lemma 14 (see Remark 15).

Since F_{d}^{ijab} is periodic and smooth with respect to $\mathbf{k}_i, \mathbf{k}_j \in \Omega^*$, $\int_{\Omega^*}\mathrm{d}\mathbf{q} F_{\mathrm{d}}^{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q})$ is also periodic and smooth with respect to $\mathbf{k}_i, \mathbf{k}_j \in \Omega^*$ using the Leibniz integral rule. According to Lemma 12, the quadrature error for this partially integrated function (the first term in Eq. (33)) decays super-algebraically as

$$\left|\mathcal{E}_{\Omega^*\times\Omega^*}\left(\int_{\Omega^*}\mathrm{d}\mathbf{q} F_{\mathrm{d}}^{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{q}), \mathcal{K}\times\mathcal{K}\right)\right| \leqslant C_l N_{\mathbf{k}}^{-l}, \quad \forall l > 0.$$

Plugging the above two estimates into Eq. (33) proves that the quadrature error in the direct term calculation scales as

$$\left|\mathcal{E}_{\Omega^*\times\Omega^*\times\Omega^*}\left(\sum_{ijab}\langle i\mathbf{k}_i, j\mathbf{k}_j | a\mathbf{k}_a, b\mathbf{k}_b\rangle\, t_{ijab}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a), \mathcal{K}\times\mathcal{K}\times\mathcal{K}\right)\right| \leqslant C N_{\mathbf{k}}^{-1}.$$

Similar analysis can be applied to the exchange term where we formulate the integrand using two changes of variables $\mathbf{k}_j \to \mathbf{k}_a + \mathbf{q}_1$ and $\mathbf{k}_i \to \mathbf{k}_a - \mathbf{q}_2$ as

$$F_{\mathrm{x}}^{ijab}(\mathbf{k}_a, \mathbf{q}_1, \mathbf{q}_2) = \langle i(\mathbf{k}_a - \mathbf{q}_2), j(\mathbf{k}_a + \mathbf{q}_1) | b(\mathbf{k}_a - \mathbf{q}_2 + \mathbf{q}_1), a\mathbf{k}_a\rangle\, t_{ijab}(\mathbf{k}_a - \mathbf{q}_2, \mathbf{k}_a + \mathbf{q}_1, \mathbf{k}_a),$$

where the momentum transfers of the ERI and the amplitude are $\mathbf{q}_1$ and $\mathbf{q}_2$, respectively. The ERI and the amplitude are smooth everywhere with respect to $\mathbf{k}_a, \mathbf{q}_1, \mathbf{q}_2 \in \Omega^*$ except at $\mathbf{q}_1 = \mathbf{0}$ and $\mathbf{q}_2 = \mathbf{0}$, respectively, both with order 0. Similar to the discussion for the direct term, the exchange term calculation is equivalent to the trapezoidal rule over F_{x}^{ijab} using uniform mesh $\mathcal{K}\times\mathcal{K}_{\mathbf{q}}\times\mathcal{K}_{\mathbf{q}}$. The associated quadrature error with each set of (i, j, a, b) can be split as

$$\begin{aligned}&\mathcal{E}_{\Omega^*\times\Omega^*\times\Omega^*}\left(F_{\mathrm{x}}^{ijab}(\mathbf{k}_a, \mathbf{q}_1, \mathbf{q}_2), \mathcal{K}\times\mathcal{K}_{\mathbf{q}}\times\mathcal{K}_{\mathbf{q}}\right)\\ =&\mathcal{E}_{\Omega^*}\left(\int_{\Omega^*\times\Omega^*}\mathrm{d}\mathbf{q}_1\,\mathrm{d}\mathbf{q}_2 F_{\mathrm{x}}^{ijab}(\mathbf{k}_a, \mathbf{q}_1, \mathbf{q}_2), \mathcal{K}\right) + \frac{|\Omega^*|}{N_{\mathbf{k}}}\sum_{\mathbf{k}_a\in\mathcal{K}}\mathcal{E}_{\Omega^*\times\Omega^*}\left(F_{\mathrm{x}}^{ijab}(\mathbf{k}_a, \mathbf{q}_1, \mathbf{q}_2), \mathcal{K}_{\mathbf{q}}\times\mathcal{K}_{\mathbf{q}}\right).\end{aligned} \tag{34}$$

The first term decays super-algebraically since $\int_{\Omega^*\times\Omega^*} \mathrm{d}\mathbf{q}_1\,\mathrm{d}\mathbf{q}_2 F_{\mathrm{x}}^{ijab}(\mathbf{k}_a,\mathbf{q}_1,\mathbf{q}_2)$ is smooth and periodic with respect to $\mathbf{k}_a \in \Omega^*$ using the Leibniz integral rule. For the second term with each fixed $\mathbf{k}_a \in \mathcal{K}$, F_{x}^{ijab} can be viewed as a product of two periodic functions, $f_1(\mathbf{k}_a,\mathbf{q}_1,\mathbf{q}_2)f_2(\mathbf{k}_a,\mathbf{q}_1,\mathbf{q}_2)$, where f_1 is smooth everywhere except at $\mathbf{q}_1 = \mathbf{0}$ with order 0 and f_2 is smooth everywhere except at $\mathbf{q}_2 = \mathbf{0}$ with order 0. Lemma 18 provides quadrature error estimates for periodic functions in such a product form, and specifically in this case we have

$$\left|\mathcal{E}_{\Omega^*\times\Omega^*}\left(F_{\mathrm{x}}^{ijab}(\mathbf{k}_a,\mathbf{q}_1,\mathbf{q}_2), \mathcal{K}_{\mathbf{q}}\times\mathcal{K}_{\mathbf{q}}\right)\right| \leqslant CN_{\mathbf{k}}^{-1}, \quad \forall \mathbf{k}_a \in \mathcal{K},$$

where constant C can be proved independent of $\mathbf{k}_a$ using the algebraic singularity characterization of f_1 and f_2 and the prefactor estimate in Lemma 18 (see Remark 19).

Plugging these two estimates into Eq. (34) proves that the quadrature error in the exchange term calculation scales as

$$\left|\mathcal{E}_{\Omega^*\times\Omega^*\times\Omega^*}\left(\sum_{ijab}\langle i\mathbf{k}_i, j\mathbf{k}_j|b\mathbf{k}_b, a\mathbf{k}_a\rangle\, t_{ijab}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a), \mathcal{K}\times\mathcal{K}\times\mathcal{K}\right)\right| \leqslant CN_{\mathbf{k}}^{-1}.$$

Combining the above estimates for the direct and exchange terms together, we have

$$|\mathcal{G}_{\mathrm{TDL}}(t) - \mathcal{G}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}}t)| = \mathcal{O}(N_{\mathbf{k}}^{-1}), \quad \forall t \in \mathbb{T}(\Omega^*)^{n_{\mathrm{occ}}\times n_{\mathrm{occ}}\times n_{\mathrm{vir}}\times n_{\mathrm{vir}}},$$

which covers the case of the exact CCD(n) amplitudes with any $n > 0$.

B.3 Proof of Lemma 6: Amplitude error in a single iteration

Consider the error in the amplitude calculation using an arbitrary amplitude $t \in \mathbb{T}(\Omega^*)^{n_{\mathrm{occ}}\times n_{\mathrm{occ}}\times n_{\mathrm{vir}}\times n_{\mathrm{vir}}}$. Fixing a set of (i,j,a,b) and $\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_a \in \mathcal{K}$, the corresponding error entry can be detailed using the amplitude mapping definitions in Eq. (25) and Eq. (27) as

$$\begin{aligned}
&[\mathcal{M}_{\mathcal{K}}\mathcal{F}_{\mathrm{TDL}}(t) - \mathcal{F}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}}t)]_{ijab,\mathbf{k}_i\mathbf{k}_j\mathbf{k}_a} \\
=&\frac{1}{\varepsilon_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b}}\frac{1}{|\Omega^*|}\mathcal{E}_{\Omega^*}\left(\sum_{kl}\langle k\mathbf{k}_k, l\mathbf{k}_l|i\mathbf{k}_i, j\mathbf{k}_j\rangle\, t_{klab}(\mathbf{k}_k,\mathbf{k}_l,\mathbf{k}_a), \mathcal{K}\right) \\
&+ \frac{1}{\varepsilon_{i\mathbf{k}_i,j\mathbf{k}_j}^{a\mathbf{k}_a,b\mathbf{k}_b}}\frac{1}{|\Omega^*|^2}\mathcal{E}_{\Omega^*\times\Omega^*}\left(\sum_{klcd}\langle k\mathbf{k}_k, l\mathbf{k}_l|c\mathbf{k}_c, d\mathbf{k}_d\rangle\, t_{ijcd}(\mathbf{k}_i,\mathbf{k}_j,\mathbf{k}_c)t_{klab}(\mathbf{k}_k,\mathbf{k}_l,\mathbf{k}_a), \mathcal{K}\times\mathcal{K}\right) \\
&+ \cdots,
\end{aligned} \tag{35}$$

where the constant terms cancel each other and the listed two quadrature errors are the errors in the 4h2p linear and 4h2p quadratic term calculations. The neglected terms are the errors in remaining linear and quadratic terms calculations which can all be similarly formulated as quadrature errors of trapezoidal rules. The problem is thus reduced to the error estimate for trapezoidal rules applied to integrands defined by different amplitude terms in the amplitude equation.

As shown in Section B.1, the linear terms can be categorized into three classes listed in Table B.1 where the integrands respectively have zero, one, and two nonsmooth points. For terms in each class, their quadrature errors can be estimated similarly and we below demonstrate the error estimate for one example in each class.

For linear terms with zero nonsmooth point, we consider the 3h3p term $\langle AK|IC\rangle\, t_{KJ}^{CB}$ detailed as,

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} \langle a\mathbf{k}_a, k\mathbf{k}_k | i\mathbf{k}_i, c\mathbf{k}_c\rangle\, t_{kjcb}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_c).$$

The ERIs and the amplitudes have momentum transfers $\mathbf{k}_i - \mathbf{k}_a = -\mathbf{q}$ and $\mathbf{k}_c - \mathbf{k}_k = \mathbf{q}$, respectively, which are independent of $\mathbf{k}_k$. Therefore, for any $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \Omega^*$, the integrand is smooth and periodic with respect to $\mathbf{k}_k$ and thus has the quadatrure error decay super-algebraically according to Lemma 12, i.e.,

$$\left|\mathcal{E}_{\Omega^*}\left(\sum_{kc} \langle a\mathbf{k}_a, k\mathbf{k}_k | i\mathbf{k}_i, c\mathbf{k}_c\rangle\, t_{kjcb}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_c), \mathcal{K}\right)\right| \leqslant C_l N_{\mathbf{k}}^{-l/3}, \quad \forall l > 0. \tag{36}$$

where constant C_l can be shown independent of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \Omega^*$ using the prefactor estimate in Lemma 12 and the uniform boundedness of integrand derivatives over $\mathbf{k}_k$ for all $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ (see Remark 13).

For linear terms with one nonsmooth point, we consider the 3h3p term $\langle AK|CI\rangle\, t_{KJ}^{CB}$ detailed as

$$\int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kc} \langle a\mathbf{k}_a, k\mathbf{k}_k | c\mathbf{k}_c, i\mathbf{k}_i\rangle\, t_{kjcb}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_c),$$

where the ERIs and the amplitudes have momentum transfers $\mathbf{k}_k - \mathbf{k}_i$ and $\mathbf{k}_c - \mathbf{k}_k = \mathbf{q}$, respectively. For any $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \Omega^*$, these ERIs are smooth everywhere in Ω^* except at $\mathbf{k}_k = \mathbf{k}_i$ with order 0, -1, or -2 depending on the relation between (k, c) and (i, a). It can then be verified that the overall integrand is smooth everywhere except at $\mathbf{k}_k = \mathbf{k}_i$ with order -2 due to the product term with $k = i, c = a$. Lemma 14 provides the quadrature error estimate for such a periodic function that has algebraic singularity at one point, and specifically in this case we have

$$\left|\mathcal{E}_{\Omega^*}\left(\sum_{kc} \langle a\mathbf{k}_a, k\mathbf{k}_k | c\mathbf{k}_c, i\mathbf{k}_i\rangle\, t_{kjcb}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_c), \mathcal{K}\right)\right| \leqslant C N_{\mathbf{k}}^{-1/3}, \tag{37}$$

where constant C can be shown independent of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ using the prefactor estimate in Lemma 14 and the algebraic singularity characterization of the ERIs and the amplitudes (see Remark 15).

For linear terms with two nonsmooth points, we consider the 4h2p term $\langle KL|IJ\rangle\, t_{KL}^{AB}$ detailed in Eq. (35). First denote the integrand with each set of (k, l) as

$$F^{kl}(\mathbf{k}_k) = \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j\rangle\, t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a). \tag{38}$$

The ERI in $F^{kl}(\mathbf{k}_k)$ is smooth everywhere except at $\mathbf{k}_k = \mathbf{k}_i$ with order 0, -1, or -2 depending on the relation between (k, l) and (i, j). The amplitude in $F^{kl}(\mathbf{k}_k)$ is smooth everywhere except at $\mathbf{k}_k = \mathbf{k}_a$ with order 0. Applying the change of variable $\mathbf{k}_k \to \mathbf{k}_i - \mathbf{q}_1$, $F^{kl}(\mathbf{q}_1)$ can be formulated as the product of two periodic functions, $f_1(\mathbf{q}_1)f_2(\mathbf{q}_1)$, where $f_1(\mathbf{q}_1)$ is nonsmooth at $\mathbf{q}_1 = \mathbf{0}$ with order $0, -1$, or -2 and $f_2(\mathbf{q}_1)$ is nonsmooth at $\mathbf{q}_1 = \mathbf{k}_i - \mathbf{k}_a$ with order 0. Quadrature error of periodic functions in such a product form is estimated by Lemma 16 when $\mathbf{k}_i \neq \mathbf{k}_a \in \mathcal{K}$ and by Lemma 14 when $\mathbf{k}_i = \mathbf{k}_a \in \mathcal{K}$ as

$$\left|\mathcal{E}_{\Omega^*}\left(F^{kl}(\mathbf{q}_1), \mathcal{K}_{\mathbf{q}}\right)\right| \leqslant C \begin{cases} N_{\mathbf{k}}^{-1} & k \neq i, l \neq j \\ N_{\mathbf{k}}^{-\frac{2}{3}} & k = i, l \neq j \text{ or } k \neq i, l = j \\ N_{\mathbf{k}}^{-\frac{1}{3}} & k = i, l = j \end{cases}$$

where constant C can be shown independent of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ using the prefactor estimates in the two lemmas and the algebraic singularity characterization of the ERIs and the amplitudes (see Remark 15 and Remark 17). Summing over the above estimates for each set of (k, l), the quadrature error in the 4h2p linear term calculation can be estimated as

$$\left|\mathcal{E}_{\Omega^*}\left(\sum_{kl} \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j\rangle\, t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a), \mathcal{K}\right)\right| \leqslant C N_{\mathbf{k}}^{-\frac{1}{3}}.$$

Similar to the linear term case, all the quadratic terms and their quadrature error estimates can be categorized into four classes according to the nonsmoothness with respect to the two intermediate momentum vectors, as listed in Table B.2.

Table B.2. Classification of quadratic amplitude terms by the nonsmooth points with respect to the intermediate momentum vectors. We use $\mathbf{k}_1, \mathbf{k}_2$ to denote the two integration variables after proper change of variables over the intermediate momentum vectors and $\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3$ to denote generic points in Ω^*. The 3h3p terms permuted by $\mathcal{P}$ are of the same class as the unpermuted ones and thus not listed. The intermediate block underlying each product indicates the origin of this product in Eq. (27).

Singular Points	Quadratic terms
None	$\underbrace{\langle LK\|DC\rangle\, t_{IL}^{AD}}_{\chi_{IC}^{AK}} t_{KJ}^{CB}$
$\mathbf{k}_1 = \mathbf{x}_1$	$\underbrace{\langle LK\|CD\rangle\, t_{IL}^{AD}}_{\chi_{IC}^{AK}} t_{KJ}^{CB}, \underbrace{\langle LK\|DC\rangle\, t_{IL}^{DA}}_{\chi_{IC}^{AK}} t_{KJ}^{CB}, \underbrace{\langle LK\|DC\rangle\, t_{IL}^{AD}}_{\chi_{IC}^{AK}} t_{KJ}^{BC}\ \underbrace{\langle KL\|CD\rangle\, t_{KL}^{AD}}_{\kappa_{C}^{A}} t_{IJ}^{CB}, \underbrace{\langle KL\|CD\rangle\, t_{IL}^{CD}}_{\kappa_{I}^{K}} t_{KJ}^{AB}$
$\mathbf{k}_1 = \mathbf{x}_1, \mathbf{k}_2 = \mathbf{x}_2$	$\underbrace{\langle LK\|CD\rangle\, t_{IL}^{DA}}_{\chi_{CI}^{AK}} t_{KJ}^{CB}, \underbrace{\langle LK\|CD\rangle\, t_{IL}^{AD}}_{\chi_{IC}^{AK}} t_{KJ}^{BC}, \underbrace{\langle KL\|DC\rangle\, t_{KL}^{AD}}_{\kappa_{C}^{A}} t_{IJ}^{CB}, \underbrace{\langle KL\|DC\rangle\, t_{IL}^{CD}}_{\kappa_{I}^{K}} t_{KJ}^{AB}$
$\mathbf{k}_1 = \mathbf{x}_1, \mathbf{k}_2 = \mathbf{x}_2, \mathbf{k}_1 \pm \mathbf{k}_2 = \mathbf{x}_3$	$\underbrace{\langle KL\|CD\rangle\, t_{IJ}^{CD}}_{\chi_{IJ}^{AB}} t_{KL}^{AB}, \underbrace{\langle LK\|DC\rangle\, t_{IL}^{DA}}_{\chi_{IC}^{AK}} t_{KJ}^{BC}, \underbrace{\langle LK\|CD\rangle\, t_{JL}^{DA}}_{\chi_{CJ}^{AK}} t_{KI}^{BC}$

For quadratic terms of the first and second class, their quadrature errors can be estimated using Lemma 12 and Lemma 14 in a similar way as for the linear terms above. In the following, we demonstrate the quadrature error estimate for the third and forth classes of quadratic terms.

For the third class, we consider the 3h3p quadratic term $\langle LK|CD\rangle\, t_{IL}^{AD} t_{KJ}^{BC}$ and denote the integrand for each set of (k, l, c, d) as

$$F_{\text{3h3p}}^{klcd}(\mathbf{k}_k, \mathbf{k}_l) = \langle l\mathbf{k}_l, k\mathbf{k}_k | c\mathbf{k}_c, d\mathbf{k}_d\rangle\, t_{ilad}(\mathbf{k}_i, \mathbf{k}_l, \mathbf{k}_a) t_{kjbc}(\mathbf{k}_k, \mathbf{k}_j, \mathbf{k}_b),$$

where $\mathbf{k}_c = \mathbf{k}_k + \mathbf{k}_j - \mathbf{k}_b$ and $\mathbf{k}_d = \mathbf{k}_l + \mathbf{k}_i - \mathbf{k}_a$. The ERI and the two amplitudes have momentum transfers as $\mathbf{k}_c - \mathbf{k}_l = \mathbf{k}_k - \mathbf{k}_l + \mathbf{k}_j - \mathbf{k}_b$, $\mathbf{k}_a - \mathbf{k}_i$, and $\mathbf{k}_b - \mathbf{k}_k$, respectively. To single out the nonsmoothness with respect to $\mathbf{k}_k$ and $\mathbf{k}_l$, we introduce two changes of variables $\mathbf{k}_k \to \mathbf{k}_b - \mathbf{q}_2$ and $\mathbf{k}_l \to \mathbf{k}_k + \mathbf{k}_j - \mathbf{k}_b + \mathbf{q}_1$ and this term calculation can be formulated using the integrand periodicity as

$$\frac{1}{N_{\mathbf{k}}^2} \sum_{\mathbf{q}_1, \mathbf{q}_2 \in \mathcal{K}_{\mathbf{q}}} \sum_{klcd} F_{\text{3h3p}}^{klcd}(\mathbf{q}_1, \mathbf{q}_2) \longrightarrow \frac{1}{|\Omega^*|^2} \int_{\Omega^* \times \Omega^*} \mathrm{d}\mathbf{q}_1\, \mathrm{d}\mathbf{q}_2 \sum_{klcd} F_{\text{3h3p}}^{klcd}(\mathbf{q}_1, \mathbf{q}_2),$$

where the integrand is smooth everywhere except at $\mathbf{q}_1 = \mathbf{0}$ and $\mathbf{q}_2 = \mathbf{0}$. This explains the classification of this term as the third class listed in Table B.2. Note that the first amplitude in F_{3h3p}^{klcd} is smooth with respect to

$\mathbf{q}_1, \mathbf{q}_2$ and thus F_{3h3p}^{klcd} can be written in a product form $f_1(\mathbf{q}_1, \mathbf{q}_2) f_2(\mathbf{q}_1, \mathbf{q}_2)$ where $f_s(\mathbf{q}_1, \mathbf{q}_2)$ with $s = 1, 2$ is smooth everywhere except at $\mathbf{q}_s = \mathbf{0}$ with order 0. Lemma 18 provides the quadrature error estimate for bivariate functions in such a product form, and specifically in this case we have

$$\left| \mathcal{E}_{\Omega^* \times \Omega^*} \left(\sum_{klcd} F_{\text{3h3p}}^{klcd}(\mathbf{q}_1, \mathbf{q}_2), \mathcal{K}_{\mathbf{q}} \times \mathcal{K}_{\mathbf{q}} \right) \right| \leqslant C N_{\mathbf{k}}^{-1},$$

where constant C can be shown independent of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ using the prefactor estimate in Lemma 18 and the algebraic singularity characterization of ERIs and amplitudes (see Remark 19).

For the forth class, we consider the 4h2p quadratic term $\langle KL|CD\rangle\, t_{IJ}^{CD} t_{KL}^{AB}$ and denote the integrand with each set of (k, l, c, d) with the change of variable $\mathbf{k}_k \to \mathbf{k}_c + \mathbf{q}_1$ and $\mathbf{k}_c \to \mathbf{k}_i + \mathbf{q}_2$ as

$$F_{\text{4h2p}}^{klcd}(\mathbf{q}_1, \mathbf{q}_2) = \langle k\mathbf{k}_k, l\mathbf{k}_l | c\mathbf{k}_c, d\mathbf{k}_d\rangle\, t_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) t_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a),$$

where the ERI is smooth everywhere except at $\mathbf{q}_1 = \mathbf{0}$ with order 0, the two amplitudes are smooth everywhere except at $\mathbf{q}_2 = \mathbf{0}$ and $\mathbf{k}_a - \mathbf{k}_i = \mathbf{q}_2 - \mathbf{q}_1$, respectively, with order 0. This explains the classification of this term as the forth class listed in Table B.2. Lemma 20 provides the quadrature error estimate for bivariate functions in such a product form, and specifically in this case we have

$$\left| \mathcal{E}_{\Omega^* \times \Omega^*} \left(\sum_{klcd} F_{\text{4h2p}}^{klcd}(\mathbf{q}_1, \mathbf{q}_2), \mathcal{K}_{\mathbf{q}} \times \mathcal{K}_{\mathbf{q}} \right) \right| \leqslant C N_{\mathbf{k}}^{-1},$$

where constant C can be shown independent of $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$ using the prefactor estimate in Lemma 20 and the algebraic singularity characterization of ERIs and amplitudes (see Remark 21).

Collecting all the above quadrature error estimates for linear and quadratic terms (see Table V.2 for a summary), we obtain the final error estimate in amplitude calculation

$$\left| [\mathcal{M}_{\mathcal{K}} \mathcal{F}_{\text{TDL}}(t) - \mathcal{F}_{N_{\mathbf{k}}}(\mathcal{M}_{\mathcal{K}} t)]_{ijab, \mathbf{k}_i \mathbf{k}_j \mathbf{k}_a} \right| \leqslant C N_{\mathbf{k}}^{-\frac{1}{3}}, \qquad \forall i, j, a, b, \forall \mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a \in \mathcal{K}.$$

It is worth pointing out that the dominant error comes from the following six linear amplitude terms,

$$\sum_{KL} \langle KL|IJ\rangle\, t_{KL}^{AB}, \quad \sum_{CD} \langle AB|CD\rangle\, t_{IJ}^{CD}, \quad \mathcal{P} \sum_{KC} \langle AK|CJ\rangle\, t_{IK}^{CB}, \quad \mathcal{P} \sum_{KC} \langle AK|CI\rangle\, t_{KJ}^{BC},$$

where the involved ERIs can have matching band indices and thus are nonsmooth at zero momentum transfer points with order -2.

B.4 Proof of Lemma 7: Error accumulation in the CCD iteration

Fixing a set of i, j, a, b and $\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_a$, we focus on one constant, one linear, and one quadratic terms in the entry $[\mathcal{F}_{N_{\mathbf{k}}}(T)]_{ijab, \mathbf{k}_i \mathbf{k}_j \mathbf{k}_a}$ detailed as follows

$$\begin{aligned}
[\mathcal{F}_{N_{\mathbf{k}}}(T)]_{ijab, \mathbf{k}_i \mathbf{k}_j \mathbf{k}_a} &= \frac{1}{\varepsilon_{i\mathbf{k}_i, j\mathbf{k}_j}^{a\mathbf{k}_a, b\mathbf{k}_b}} \langle a\mathbf{k}_a, b\mathbf{k}_b | i\mathbf{k}_i, j\mathbf{k}_j\rangle \\
&+ \frac{1}{\varepsilon_{i\mathbf{k}_i, j\mathbf{k}_j}^{a\mathbf{k}_a, b\mathbf{k}_b}} \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{k}_k \in \mathcal{K}} \sum_{kl} \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j\rangle\, T_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) \\
&+ \frac{1}{\varepsilon_{i\mathbf{k}_i, j\mathbf{k}_j}^{a\mathbf{k}_a, b\mathbf{k}_b}} \frac{1}{N_{\mathbf{k}}^2} \sum_{\mathbf{k}_k \mathbf{k}_c \in \mathcal{K}} \sum_{klcd} \langle k\mathbf{k}_k, l\mathbf{k}_l | c\mathbf{k}_c, d\mathbf{k}_d\rangle\, T_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) T_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) \\
&+ \cdots,
\end{aligned} \tag{39}$$

where the linear and quadratic terms come from the 4h2p term $\chi_{IJ}^{KL} t_{KL}^{AB}$ in Eq. (25) and the neglected terms above are the other linear and quadratic terms included in Eq. (25).

In the subtraction $\mathcal{F}_{N_\mathbf{k}}(T) - \mathcal{F}_{N_\mathbf{k}}(S)$, the constant terms above in these two maps cancel each other. The subtraction between the two 4h2p linear terms can be formulated and bounded as

$$\begin{aligned}
&\left| \frac{1}{\varepsilon_{i\mathbf{k}_i, j\mathbf{k}_j}^{a\mathbf{k}_a, b\mathbf{k}_b}} \frac{1}{N_\mathbf{k}} \sum_{\mathbf{k}_k \in \mathcal{K}} \sum_{kl} \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j \rangle \left(T_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) - S_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) \right) \right| \\
\leqslant & C \frac{1}{N_\mathbf{k}} \sum_{\mathbf{k}_k \in \mathcal{K}} \sum_{kl} \left| \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j \rangle \right| \|T - S\|_\infty \\
\leqslant & C \|T - S\|_\infty \int_{\Omega^*} \mathrm{d}\mathbf{k}_k \sum_{kl} \left| \langle k\mathbf{k}_k, l\mathbf{k}_l | i\mathbf{k}_i, j\mathbf{k}_j \rangle \right| \leqslant C \|T - S\|_\infty.
\end{aligned}$$

Similar estimate can be obtained for all the other linear terms in the amplitude map $\mathcal{F}_{N_\mathbf{k}}$. The subtraction between the two 4h2p quadratic terms can be formulated and bounded as

$$\begin{aligned}
&\left| \frac{1}{\varepsilon_{i\mathbf{k}_i, j\mathbf{k}_j}^{a\mathbf{k}_a, b\mathbf{k}_b}} \frac{1}{N_\mathbf{k}^2} \sum_{\mathbf{k}_k \mathbf{k}_c \in \mathcal{K}} \sum_{klcd} \langle k\mathbf{k}_k, l\mathbf{k}_l | c\mathbf{k}_c, d\mathbf{k}_d \rangle \left(T_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) T_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) - S_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) S_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) \right) \right| \\
\leqslant & C \int_{\Omega^* \times \Omega^*} \mathrm{d}\mathbf{k}_k \, \mathrm{d}\mathbf{k}_c \sum_{klcd} \left| \langle k\mathbf{k}_k, l\mathbf{k}_l | c\mathbf{k}_c, d\mathbf{k}_d \rangle \right| \left(\|T\|_\infty + \|S\|_\infty \right) \|T - S\|_\infty \\
\leqslant & C \left(\|T\|_\infty + \|S\|_\infty \right) \|T - S\|_\infty,
\end{aligned}$$

where the first inequality uses the estimate

$$\begin{aligned}
&\left| T_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) T_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) - S_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) S_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) \right| \\
\leqslant & \left| T_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) \left(T_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) - S_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) \right) \right| + \left| \left(T_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) - S_{ijcd}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_c) \right) S_{klab}(\mathbf{k}_k, \mathbf{k}_l, \mathbf{k}_a) \right| \\
\leqslant & \|T\|_\infty \|T - S\|_\infty + \|S\|_\infty \|T - S\|_\infty.
\end{aligned}$$

Similar estimate can be obtained for all the other quadratic terms in the amplitude map $\mathcal{F}_{N_\mathbf{k}}$. Collecting all these estimates together, we have

$$\begin{aligned}
\|\mathcal{F}_{N_\mathbf{k}}(T) - \mathcal{F}_{N_\mathbf{k}}(S)\|_\infty = & \max_{ijab, \mathbf{k}_i \mathbf{k}_j \mathbf{k}_a \in \mathcal{K}} \left| \left[\mathcal{F}_{N_\mathbf{k}}(T) - \mathcal{F}_{N_\mathbf{k}}(S) \right]_{ijab, \mathbf{k}_i \mathbf{k}_j \mathbf{k}_a} \right| \\
& \leqslant C \left(1 + \|T\|_\infty + \|S\|_\infty \right) \|T - S\|_\infty.
\end{aligned}$$

Appendix C PROOF OF COROLLARY 3

As discussed in Section VII, it is possible in general that the CCD amplitude equation may have multiple solutions or its fixed point iteration may diverge. In these cases, the finite size error in CCD energy calculation can be ill-defined and not connected to CCD(n) we have analyzed. Here, we consider the ideal case where $T = \mathcal{F}_{N_\mathbf{k}}(T)$ for any sufficiently large $N_\mathbf{k}$ and $t = \mathcal{F}_{\text{TDL}}(t)$ both have unique solutions, denoted as $T_*^{N_\mathbf{k}}$ and t_*, and the corresponding fixed point iterations converge in the sense of the $\|\cdot\|_\infty$-norm, i.e.,

$$\lim_{n \to \infty} \|T_n - T_*^{N_\mathbf{k}}\|_\infty = 0 \qquad \text{and} \qquad \lim_{n \to \infty} \|t_n - t_*\|_\infty = 0.$$

In general, a common sufficient condition that guarantees the convergence of a fixed point iteration is that the target mapping is contractive (to be specified later) in a domain that contains the solution point and the initial guess also lies in this domain. Following this practice, we make four assumptions:

- $\mathcal{F}_{\text{TDL}}$ is a contraction map in a domain $\mathbb{B}_{\text{TDL}} \subset \mathbb{T}(\Omega^*)^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}}$ that contains t_* and the initial guess $\mathbf{0}$, i.e.,

$$\begin{aligned}&\mathcal{F}_{\text{TDL}}(t) \in \mathbb{B}_{\text{TDL}}, \quad \forall t \in \mathbb{B}_{\text{TDL}},\\ &\|\mathcal{F}_{\text{TDL}}(t) - \mathcal{F}_{\text{TDL}}(s)\|_\infty \leqslant L\|t-s\|_\infty, \quad \forall t,s \in \mathbb{B}_{\text{TDL}},\end{aligned}$$

with a constant $L<1$. This assumption guarantees that $\{t_n\}$ lies in $\mathbb{B}_{\text{TDL}}$ and converges to t_*.

- $\mathcal{F}_{N_\mathbf{k}}$ with sufficiently large $N_\mathbf{k}$ is a contraction map in a domain $\mathbb{B}_{N_\mathbf{k}} \subset \mathbb{C}^{n_{\text{occ}}\times n_{\text{occ}}\times n_{\text{vir}}\times n_{\text{vir}}\times N_\mathbf{k}\times N_\mathbf{k}\times N_\mathbf{k}}$ that contains $T_*^{N_\mathbf{k}}$ and the initial guess $\mathbf{0}$, i.e.,

$$\begin{aligned}&\mathcal{F}_{N_\mathbf{k}}(T) \in \mathbb{B}_{N_\mathbf{k}}, \quad \forall T \in \mathbb{B}_{N_\mathbf{k}},\\ &\|\mathcal{F}_{N_\mathbf{k}}(T) - \mathcal{F}_{N_\mathbf{k}}(S)\|_\infty \leqslant L\|T-S\|_\infty, \quad \forall T,S \in \mathbb{B}_{N_\mathbf{k}},\end{aligned}$$

with a constant $L<1$. This assumption guarantees that $\{T_n^{N_\mathbf{k}}\}$ lies in $\mathbb{B}_{N_\mathbf{k}}$ and converges to $T_*^{N_\mathbf{k}}$.

- For sufficiently large $N_\mathbf{k}$, the domains in the above two assumptions satisfy that

$$\mathcal{M}_\mathcal{K}\mathbb{B}_{\text{TDL}} := \{\mathcal{M}_\mathcal{K} t : t \in \mathbb{B}_{\text{TDL}}\} \subset \mathbb{B}_{N_\mathbf{k}}. \tag{40}$$

  Theorem 1 proves that for each fixed n the finite-size amplitude $T_n^{N_\mathbf{k}}$ converges to t_n in the sense of

$$\lim_{N_\mathbf{k}\to\infty} \|\mathcal{M}_\mathcal{K} t_n - T_n^{N_\mathbf{k}}\|_\infty = 0,$$

  showing that $\{T_n^{N_\mathbf{k}}\} \subset \mathbb{B}_{N_\mathbf{k}}$ converges to $\{t_n\} \subset \mathbb{B}_{\text{TDL}}$ with $\mathcal{K} \to \Omega^*$. Intuitively, this argument suggests certain closeness between $\mathbb{B}_{N_\mathbf{k}}$ and $\mathcal{M}_\mathcal{K}\mathbb{B}_{\text{TDL}}$ which leads to the assumption here.

- Note that the error estimate in Lemma 6 has prefactor C dependent on the amplitude t. For the whole set of iterates $\{t_n\}$, we make a stronger assumption that there exists a constant C such that

$$\|\mathcal{M}_\mathcal{K}\mathcal{F}_{\text{TDL}}(t_n) - \mathcal{F}_{N_\mathbf{k}}(\mathcal{M}_\mathcal{K} t_n)\|_\infty \leqslant C N_\mathbf{k}^{-\frac{1}{3}}, \quad \forall n > 0. \tag{41}$$

Under these assumptions, the finite-size error in the CCD energy calculation can be estimated as

$$\begin{aligned}\left|E_{\text{CCD}}^{\text{TDL}} - E_{\text{CCD}}^{N_\mathbf{k}}\right| &= \left|\mathcal{G}_{\text{TDL}}(t_*) - \mathcal{G}_{N_\mathbf{k}}(T_*^{N_\mathbf{k}})\right|\\ &\leqslant \left|\mathcal{G}_{N_\mathbf{k}}(\mathcal{M}_\mathcal{K} t_*) - \mathcal{G}_{N_\mathbf{k}}(T_*^{N_\mathbf{k}})\right| + \left|\mathcal{G}_{\text{TDL}}(t_*) - \mathcal{G}_{N_\mathbf{k}}(\mathcal{M}_\mathcal{K} t_*)\right|\\ &\leqslant C\left\|\mathcal{M}_\mathcal{K} t_* - T_*^{N_\mathbf{k}}\right\|_\infty + C N_\mathbf{k}^{-1},\end{aligned} \tag{42}$$

where the last inequality uses the boundedness of linear operator $\mathcal{G}_{N_\mathbf{k}}$ and Lemma 5. To estimate the finite-size error in the converged amplitudes, $\left\|\mathcal{M}_\mathcal{K} t_* - T_*^{N_\mathbf{k}}\right\|_\infty$, we consider the error splitting Eq. (13) for the amplitude calculation at the n-th fixed point iteration as

$$\begin{aligned}\left\|\mathcal{M}_\mathcal{K} t_n - T_n^{N_\mathbf{k}}\right\|_\infty &\leqslant \|\mathcal{M}_\mathcal{K}\mathcal{F}_{\text{TDL}}(t_{n-1}) - \mathcal{F}_{N_\mathbf{k}}(\mathcal{M}_\mathcal{K} t_{n-1})\|_\infty + \left\|\mathcal{F}_{N_\mathbf{k}}(T_{n-1}^{N_\mathbf{k}}) - \mathcal{F}_{N_\mathbf{k}}(\mathcal{M}_\mathcal{K} t_{n-1})\right\|_\infty\\ &\leqslant C N_\mathbf{k}^{-\frac{1}{3}} + L\left\|\mathcal{M}_\mathcal{K} t_{n-1} - T_{n-1}^{N_\mathbf{k}}\right\|_\infty,\end{aligned}$$

where the last estimate uses the assumption in Eq. (41) for the first term and the assumptions that $\mathcal{F}_{N_{\mathbf{k}}}$ is a contraction map and $\mathcal{M}_{\mathcal{K}}t_{n-1} \in \mathbb{B}_{N_{\mathbf{k}}}$ for the second term. Since the initial guesses in the finite and the TDL cases satisfy $\|\mathcal{M}_{\mathcal{K}}t_0 - T_0^{N_{\mathbf{k}}}\|_\infty = 0$, we can recursively derive that

$$\left\|\mathcal{M}_{\mathcal{K}}t_n - T_n^{N_{\mathbf{k}}}\right\|_\infty \leqslant C\frac{1-L^n}{1-L}N_{\mathbf{k}}^{-\frac{1}{3}},$$

and thus

$$\left\|\mathcal{M}_{\mathcal{K}}t_* - T_*^{N_{\mathbf{k}}}\right\|_\infty = \lim_{n\to\infty}\left\|\mathcal{M}_{\mathcal{K}}t_n - T_n^{N_{\mathbf{k}}}\right\|_\infty \leqslant CN_{\mathbf{k}}^{-\frac{1}{3}}.$$

Plugging this estimate into Eq. (42) then finishes the proof.

Appendix D QUADRATURE ERROR ESTIMATE FOR PERIODIC FUNCTION WITH ALGEBRAIC SINGULARITY

This section consists of five lemmas that provide the quadrature error estimates for trapezoidal rules over integrands in five different classes as listed in Table V.1. All the integrands are either smooth or built by univariate/multivariate functions that have algebraic singularity at one single point. In addition to the asymptotic scaling of the quadrature errors, our finite-size error analysis also needs quantitative descriptions about the relation between prefactors in the estimate and the smoothness properties of the integrand.

For a univariate function $f(\mathbf{x})$ that is smooth everywhere in V except at $\mathbf{x} = \mathbf{x}_0$ with algebraic singularity of order γ, we define a constant

$$\begin{aligned}\mathcal{H}^l_{V,\mathbf{x}_0}(f) &= \min\left\{C : |\partial^{\boldsymbol{\alpha}}_{\mathbf{x}} f(\mathbf{x})| \leqslant C|\mathbf{x}-\mathbf{x}_0|^{\gamma-|\boldsymbol{\alpha}|}, \forall|\boldsymbol{\alpha}| \leqslant l, \forall \mathbf{x}\in V\setminus\{\mathbf{x}_0\}\right\}\\ &= \max_{|\boldsymbol{\alpha}|\leqslant l}\left\|\left(\partial^{\boldsymbol{\alpha}}_{\mathbf{x}} f(\mathbf{x})\right)/|\mathbf{x}-\mathbf{x}_0|^{\gamma-|\boldsymbol{\alpha}|}\right\|_{L^\infty(V)}.\end{aligned} \tag{43}$$

For a multivariate function $f(\mathbf{x},\mathbf{y})$ that is smooth everywhere in $V_X \times V_Y$ except at $\mathbf{x} = \mathbf{x}_0$ with algebraic singularity of order γ, we define a constant

$$\begin{aligned}\mathcal{H}^l_{V_X\times V_Y,(\mathbf{x}_0,\cdot)}(f) &= \min\left\{C : \left|\partial^{\boldsymbol{\alpha}}_{\mathbf{x}}\partial^{\boldsymbol{\beta}}_{\mathbf{y}} f(\mathbf{x},\mathbf{y})\right| \leqslant C|\mathbf{x}-\mathbf{x}_0|^{\gamma-|\boldsymbol{\alpha}|}, \forall|\boldsymbol{\alpha}|,|\boldsymbol{\beta}| \leqslant l, \forall \mathbf{x}\in V_X\setminus\{\mathbf{x}_0\}, \mathbf{y}\in V_Y\right\}\\ &= \max_{|\boldsymbol{\alpha}|\leqslant l}\left\|\left(\partial^{\boldsymbol{\alpha}}_{\mathbf{x}}\partial^{\boldsymbol{\beta}}_{\mathbf{y}} f(\mathbf{x},\mathbf{y})\right)/|\mathbf{x}-\mathbf{x}_0|^{\gamma-|\boldsymbol{\alpha}|}\right\|_{L^\infty(V\times V)},\end{aligned} \tag{44}$$

where "$\cdot$" in the subscript "$(\mathbf{x}_0,\cdot)$" is a placeholder to indicate the smooth variable. Using these two quantities, we have following function estimates that will be extensively used in this section

$$|\partial^{\boldsymbol{\alpha}}_{\mathbf{x}} f(\mathbf{x})| \leqslant \mathcal{H}^l_{V,\mathbf{x}_0}(f)|\mathbf{x}-\mathbf{x}_0|^{\gamma-|\boldsymbol{\alpha}|}, \qquad \forall l \geqslant |\boldsymbol{\alpha}|,\ \forall \mathbf{x}\in V\setminus\{\mathbf{x}_0\}, \tag{45}$$

$$\left|\partial^{\boldsymbol{\alpha}}_{\mathbf{x}}\partial^{\boldsymbol{\beta}}_{\mathbf{y}} f(\mathbf{x},\mathbf{y})\right| \leqslant \mathcal{H}^l_{V_X\times V_Y,(\mathbf{x}_0,\cdot)}(f)|\mathbf{x}-\mathbf{x}_0|^{\gamma-|\boldsymbol{\alpha}|}, \qquad \forall l \geqslant |\boldsymbol{\alpha}|,|\boldsymbol{\beta}|,\ \forall \mathbf{x}\in V_X\setminus\{\mathbf{x}_0\}, \mathbf{y}\in V_Y. \tag{46}$$

The following lemma is the standard result that the quadrature error of a trapezoidal rule applied to smooth and periodic functions decays super-algebraically. For completeness, we include a proof using the Poisson summation formula.

Lemma 12. *Let $f(\mathbf{x})$ be smooth and periodic in $V = [-\frac{1}{2},\frac{1}{2}]^d$. The quadrature error of a trapezoidal rule using an m^d-sized uniform mesh $\mathcal{X}$ in V decays super-algebraically as*

$$|\mathcal{E}_V(f,\mathcal{X})| \leqslant C_l m^{-l}, \quad \forall l > 0.$$

Proof. Denote the Fourier transform of $f(\mathbf{x})$ as

$$f(\mathbf{x}) = \frac{1}{|V|}\sum_{\mathbf{k}\in 2\pi\mathbb{Z}^d} \hat{f}(\mathbf{k})e^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}} \quad \text{with} \quad \hat{f}(\mathbf{k}) = \int_V f(\mathbf{x})e^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}}\,\mathrm{d}\mathbf{x}.$$

Let $\mathcal{X}_0$ be the m^d-sized uniform mesh in V that contains $\mathbf{x} = \mathbf{0}$ and let $\mathcal{X} = \mathcal{X}_0 + \mathbf{x}_0$. Denote the unit length $h = 1/m$. Using these notations, we have

$$\begin{aligned}
&\mathcal{I}_V(f) = \hat{f}(\mathbf{0}),\\
&\mathcal{Q}_V(f,\mathcal{X}) = \frac{|V|}{m^d}\sum_{\mathbf{x}\in\mathcal{X}_0} f(\mathbf{x}+\mathbf{x}_0) = \sum_{\mathbf{k}\in 2\pi\mathbb{Z}^d}\hat{f}(\mathbf{k})e^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}_0}\frac{1}{m^d}\sum_{\mathbf{x}\in\mathcal{X}_0} e^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}} = \sum_{\mathbf{k}\in 2\pi\mathbb{Z}^d}\hat{f}\left(\frac{\mathbf{k}}{h}\right)e^{\mathrm{i}\frac{\mathbf{k}}{h}\cdot\mathbf{x}_0},
\end{aligned}$$

and thus the quadrature error of the trapezoidal rule using $\mathcal{X}$ can be estimated as

$$|\mathcal{E}_V(f,\mathcal{X})| \leqslant \sum_{\mathbf{k}\in 2\pi\mathbb{Z}^d\setminus\{\mathbf{0}\}}\left|\hat{f}\left(\frac{\mathbf{k}}{h}\right)\right|. \tag{47}$$

Based on the periodicity and smoothness of $f(\mathbf{x})$ in V, we can use integration by part to estimate the Fourier transform coefficient as

$$\begin{aligned}
\left|\hat{f}\left(\frac{\mathbf{k}}{h}\right)\right| &= \left|\int_V \mathrm{d}\mathbf{x} f(\mathbf{x})e^{-\mathrm{i}\frac{\mathbf{k}}{h}\cdot\mathbf{x}}\right|\\
&= \frac{h^{|\boldsymbol{\alpha}|}}{|\mathbf{k}|^{\boldsymbol{\alpha}}}\left|\int_V \mathrm{d}\mathbf{x}\frac{\partial^{\boldsymbol{\alpha}}}{\partial\mathbf{x}^{\boldsymbol{\alpha}}}f(\mathbf{x})e^{-\mathrm{i}\frac{\mathbf{k}}{h}\cdot\mathbf{x}}\right|\\
&\leqslant \frac{h^{|\boldsymbol{\alpha}|}}{|\mathbf{k}|^{\boldsymbol{\alpha}}}\int_V \mathrm{d}\mathbf{x}\left|\frac{\partial^{\boldsymbol{\alpha}}}{\partial\mathbf{x}^{\boldsymbol{\alpha}}}f(\mathbf{x})\right| = C_{\boldsymbol{\alpha}}\frac{h^{|\boldsymbol{\alpha}|}}{|\mathbf{k}|^{\boldsymbol{\alpha}}},
\end{aligned} \tag{48}$$

with any derivative order $\boldsymbol{\alpha} \geqslant 0$ where $|\boldsymbol{\alpha}| = \sum_i \boldsymbol{\alpha}_i$ and $|\mathbf{k}|^{\boldsymbol{\alpha}} = \prod_i |\mathbf{k}_i|^{\boldsymbol{\alpha}_i}$. Plugging this estimate into Eq. (47), we obtain

$$|\mathcal{E}_V(f,\mathcal{X})| \leqslant C_{\boldsymbol{\alpha}}h^{|\boldsymbol{\alpha}|}\sum_{\mathbf{k}\in 2\pi\mathbb{Z}^d\setminus\{\mathbf{0}\}}\frac{1}{|\mathbf{k}|^{\boldsymbol{\alpha}}},$$

which then proves the lemma by choosing an arbitrary $\boldsymbol{\alpha}$ with $|\boldsymbol{\alpha}| = l > d$. ☐

Remark 13. *If we replace $f(\mathbf{x})$ by $f(\mathbf{x},\mathbf{y})$ defined in $V\times V_Y$ which is smooth and periodic with respect to $\mathbf{x}$ for each $\mathbf{y}\in V_Y$ and satisfies $\sup_{\mathbf{x}\in V,\mathbf{y}\in V_Y}|\partial_{\mathbf{x}}^{\boldsymbol{\alpha}} f(\mathbf{x},\mathbf{y})| < \infty$ for any $\boldsymbol{\alpha}\geqslant 0$, Lemma 12 can be generalized as*

$$|\mathcal{E}_V(f(\cdot,\mathbf{y}),\mathcal{X})| \leqslant C_l m^{-l}, \quad \forall l > 0, \forall \mathbf{y}\in V_Y,$$

where constant C_l is independent of $\mathbf{y}\in V_Y$ based on the prefactor estimate in Eq. (48).

Lemma 14. *Let $f(\mathbf{x})$ be periodic with respect to $V = [-\frac{1}{2},\frac{1}{2}]^d$ and smooth everywhere except at $\mathbf{x} = \mathbf{0}$ with order $\gamma \geqslant -d+1$. At $\mathbf{x}=\mathbf{0}$, $f(\mathbf{x})$ is set to 0. The quadrature error of a trapezoidal rule using an m^d-sized uniform mesh $\mathcal{X}$ that contains $\mathbf{x}=\mathbf{0}$ can be estimated as*

$$|\mathcal{E}_V(f,\mathcal{X})| \leqslant C\mathcal{H}_{V,\mathbf{0}}^{d+\max(1,\gamma)}(f)m^{-(d+\gamma)}.$$

If $f(\mathbf{0})$ is set to an $\mathcal{O}(1)$ value in the calculation, it introduces additional $\mathcal{O}(m^{-d})$ quadrature error.

Proof. Define a cutoff function $\psi \in \mathbb{C}_c^\infty(\mathbb{R}^n)$ satisfying

$$\psi(\mathbf{x}) = \begin{cases} 1, & |\mathbf{x}| \leqslant \frac{1}{2} \\ 0, & |\mathbf{x}| \geqslant 1 \end{cases},$$

and denote its scaling as $\psi_L(\mathbf{x}) = \psi(\mathbf{x}/L)$ that is compactly supported in $|\mathbf{x}| \leqslant L$. Let $h = 1/m$ be the unit length of the uniform mesh $\mathcal{X}$. Using the cutoff function, we split $f(\mathbf{x})$ as

$$f(\mathbf{x}) = f(\mathbf{x})\psi_{\frac{1}{2}}(\mathbf{x}) + f(\mathbf{x})(1 - \psi_{\frac{1}{2}}(\mathbf{x})),$$

where the first term is compactly supported in V and the second term is smooth in V and satisfies the periodic boundary condition on ∂V. Accordingly, the quadrature error can be split into

$$\mathcal{E}_V(f, \mathcal{X}) = \mathcal{E}_V(f\psi_{\frac{1}{2}}, \mathcal{X}) + \mathcal{E}_V(f(1 - \psi_{\frac{1}{2}}), \mathcal{X}),$$

where the second error decays super-algebraically with respect to m according to Lemma 12. The problem is reduced to estimating the first quadrature error for a localized integrand $f\psi_{\frac{1}{2}}$. In the following discussion, we abuse the notation f to denote the localized function $f\psi_{\frac{1}{2}}$ and assume that f is compactly supported in V and smooth everywhere except at $\mathbf{0}$ with order γ.

Since $f(\mathbf{0}) = 0$, the trapezoidal rule over $f(\mathbf{x})$ using $\mathcal{X}$ satisfies

$$\mathcal{Q}_V(f, \mathcal{X}) = \mathcal{Q}_V(f(1 - \psi_h), \mathcal{X}),$$

and accordingly its quadrature error can be split as

$$\begin{aligned} \mathcal{E}_V(f, \mathcal{X}) &= \mathcal{I}_V(f\psi_h) + \mathcal{I}_V(f(1 - \psi_h)) - \mathcal{Q}_V(f(1 - \psi_h), \mathcal{X}) \\ &= \mathcal{I}_V(f\psi_h) + \mathcal{E}_V(f(1 - \psi_h), \mathcal{X}). \end{aligned} \tag{49}$$

The first part in Eq. (49) can be estimated as

$$|\mathcal{I}_V(f\psi_h)| \leqslant C \int_{|\mathbf{x}| \leqslant h} |f(\mathbf{x})| \, \mathrm{d}\mathbf{x} \leqslant C\mathcal{H}^0_{V,\mathbf{0}}(f) \int_{|\mathbf{x}| \leqslant h} |\mathbf{x}|^\gamma \, \mathrm{d}\mathbf{x} \leqslant C\mathcal{H}^0_{V,\mathbf{0}}(f) h^{d+\gamma}, \tag{50}$$

using the algebraic singularity characterization Eq. (45) for $f(\mathbf{x})$ at $\mathbf{x} = \mathbf{0}$. The second part in Eq. (49) can be reformulated using the Poisson summation formula as

$$\mathcal{E}_V\left(f(1 - \psi_h), \mathcal{X}\right) = -\sum_{\mathbf{k} \in 2\pi\mathbb{Z}^d \setminus \{\mathbf{0}\}} \hat{f}_{\psi,h}\left(\frac{\mathbf{k}}{h}\right), \tag{51}$$

where $f_{\psi,h} = f(1 - \psi_h)$ and its Fourier transform can be estimated as

$$\begin{aligned} \left|\hat{f}_{\psi,h}\left(\frac{\mathbf{k}}{h}\right)\right| &= \frac{h^{|\boldsymbol{\alpha}|}}{|\mathbf{k}|^{\boldsymbol{\alpha}}} \left| \int_{\mathbb{R}^d} \mathrm{d}\mathbf{x} \frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}^{\boldsymbol{\alpha}}} f(\mathbf{x})(1 - \psi_h(\mathbf{x})) e^{-\mathrm{i}\frac{\mathbf{k}}{h}\cdot\mathbf{x}} \right| \\ &\leqslant \frac{h^{|\boldsymbol{\alpha}|}}{|\mathbf{k}|^{\boldsymbol{\alpha}}} \int_{\mathbb{R}^d} \mathrm{d}\mathbf{x} \left| \frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}^{\boldsymbol{\alpha}}} f(\mathbf{x})(1 - \psi_h(\mathbf{x})) \right|, \end{aligned}$$

with any derivative order $\boldsymbol{\alpha} \geqslant 0$. The derivative in the last integral can be further expanded as

$$\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}^{\boldsymbol{\alpha}}} f(\mathbf{x})(1 - \psi_h(\mathbf{x})) = \sum_{\boldsymbol{\alpha}_1 + \boldsymbol{\alpha}_2 = \boldsymbol{\alpha}} \binom{\boldsymbol{\alpha}}{\boldsymbol{\alpha}_1} \frac{\partial^{\boldsymbol{\alpha}_1}}{\partial \mathbf{x}^{\boldsymbol{\alpha}_1}} (1 - \psi_h(\mathbf{x})) \frac{\partial^{\boldsymbol{\alpha}_2}}{\partial \mathbf{x}^{\boldsymbol{\alpha}_2}} f(\mathbf{x}).$$

Using the locality of $1-\psi_h(\mathbf{x})$ and $f(\mathbf{x})$ and the inequality $|\partial_{\mathbf{x}}^{\boldsymbol{\beta}} f(\mathbf{x})| \leqslant \mathcal{H}_{V,\mathbf{0}}^{|\boldsymbol{\alpha}|}(f)|\mathbf{x}|^{\gamma-|\boldsymbol{\beta}|}$ with any $\boldsymbol{\beta} \leqslant \boldsymbol{\alpha}$, we can estimate this derivative as

$$\left|\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}^{\boldsymbol{\alpha}}} f(\mathbf{x})(1-\psi_h(\mathbf{x}))\right| \leqslant C\mathcal{H}_{V,\mathbf{0}}^{|\boldsymbol{\alpha}|}(f) \begin{cases} 0, & |\mathbf{x}| \leqslant \frac{1}{2}h \\ h^{\gamma-|\boldsymbol{\alpha}|}, & \frac{1}{2}h \leqslant |\mathbf{x}| \leqslant h \\ |\mathbf{x}|^{\gamma-|\boldsymbol{\alpha}|}, & h \leqslant |\mathbf{x}| \leqslant \frac{1}{2} \\ 0, & |\mathbf{x}| > \frac{1}{2} \end{cases}.$$

Using this estimate, the associated integral can be bounded as

$$\begin{aligned} \int_{\mathbb{R}^d} \mathrm{d}\mathbf{x} \left|\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}^{\boldsymbol{\alpha}}} f(\mathbf{x})(1-\psi_h(\mathbf{x}))\right| &\leqslant C\mathcal{H}_{V,\mathbf{0}}^{|\boldsymbol{\alpha}|}(f)\left(\int_{\frac{1}{2}h}^{h} h^{\gamma-|\boldsymbol{\alpha}|} r^{d-1}\,\mathrm{d}r + \int_h^{\frac{1}{2}} r^{\gamma-|\boldsymbol{\alpha}|+d-1}\,\mathrm{d}r\right) \\ &\leqslant C\mathcal{H}_{V,\mathbf{0}}^{|\boldsymbol{\alpha}|}(f)(h^{\gamma+d-|\boldsymbol{\alpha}|}+1). \end{aligned}$$

Plugging this estimate into Eq. (51), we obtain

$$|\mathcal{E}_V(f(1-\psi_h),\mathcal{X})| \leqslant C\mathcal{H}_{V,\mathbf{0}}^{|\boldsymbol{\alpha}|}(f) \sum_{\mathbf{k}\in 2\pi\mathbb{Z}^d\setminus\{\mathbf{0}\}} \frac{1}{|\mathbf{k}|^{\boldsymbol{\alpha}}}\left(h^{\gamma+d}+h^{|\boldsymbol{\alpha}|}\right).$$

Choosing an arbitrary $\boldsymbol{\alpha}$ with $|\boldsymbol{\alpha}| = \max(d+1, d+\gamma)$, we obtain

$$|\mathcal{E}_V(f(1-\psi_h),\mathcal{X})| \leqslant C\mathcal{H}_{V,\mathbf{0}}^{|\boldsymbol{\alpha}|}(f)h^{\gamma+d},$$

which together with Eq. (49) and Eq. (50) proves the lemma. □

Remark 15. *If we replace $f(\mathbf{x})$ by $f(\mathbf{x},\mathbf{y})$ defined in $V\times V_Y$ which is smooth everywhere in $V\times V_Y$ except at $\mathbf{x}=\mathbf{0}$ with order γ, Lemma 14 can be generalized to*

$$|\mathcal{E}_V(f(\cdot,\mathbf{y}),\mathcal{X})| \leqslant C\mathcal{H}_{V\times V_Y,(\mathbf{0},\cdot)}^{d+\max(1,\gamma)}(f)m^{-(d+\gamma)}, \quad \forall \mathbf{y}\in V_Y,$$

where the prefactor applies uniformly across all $\mathbf{y}\in V_Y$. This generalization can be obtained using the prefactor estimate in Lemma 14 and the fact that

$$\mathcal{H}_{V,\mathbf{0}}^{l}(f(\cdot,\mathbf{y})) \leqslant \mathcal{H}_{V\times V_Y,(\mathbf{0},\cdot)}^{l}(f), \quad \forall l\geqslant 0, \forall \mathbf{y}\in V_Y,$$

based on the definitions of the two quantities in Eq. (43) and Eq. (44).

Lemma 16. *Let $f(\mathbf{x}) = f_1(\mathbf{x})f_2(\mathbf{x})$ where $f_1(\mathbf{x})$ and $f_2(\mathbf{x})$ are periodic with respect to $V=[-\frac{1}{2},\frac{1}{2}]^d$ and*

- *$f_1(\mathbf{x})$ is smooth everywhere except at $\mathbf{x}=\mathbf{z}_1=\mathbf{0}$ with order $\gamma\leqslant 0$,*
- *$f_2(\mathbf{x})$ is smooth everywhere except at $\mathbf{x}=\mathbf{z}_2\neq\mathbf{0}$ with order 0.*

Consider an m^d-sized uniform mesh $\mathcal{X}$ in V. Assume that $\mathcal{X}$ satisfies that $\mathbf{z}_1,\mathbf{z}_2$ are either on the mesh or $\Theta(m^{-1})$ away from any mesh points, and m is sufficiently large that $|\mathbf{z}_1-\mathbf{z}_2| = \Omega(m^{-1})$. At $\mathbf{x}=\mathbf{z}_1$ and $\mathbf{x}=\mathbf{z}_2$, $f(\mathbf{x})$ is set to 0. The trapezoidal rule using $\mathcal{X}$ has quadrature error

$$|\mathcal{E}_V(f,\mathcal{X})| \leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{d+1}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{d+1}(f_2)m^{-(d+\gamma)}.$$

If $f(\mathbf{z}_1)$ and $f(\mathbf{z}_2)$ are set to arbitrary $\mathcal{O}(1)$ values, it introduces additional $\mathcal{O}(m^{-d})$ quadrature error.

Proof. First, we can introduce a proper translation $f(\mathbf{x}) \to f(\mathbf{x}-\mathbf{x}_0)$ and move the nonsmooth points $\mathbf{z}_1$ and $\mathbf{z}_2$ to both lie in the smaller cube $[-\frac{1}{4}, \frac{1}{4}]^d$ in V. Assume such a translation has been applied to f and the mesh $\mathcal{X}$, which does not change the integral value due to the function periodicity. Using the cut-off function $\psi_{\frac{1}{2}}(\mathbf{x})$, we split $f(\mathbf{x})$ as

$$f(\mathbf{x}) = f(\mathbf{x})\psi_{\frac{1}{2}}^2(\mathbf{x}) + f(\mathbf{x})(1-\psi_{\frac{1}{2}}^2(\mathbf{x})), \tag{52}$$

where the quadrature error for the second term decays super-algebraically according to the analysis in the proof of Lemma 14. The problem is reduced to the quadrature error estimate for the localized integrand $f\psi_{\frac{1}{2}}^2$ which can be decomposed as $f\psi_{\frac{1}{2}}^2 = (f_1\psi_{\frac{1}{2}})(f_2\psi_{\frac{1}{2}})$. In the following, we abuse the notation f_i to denote $f_i\psi_{\frac{1}{2}}$ and f to denote $f\psi_{\frac{1}{2}}^2$, and then assume that f_1 and f_2 is are both compactly supported in V and smooth everywhere except at $\mathbf{z}_1$ and $\mathbf{z}_2$ with order γ and 0, respectively. Denote $h = 1/m$ be the unit length of $\mathcal{X}$. The remaining analysis follows the proof of Lemma 14.

First, the quadrature error of the trapezoidal rule can be reformulated as

$$\mathcal{E}_V(f,\mathcal{X}) = \left(\mathcal{I}_V\left(f\psi_{h,1}\right) + \mathcal{I}_V\left(f\psi_{h,2}\right)\right) + \mathcal{E}_V\left(f(1-\psi_{h,1})(1-\psi_{h,2}),\mathcal{X}\right), \tag{53}$$

where $\psi_{h,1}(\mathbf{x})$ and $\psi_{h,2}(\mathbf{x})$ are two scaled and shifted cutoff functions centered at $\mathbf{z}_1$ and $\mathbf{z}_2$, respectively, with cutoff radius C_1h as

$$\psi_{h,1}(\mathbf{x}) = \psi\left(\frac{\mathbf{x}-\mathbf{z}_1}{C_1h}\right), \qquad \psi_{h,2}(\mathbf{x}) = \psi\left(\frac{\mathbf{x}-\mathbf{z}_2}{C_1h}\right),$$

where C_1 is a constant such that the two balls $B(\mathbf{z}_1, C_1h)$ and $B(\mathbf{z}_2, C_1h)$ do not overlap with each other or with any mesh points in $\mathcal{X}$ other than $\mathbf{z}_1$ and $\mathbf{z}_2$. Here we use $B(\mathbf{x}, r)$ to denote a ball centered at $\mathbf{x}$ with radius r. Such C_1 exists based on the assumptions over $\mathcal{X}$ in the lemma.

The first part in Eq. (53) can be estimated directly as

$$\begin{aligned}
&\left|\mathcal{I}_V\left(f\psi_{h,1}\right) + \mathcal{I}_V\left(f\psi_{h,2}\right)\right| \\
\leqslant& \int_{B(\mathbf{z}_1,C_1h)} \mathrm{d}\mathbf{x}|f(\mathbf{x})| + \int_{B(\mathbf{z}_2,C_1h)} \mathrm{d}\mathbf{x}|f(\mathbf{x})| \\
\leqslant& C\mathcal{H}^0_{V,\mathbf{z}_1}(f_1)\mathcal{H}^0_{V,\mathbf{z}_2}(f_2)\left(\int_{B(\mathbf{z}_1,C_1h)} \mathrm{d}\mathbf{x}|\mathbf{x}-\mathbf{z}_1|^\gamma + \int_{B(\mathbf{z}_2,C_1h)} \mathrm{d}\mathbf{x}h^\gamma|\mathbf{x}-\mathbf{z}_2|^0\right) \\
\leqslant& C\mathcal{H}^0_{V,\mathbf{z}_1}(f_1)\mathcal{H}^0_{V,\mathbf{z}_2}(f_2)h^{d+\gamma},
\end{aligned}$$

where the second inequality uses the two estimates from Eq. (45) as

$$\begin{aligned}
|f_1(\mathbf{x})| &\leqslant \mathcal{H}^0_{V,\mathbf{z}_1}(f_1)|\mathbf{x}-\mathbf{z}_1|^\gamma, \\
|f_2(\mathbf{x})| &\leqslant \mathcal{H}^0_{V,\mathbf{z}_2}(f_2)|\mathbf{x}-\mathbf{z}_2|^0.
\end{aligned}$$

The second part in Eq. (53) can be estimated using Poisson summation formula with any $\boldsymbol{\alpha} \geqslant \mathbf{0}$ as

$$
\begin{aligned}
&\left|\mathcal{E}_V\left(f(1-\psi_{h,1})(1-\psi_{h,2}), \mathcal{X}\right)\right| \\
\leqslant & \sum_{\mathbf{k} \in 2\pi\mathbb{Z}^d \setminus \{\mathbf{0}\}} \left|\hat{f}_{\psi,h}\left(\frac{\mathbf{k}}{h}\right)\right| \\
\leqslant & \sum_{\mathbf{k} \in 2\pi\mathbb{Z}^d \setminus \{\mathbf{0}\}} \frac{h^{|\boldsymbol{\alpha}|}}{|\mathbf{k}|^{\boldsymbol{\alpha}}} \int_{\mathbb{R}^d} \mathrm{d}\mathbf{x} \left|\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}^{\boldsymbol{\alpha}}}\left(f(\mathbf{x})(1-\psi_{h,1}(\mathbf{x}))(1-\psi_{h,2}(\mathbf{x}))\right)\right| \\
\leqslant & C \sum_{\mathbf{k} \in 2\pi\mathbb{Z}^d \setminus \{\mathbf{0}\}} \frac{h^{|\boldsymbol{\alpha}|}}{|\mathbf{k}|^{\boldsymbol{\alpha}}} \int_{\mathbb{R}^d} \mathrm{d}\mathbf{x} \sum_{\boldsymbol{\alpha}_1+\boldsymbol{\alpha}_2+\boldsymbol{\alpha}_3=\boldsymbol{\alpha}} \left|\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_1}(1-\psi_{h,1}(\mathbf{x}))\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_2}(1-\psi_{h,2}(\mathbf{x}))\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_3} f(\mathbf{x})\right|.
\end{aligned}
\tag{54}
$$

To estimate the last integral for each set of $(\boldsymbol{\alpha}_1, \boldsymbol{\alpha}_2, \boldsymbol{\alpha}_3)$, we consider three cases:

- $\boldsymbol{\alpha}_1 > 0$. Since $1-\psi_{h,1}(\mathbf{x})$ is constant inside $B(\mathbf{z}_1, \frac{1}{2}C_1 h)$ and outside $B(\mathbf{z}_1, C_1 h)$, its derivatives are nonzero only in the annulus $B(\mathbf{z}_1, C_1) \setminus B(\mathbf{z}_1, \frac{1}{2}C_1 h)$. The derivative term in Eq. (54) in this case can be estimated as

$$
|\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_1}(1-\psi_{h,1})\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_2}(1-\psi_{h,2})\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_3} f| \leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{|\boldsymbol{\alpha}|}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{|\boldsymbol{\alpha}|}(f_2) \begin{cases} 0 & |\mathbf{x}-\mathbf{z}_1| < \frac{1}{2}C_1 h \\ 0 & |\mathbf{x}-\mathbf{z}_1| > C_1 h \\ h^{\gamma-|\boldsymbol{\alpha}|} & \text{otherwise} \end{cases}.
$$

  The estimate for $\mathbf{x}$ in the annulus around $\mathbf{z}_1$ above uses for any $\boldsymbol{\beta} \geqslant \mathbf{0}$

$$
\begin{aligned}
\left|\partial_{\mathbf{x}}^{\boldsymbol{\beta}} f(\mathbf{x})\right| &\leqslant C \sum_{\boldsymbol{\beta}_1+\boldsymbol{\beta}_2=\boldsymbol{\beta}} \left|\partial_{\mathbf{x}}^{\boldsymbol{\beta}_1} f_1(\mathbf{x}) \partial_{\mathbf{x}}^{\boldsymbol{\beta}_2} f_2(\mathbf{x})\right| \\
&\leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{|\boldsymbol{\beta}|}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{|\boldsymbol{\beta}|}(f_2) \sum_{\boldsymbol{\beta}_1+\boldsymbol{\beta}_2=\boldsymbol{\beta}} |\mathbf{x}-\mathbf{z}_1|^{\gamma-|\boldsymbol{\beta}_1|}|\mathbf{x}-\mathbf{z}_2|^{-|\boldsymbol{\beta}_2|} \\
&\leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{|\boldsymbol{\beta}|}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{|\boldsymbol{\beta}|}(f_2) \sum_{\boldsymbol{\beta}_1+\boldsymbol{\beta}_2=\boldsymbol{\beta}} h^{\gamma-|\boldsymbol{\beta}_1|}h^{-|\boldsymbol{\beta}_2|} \\
&\leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{|\boldsymbol{\beta}|}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{|\boldsymbol{\beta}|}(f_2) h^{\gamma-|\boldsymbol{\beta}|},
\end{aligned}
$$

  where the third inequality uses the fact $|\mathbf{x}-\mathbf{z}_1| = \mathcal{O}(h)$ and $|\mathbf{x}-\mathbf{z}_2| = \Omega(h)$.

- $\boldsymbol{\alpha}_2 > 0$. Similar to the first case, we could get an estimate of the derivative as

$$
|\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_1}(1-\psi_{h,1})\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_2}(1-\psi_{h,2})\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_3} f| \leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{|\boldsymbol{\alpha}|}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{|\boldsymbol{\alpha}|}(f_2) \begin{cases} 0 & |\mathbf{x}-\mathbf{z}_2| < \frac{1}{2}C_1 h \\ 0 & |\mathbf{x}-\mathbf{z}_2| > C_1 h \\ h^{\gamma-|\boldsymbol{\alpha}|} & \text{otherwise} \end{cases}.
$$

- $\boldsymbol{\alpha}_1 = \boldsymbol{\alpha}_2 = \mathbf{0}, \boldsymbol{\alpha}_3 = \boldsymbol{\alpha}$. We could use the estimate in the first case for $\partial_{\mathbf{x}}^{\boldsymbol{\alpha}} f(\mathbf{x})$ to get

$$
|(1-\psi_{h,1})(1-\psi_{h,2})\partial_{\mathbf{x}}^{\boldsymbol{\alpha}} f| \leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{|\boldsymbol{\alpha}|}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{|\boldsymbol{\alpha}|}(f_2) \begin{cases} 0 & |\mathbf{x}-\mathbf{z}_1| < \frac{1}{2}C_1 h \\ 0 & |\mathbf{x}-\mathbf{z}_2| < \frac{1}{2}C_1 h \\ \sum_{\boldsymbol{\beta}_1+\boldsymbol{\beta}_2=\boldsymbol{\alpha}} |\mathbf{x}-\mathbf{z}_1|^{\gamma-|\boldsymbol{\beta}_1|}|\mathbf{x}-\mathbf{z}_2|^{-|\boldsymbol{\beta}_2|} & \text{otherwise with } |\mathbf{x}| < 1 \end{cases}.
$$

Based on the analysis of the three cases above, we get an estimate of the integral as

$$\int_{\mathbb{R}^d} \mathrm{d}\mathbf{x}\, |\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_1}(1-\psi_{h,1})\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_2}(1-\psi_{h,2})\partial_{\mathbf{x}}^{\boldsymbol{\alpha}_3} f| \leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{|\boldsymbol{\alpha}|}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{|\boldsymbol{\alpha}|}(f_2)\begin{cases} h^{\gamma-|\boldsymbol{\alpha}|+d} & \boldsymbol{\alpha}_1 > 0 \text{ or } \boldsymbol{\alpha}_2 > 0 \\ 1 + h^{\gamma-|\boldsymbol{\alpha}|+d} & \boldsymbol{\alpha}_1 = \boldsymbol{\alpha}_2 = \mathbf{0} \end{cases}.$$

The estimate for the case with $\boldsymbol{\alpha}_1 = \boldsymbol{\alpha}_2 = \mathbf{0}$ can be obtained directly when $\boldsymbol{\beta}_1 = \boldsymbol{\alpha}$ and be bounded as follows when $\boldsymbol{\beta}_1 < \boldsymbol{\alpha}$

$$\begin{aligned}
&\int_{B(\mathbf{0},2)\setminus(B(\mathbf{z}_1,\frac{1}{2}C_1h)\cup B(\mathbf{z}_2,\frac{1}{2}C_1h))} \mathrm{d}\mathbf{x}|\mathbf{x}-\mathbf{z}_1|^{\gamma-|\boldsymbol{\beta}_1|}|\mathbf{x}-\mathbf{z}_2|^{|\boldsymbol{\beta}_1|-|\boldsymbol{\alpha}|} \\
\leqslant& \left(\int_{B(\mathbf{0},2)\setminus B(\mathbf{z}_1,\frac{1}{2}C_1h)} \mathrm{d}\mathbf{x}|\mathbf{x}-\mathbf{z}_1|^{p\gamma-p|\boldsymbol{\beta}_1|}\right)^{\frac{1}{p}} \left(\int_{B(\mathbf{0},2)\setminus B(\mathbf{z}_2,\frac{1}{2}C_1h)} \mathrm{d}\mathbf{x}|\mathbf{x}-\mathbf{z}_2|^{q|\boldsymbol{\beta}_1|-q|\boldsymbol{\alpha}|}\right)^{\frac{1}{q}} \\
\leqslant& C\left(1+h^{p\gamma-p|\boldsymbol{\beta}_1|+d}\right)^{\frac{1}{p}}\left(1+h^{q|\boldsymbol{\beta}_1|-q|\boldsymbol{\alpha}|+d}\right)^{\frac{1}{q}} \\
\leqslant& C\left(1+h^{\gamma-|\boldsymbol{\beta}_1|+\frac{1}{p}d}+h^{|\boldsymbol{\beta}_1|-|\boldsymbol{\alpha}|+\frac{1}{q}d}+h^{\gamma-|\boldsymbol{\alpha}|+d}\right) \leqslant C\left(1+h^{\gamma-|\boldsymbol{\alpha}|+d}\right),
\end{aligned}$$

where the last estimate assumes $|\boldsymbol{\alpha}| \geqslant \gamma + d$ and is obtained by setting the Hölder inequality exponents as

$$\begin{cases} p = 1, q = \infty & \text{when } d+\gamma-|\boldsymbol{\beta}_1| \leqslant 0 \\ p = \frac{d}{|\boldsymbol{\beta}_1|-\gamma}, q = \frac{d}{d+\gamma-|\boldsymbol{\beta}_1|} & \text{when } d+\gamma-|\boldsymbol{\beta}_1| > 0 \end{cases}.$$

Plugging this estimate into Eq. (54) and choosing an arbitrary $\boldsymbol{\alpha}$ with $|\boldsymbol{\alpha}| = d+1$, we obtain

$$|\mathcal{E}_V\left(f(1-\psi_{h,1})(1-\psi_{h,2}),\mathcal{X}\right)| \leqslant C\mathcal{H}_{V,\mathbf{z}_1}^{d+1}(f_1)\mathcal{H}_{V,\mathbf{z}_2}^{d+1}(f_2)h^{d+\gamma},$$

which together with Eq. (53) proves the lemma. □

Remark 17. *If we replace $f_i(\mathbf{x})$ with $i = 1, 2$ by $f_i(\mathbf{x}, \mathbf{y})$ defined in $V \times V_Y$ which is smooth everywhere in $V \times V_Y$ except at $\mathbf{x} = \mathbf{z}_i$ with order γ and 0, respectively, Lemma 16 can be generalized to*

$$|\mathcal{E}_V(f_1(\cdot,\mathbf{y})f_2(\cdot,\mathbf{y}),\mathcal{X})| \leqslant C\mathcal{H}_{V\times V_Y,(\mathbf{0},\cdot)}^{d+1}(f_1)\mathcal{H}_{V\times V_Y,(\mathbf{0},\cdot)}^{d+1}(f_2)m^{-(d+\gamma)}, \quad \forall \mathbf{y} \in V_Y,$$

where the prefactor applies uniformly across all $\mathbf{y} \in V_Y$. This generalization can be obtained using the prefactor characterization in Lemma 14 and a similar discussion as in Remark 15.

Lemma 18. *Let $f(\mathbf{x}_1, \mathbf{x}_2) = f_1(\mathbf{x}_1, \mathbf{x}_2)f_2(\mathbf{x}_1, \mathbf{x}_2)$ where $f_1(\mathbf{x}_1, \mathbf{x}_2)$ and $f_2(\mathbf{x}_1, \mathbf{x}_2)$ are periodic with respect to $\mathbf{x}_1, \mathbf{x}_2 \in V = [-\frac{1}{2}, \frac{1}{2}]^d$ and*

- *$f_1(\mathbf{x}_1, \mathbf{x}_2)$ is smooth everywhere except at $\mathbf{x}_1 = \mathbf{0}$ with order γ_1,*
- *$f_2(\mathbf{x}_1, \mathbf{x}_2)$ is smooth everywhere except at $\mathbf{x}_2 = \mathbf{0}$ with order γ_2.*

Consider an m^d-sized uniform mesh $\mathcal{X}$ in V that contains $\mathbf{x} = \mathbf{0}$. At $\mathbf{x}_1 = \mathbf{0}$ or $\mathbf{x}_2 = \mathbf{0}$, $f(\mathbf{x}_1, \mathbf{x}_2)$ is set to zero. The trapezoidal rule using $\mathcal{X} \times \mathcal{X}$ for $f(\mathbf{x}_1, \mathbf{x}_2)$ has quadrature error

$$|\mathcal{E}_{V\times V}(f,\mathcal{X}\times\mathcal{X})| \leqslant C\mathcal{H}_{V\times V,(\mathbf{0},\cdot)}^{d+\max(1,\gamma_1,\gamma_2)}(f_1)\mathcal{H}_{V\times V,(\cdot,\mathbf{0})}^{d+\max(1,\gamma_1,\gamma_2)}(f_2)m^{-(d+\min_i \gamma_i)},$$

If $f(\mathbf{x}_1, \mathbf{0})$ and $f(\mathbf{0}, \mathbf{x}_2)$ are set to arbitrary $\mathcal{O}(1)$ values, it introduces additional $\mathcal{O}(m^{-d})$ quadrature error.

Proof. The quadrature error for f can be split into two parts

$$\mathcal{E}_{V\times V}(f, \mathcal{X}\times\mathcal{X}) = \mathcal{E}_V\left(\int_V \mathrm{d}\mathbf{x}_2 f(\cdot, \mathbf{x}_2), \mathcal{X}\right) + \frac{|V|}{m^d}\sum_{\mathbf{x}_1\in\mathcal{X}} \mathcal{E}_V\left(f(\mathbf{x}_1, \cdot), \mathcal{X}\right). \tag{55}$$

For the first quadrature error, $\int_V \mathrm{d}\mathbf{x}_2 f(\mathbf{x}_1, \mathbf{x}_2)$ as a function of $\mathbf{x}_1$ is periodic with respect to V. Using the Leibniz integral rule, we can show that the integrand is smooth everywhere in V except at $\mathbf{x}_1 = \mathbf{0}$ and its derivatives can be estimated as

$$\begin{aligned}
\left|\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}_1^{\boldsymbol{\alpha}}}\int_V \mathrm{d}\mathbf{x}_2 f(\mathbf{x}_1, \mathbf{x}_2)\right| &= \left|\int_V \mathrm{d}\mathbf{x}_2 \frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{x}_1^{\boldsymbol{\alpha}}} f(\mathbf{x}_1, \mathbf{x}_2)\right| \\
&\leqslant C\int_V \mathrm{d}\mathbf{x}_2 \sum_{\boldsymbol{\alpha}_1+\boldsymbol{\alpha}_2=\boldsymbol{\alpha}} \left|\partial_{\mathbf{x}_1}^{\boldsymbol{\alpha}_1} f_1(\mathbf{x}_1, \mathbf{x}_2)\right| \left|\partial_{\mathbf{x}_1}^{\boldsymbol{\alpha}_2} f_2(\mathbf{x}_1, \mathbf{x}_2)\right| \\
&\leqslant C\int_V \mathrm{d}\mathbf{x}_2 \sum_{\boldsymbol{\alpha}_1+\boldsymbol{\alpha}_2=\boldsymbol{\alpha}} \left(\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\mathbf{0},\cdot)}(f_1)|\mathbf{x}_1|^{\gamma_1-|\boldsymbol{\alpha}_1|}\right)\left(\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\cdot,\mathbf{0})}(f_2)|\mathbf{x}_2|^{\gamma_2}\right) \\
&\leqslant C\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\cdot,\mathbf{0})}(f_2)|\mathbf{x}_1|^{\gamma_1-|\boldsymbol{\alpha}|},
\end{aligned}$$

where the third inequality uses the estimate in Eq. (44) for multivariate functions with algebraic singularity. This derivative estimate suggests that $\int_V \mathrm{d}\mathbf{x}_2 f(\mathbf{x}_1, \mathbf{x}_2)$ is smooth everywhere in V except at $\mathbf{x}_1 = \mathbf{0}$ with order γ_1, and its algebraic singularity characterization satisfies,

$$\mathcal{H}^l_{V,\mathbf{0}}\left(\int_V \mathrm{d}\mathbf{x}_2 f(\cdot, \mathbf{x}_2)\right) \leqslant C\mathcal{H}^l_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^l_{V\times V,(\cdot,\mathbf{0})}(f_2), \quad \forall l \geqslant 0.$$

Lemma 14 then gives

$$\left|\mathcal{E}_V\left(\int_V \mathrm{d}\mathbf{x}_2 f(\cdot, \mathbf{x}_2), \mathcal{X}\right)\right| \leqslant C\mathcal{H}^{d+\max(1,\gamma_1)}_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^{d+\max(1,\gamma_1)}_{V\times V,(\cdot,\mathbf{0})}(f_2)m^{-(d+\gamma_1)}. \tag{56}$$

For the second quadrature error with each $\mathbf{x}_1 \in \mathcal{X}$, $f(\mathbf{x}_1, \mathbf{x}_2)$ as a function of $\mathbf{x}_2$ is periodic and smooth everywhere except at $\mathbf{x}_2 = \mathbf{0}$ with order γ_2. Fixing $\mathbf{x}_1$, Lemma 14 gives

$$|\mathcal{E}_V\left(f(\mathbf{x}_1, \cdot), \mathcal{X}\right)| \leqslant C\mathcal{H}^{d+\max(1,\gamma_2)}_{V,\mathbf{0}}(f(\mathbf{x}_1, \cdot))m^{-(d+\gamma_2)},$$

where the characterization prefactor with $l = d + \max(1, \gamma_2)$ and any $\mathbf{x}_1 \in V$ can be further estimated by its definition in Eq. (43) as

$$\begin{aligned}
\mathcal{H}^l_{V,\mathbf{0}}(f(\mathbf{x}_1, \cdot)) &= \max_{|\boldsymbol{\alpha}|\leqslant l}\left\|\frac{\partial^{\boldsymbol{\alpha}}_{\mathbf{x}_2} f(\mathbf{x}_1, \mathbf{x}_2)}{|\mathbf{x}_2|^{\gamma_2-|\boldsymbol{\alpha}|}}\right\|_{L^\infty(V)} \\
&\leqslant C\max_{|\boldsymbol{\alpha}|\leqslant l}\sum_{\boldsymbol{\alpha}_1+\boldsymbol{\alpha}_2=\boldsymbol{\alpha}}\left\|\partial^{\boldsymbol{\alpha}_1}_{\mathbf{x}_2} f_1(\mathbf{x}_1, \mathbf{x}_2)\frac{\partial^{\boldsymbol{\alpha}_2}_{\mathbf{x}_2} f_2(\mathbf{x}_1, \mathbf{x}_2)}{|\mathbf{x}_2|^{\gamma_2-|\boldsymbol{\alpha}_2|}}|\mathbf{x}_2|^{|\boldsymbol{\alpha}_1|}\right\|_{L^\infty(V)} \\
&\leqslant C\max_{|\boldsymbol{\alpha}|\leqslant l}\sum_{\boldsymbol{\alpha}_1+\boldsymbol{\alpha}_2=\boldsymbol{\alpha}}\left(\mathcal{H}^l_{V\times V,(\mathbf{0},\cdot)}(f_1)|\mathbf{x}_1|^{\gamma_1}\right)\left(\mathcal{H}^l_{V\times V,(\cdot,\mathbf{0})}(f_2)\right)\left\||\mathbf{x}_2|^{|\boldsymbol{\alpha}_1|}\right\|_{L^\infty(V)} \\
&\leqslant C\mathcal{H}^l_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^l_{V\times V,(\cdot,\mathbf{0})}(f_2)|\mathbf{x}_1|^{\gamma_1},
\end{aligned} \tag{57}$$

where the third inequality uses the estimate in Eq. (44).

Plugging the two estimates Eq. (56) and Eq. (57) above into Eq. (55), we have

$$\begin{aligned}|\mathcal{E}_{V\times V}(f,\mathcal{X}\times\mathcal{X})| &\leqslant C\mathcal{H}^{l_*}_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^{l_*}_{V\times V,(\cdot,\mathbf{0})}(f_2)\left(m^{-(d+\gamma_1)}+\frac{|V|}{m^d}\sum_{\mathbf{x}_1\in\mathcal{X}}|\mathbf{x}_1|^{\gamma_1}m^{-(d+\gamma_2)}\right)\\ &\leqslant C\mathcal{H}^{l_*}_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^{l_*}_{V\times V,(\cdot,\mathbf{0})}(f_2)m^{-(d+\min_i\gamma_i)},\end{aligned}$$

where $l_* = d+\max(1,\gamma_1,\gamma_2)$. This finishes the proof. □

Remark 19. *If we replace $f_i(\mathbf{x}_1,\mathbf{x}_2)$ by $f_i(\mathbf{x}_1,\mathbf{x}_2,\mathbf{y})$ that are smooth and periodic with respect to $\mathbf{y}\in V_Y$, Lemma 18 can be generalized as*

$$|\mathcal{E}_{V\times V}(f(\cdot,\cdot,\mathbf{y}),\mathcal{X}\times\mathcal{X})| \leqslant C\mathcal{H}^{d+\max(1,\gamma_1,\gamma_2)}_{V\times V\times V_Y,(\mathbf{0},\cdot,\cdot)}(f_1)\mathcal{H}^{d+\max(1,\gamma_1,\gamma_2)}_{V\times V\times V_Y,(\cdot,\mathbf{0}),\cdot}(f_2)m^{-(d+\min_i\gamma_i)},\ \forall\mathbf{y}\in V_Y,$$

where the prefactor applies uniformly across all $\mathbf{y}\in V_Y$.

Lemma 20. *Let $f(\mathbf{x}_1,\mathbf{x}_2) = f_1(\mathbf{x}_1,\mathbf{x}_2)f_2(\mathbf{x}_1,\mathbf{x}_2)f_3(\mathbf{x}_1,\mathbf{x}_2-\mathbf{x}_1)$ satisfy that*

- *$f_1(\mathbf{x}_1,\mathbf{x}_2)$ and $f_2(\mathbf{x}_1,\mathbf{x}_2)$ are the same as in Lemma 18,*
- *$f_3(\mathbf{x}_1,\mathbf{z})$ is periodic with respect to $\mathbf{x}_1,\mathbf{z}\in V$ and smooth everywhere except at $\mathbf{z}=\mathbf{0}$ with order 0.*

Consider a uniform mesh $\mathcal{X}$ in V that is of size m^d and contains $\mathbf{x}=\mathbf{0}$. At $\mathbf{x}_1=\mathbf{0}$ or $\mathbf{x}_2=\mathbf{0}$, $f(\mathbf{x}_1,\mathbf{x}_2)$ is set to 0. The trapezoidal rule using $\mathcal{X}\times\mathcal{X}$ for $f(\mathbf{x}_1,\mathbf{x}_2)$ has quadrature error

$$|\mathcal{E}_{V\times V}(f,\mathcal{X}\times\mathcal{X})| \leqslant C\mathcal{H}^{d+\max(1,\gamma_1,\gamma_2)}_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^{d+\max(1,\gamma_1,\gamma_2)}_{V\times V,(\cdot,\mathbf{0})}(f_2)\mathcal{H}^{d+\max(1,\gamma_1,\gamma_2)}_{V\times V,(\cdot,\mathbf{0})}(f_3)m^{-(d+\min_i\gamma_i)}.$$

If $f(\mathbf{x}_1,\mathbf{0})$ and $f(\mathbf{0},\mathbf{x}_2)$ are set to arbitrary $\mathcal{O}(1)$ values, it introduces additional $\mathcal{O}(m^{-d})$ quadrature error.

Proof. For this new function, we still split the quadrature error into the two parts as

$$\mathcal{E}_{V\times V}(f,\mathcal{X}\times\mathcal{X}) = \mathcal{E}_V\left(\int_V \mathrm{d}\mathbf{x}_2 f(\cdot,\mathbf{x}_2),\mathcal{X}\right)+\frac{|V|}{m^d}\sum_{\mathbf{x}_1\in\mathcal{X}}\mathcal{E}_V\left(f(\mathbf{x}_1,\cdot),\mathcal{X}\right). \tag{58}$$

In the first part of the quadrature error, $\int_V \mathrm{d}\mathbf{x}_2 f(\mathbf{x}_1,\mathbf{x}_2)$ as a function of $\mathbf{x}_1$ is periodic with respect to V and below we first show it to be smooth everywhere except at $\mathbf{x}_1=\mathbf{0}$ with order γ_1.

In order to exploit the existing nonsmoothness analysis in Lemma 11, we define an auxiliary function

$$F(\mathbf{x}_1,\mathbf{y}) = \int_V \mathrm{d}\mathbf{x}_2 f_1(\mathbf{y},\mathbf{x}_2)f_2(\mathbf{y},\mathbf{x}_2)f_3(\mathbf{y},\mathbf{x}_2-\mathbf{x}_1),$$

which satisfies that $\int_V \mathrm{d}\mathbf{x}_2 f(\mathbf{x}_1,\mathbf{x}_2) = F(\mathbf{x}_1,\mathbf{x}_1)$. First fixing $\mathbf{y}$, the integrand for $F(\mathbf{x}_1,\mathbf{y})$ as a function of $\mathbf{x}_1,\mathbf{x}_2$ meets the condition in Lemma 11 and thus $F(\mathbf{x}_1,\mathbf{y})$ is smooth everywhere with respect to $\mathbf{x}_1\in V$ except at $\mathbf{x}_1=\mathbf{0}$ with order 0. Next, fix $\mathbf{x}_1$ and consider a point $\mathbf{y}_0\neq\mathbf{0}\in V$. It can be verified that there is an open domain containing $\mathbf{y}_0$ where with any $\mathbf{y}$ in this domain the integrand is smooth with respect to $\mathbf{x}_2$ except at $\mathbf{x}_2=\mathbf{0}$ and $\mathbf{x}_2=\mathbf{x}_1$, and the absolute integrand is bounded by $C|\mathbf{x}_2|^{\gamma_2}$ (from the boundedness of f_1, f_3 and the algebraic singularity of f_2) which is integrable in V. This meets the condition for the Leibniz integral rule around $\mathbf{y}_0$ and thus $F(\mathbf{x}_1,\mathbf{y})$ is smooth with respect to $\mathbf{y}\in V$ except at $\mathbf{y}=\mathbf{0}$. These two discussions thus show that $F(\mathbf{x}_1,\mathbf{y})$ is smooth at any points with $\mathbf{x}_1\neq\mathbf{0},\mathbf{y}\neq\mathbf{0}$.

Furthermore, any partial derivative of $F(\mathbf{x}_1,\mathbf{y})$ over $\mathbf{y}$ in $V\setminus\{\mathbf{0}\}$ can be estimated as

$$\begin{aligned}\left|\partial_{\mathbf{y}}^{\boldsymbol{\alpha}} F(\mathbf{x}_1,\mathbf{y})\right| &\leqslant C\int_V \mathrm{d}\mathbf{x}_2 \sum_{\boldsymbol{\alpha}_1+\boldsymbol{\alpha}_2+\boldsymbol{\alpha}_3=\boldsymbol{\alpha}} \left|\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1} f_1(\mathbf{y},\mathbf{x}_2)\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_2} f_2(\mathbf{y},\mathbf{x}_2)\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_3} f_3(\mathbf{y},\mathbf{x}_2-\mathbf{x}_1)\right| \\ &\leqslant C\int_V \mathrm{d}\mathbf{x}_2 \sum_{\boldsymbol{\alpha}_1+\boldsymbol{\alpha}_2+\boldsymbol{\alpha}_3=\boldsymbol{\alpha}} \mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\mathbf{0},\cdot)}(f_1)|\mathbf{y}|^{\gamma_1-|\boldsymbol{\alpha}_1|}\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\cdot,\mathbf{0})}(f_2)|\mathbf{x}_2|^{\gamma_2}\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\cdot,\mathbf{0})}(f_3) \\ &\leqslant C\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\cdot,\mathbf{0})}(f_2)\mathcal{H}^{|\boldsymbol{\alpha}|}_{V\times V,(\cdot,\mathbf{0})}(f_3)|\mathbf{y}|^{\gamma_1-|\boldsymbol{\beta}|} \\ &= CC_{f_1,f_2,f_3,|\boldsymbol{\alpha}|}|\mathbf{y}|^{\gamma_1-|\boldsymbol{\beta}|},\end{aligned}$$

where $C_{f_1,f_2,f_3,|\boldsymbol{\alpha}|}$ denotes the product of the three algebraic singularity prefactors for brevity.

Substituting $\mathbf{y}$ by $\mathbf{x}_1$ in the above estimate then shows that $F(\mathbf{x}_1,\mathbf{x}_1) = \int_V \mathrm{d}\mathbf{x}_2 f(\mathbf{x}_1,\mathbf{x}_2)$ is smooth everywhere except at $\mathbf{x}_1=\mathbf{0}$ with order γ_1. Lemma 14 then gives

$$\left|\mathcal{E}_V\left(\int_V \mathrm{d}\mathbf{x}_2 f(\cdot,\mathbf{x}_2),\mathcal{X}\right)\right| \leqslant CC_{f_1,f_2,f_3,l_1}m^{-(d+\gamma_1)}, \quad \text{with } l_1 = d+\max(1,\gamma_1). \tag{59}$$

For the second part of the quadrature error with each $\mathbf{x}_1\in\mathcal{X}$, $f(\mathbf{x}_1,\mathbf{x}_2)$ as a function of $\mathbf{x}_2$ is periodic and smooth everywhere except at $\mathbf{x}_2=\mathbf{0}$ and $\mathbf{x}_2=\mathbf{x}_1$ with orders γ_2 and 0, respectively. Applying Lemma 16 and Remark 17 with the decomposition $f=(f_1f_2)(f_3)$ then shows that

$$\begin{aligned}\left|\mathcal{E}_V\left(f(\mathbf{x}_1,\cdot),\mathcal{X}\right)\right| &\leqslant C\mathcal{H}^{l_2}_{V,\mathbf{0}}((f_1f_2)(\mathbf{x}_1,\cdot))\mathcal{H}^{l_2}_{V\times V,(\cdot,\mathbf{0})}(f_3)m^{-(d+\gamma_2)} \\ &\leqslant C\mathcal{H}^{l_2}_{V\times V,(\mathbf{0},\cdot)}(f_1)\mathcal{H}^{l_2}_{V\times V,(\cdot,\mathbf{0})}(f_2)|\mathbf{x}_1|^{\gamma_1}\mathcal{H}^{l_2}_{V\times V,(\cdot,\mathbf{0})}(f_3)m^{-(d+\gamma_2)} \\ &\leqslant CC_{f_1,f_2,f_3,l_2}|\mathbf{x}_1|^{\gamma_1}m^{-(d+\gamma_2)},\end{aligned} \tag{60}$$

where $l_2=d+\max(1,\gamma_2)$ and the second inequality is derived as

$$\begin{aligned}\mathcal{H}^{l_2}_{V,\mathbf{0}}((f_1f_2)(\mathbf{x}_1,\cdot)) &= \max_{|\boldsymbol{\alpha}|\leqslant l_2}\left\|\frac{\partial^{\boldsymbol{\alpha}}_{\mathbf{x}_2}\left(f_1(\mathbf{x}_1,\mathbf{x}_2)f_2(\mathbf{x}_1,\mathbf{x}_2)\right)}{|\mathbf{x}_2|^{\gamma_2-|\boldsymbol{\alpha}|}}\right\|_{L^\infty(V)} \\ &\leqslant C\max_{|\boldsymbol{\alpha}|\leqslant l_2}\sum_{\boldsymbol{\alpha}_1+\boldsymbol{\alpha}_2=\boldsymbol{\alpha}}\left\|\partial^{\boldsymbol{\alpha}_1}_{\mathbf{x}_2}f_1(\mathbf{x}_1,\mathbf{x}_2)\frac{\partial^{\boldsymbol{\alpha}_2}_{\mathbf{x}_2}f_2(\mathbf{x}_1,\mathbf{x}_2)}{|\mathbf{x}_2|^{\gamma_2-|\boldsymbol{\alpha}_2|}}|\mathbf{x}_2|^{|\boldsymbol{\alpha}_1|}\right\|_{L^\infty(V)} \\ &\leqslant C\left(\mathcal{H}^{l_2}_{V\times V,(\mathbf{0},\cdot)}(f_1)|\mathbf{x}_1|^{\gamma_1}\right)\mathcal{H}^{l_2}_{V\times V,(\cdot,\mathbf{0})}(f_2).\end{aligned}$$

Plugging the two estimates Eq. (59) and Eq. (60) into Eq. (55), we have

$$\begin{aligned}\left|\mathcal{E}_{V\times V}(f,\mathcal{X}\times\mathcal{X})\right| &\leqslant CC_{f_1,f_2,f_3,l_*}\left(m^{-(d+\gamma_1)}+\frac{|V|}{m^d}\sum_{\mathbf{x}_1\in\mathcal{X}}|\mathbf{x}_1|^{\gamma_1}m^{-(d+\gamma_2)}\right) \\ &\leqslant CC_{f_1,f_2,f_3,l_*}m^{-(d+\min_i\gamma_i)}\end{aligned}$$

with $l_*=d+\max(1,\gamma_1,\gamma_2)$. This finishes the proof. □

Remark 21. *If we replace $f_i(\mathbf{x}_1,\mathbf{x}_2)$ by $f_i(\mathbf{x}_1,\mathbf{x}_2,\mathbf{y})$ that are smooth and periodic with respect to $\mathbf{y}\in V_Y$, Lemma 20 can be generalized as*

$$\left|\mathcal{E}_{V\times V}(f(\cdot,\cdot,\mathbf{y}),\mathcal{X}\times\mathcal{X})\right| \leqslant CC_{f_1,f_2,f_3}m^{-(d+\min_i\gamma_i)},\ \forall\mathbf{y}\in V_Y,$$

where prefactor $C_{f_1,f_2,f_3} = \mathcal{H}^{d+\max(1,\gamma_1,\gamma_2)}_{V\times V\times V_Y,(\mathbf{0},\cdot,\cdot)}(f_1)\mathcal{H}^{d+\max(1,\gamma_1,\gamma_2)}_{V\times V\times V_Y,(\cdot,\mathbf{0},\cdot)}(f_2)\mathcal{H}^{d+\max(1,\gamma_1,\gamma_2)}_{V\times V\times V_Y,(\cdot,\mathbf{0},\cdot)}(f_3)$ applies uniformly across all $\mathbf{y}\in V_Y$.

Appendix E PROOF OF LEMMA 11: SINGULARITY STRUCTURE OF FUNCTIONS IN INTEGRAL FORM

In this proof, we assume $\gamma_2 = \min_i \gamma_i \leqslant \gamma_1$. Otherwise if $\gamma_2 > \gamma_1$, the target function can be reformulated as

$$F(\mathbf{y},\mathbf{z}) = \int_{V-\mathbf{y}} \mathrm{d}\mathbf{x} f(\mathbf{x}+\mathbf{y},\mathbf{x},\mathbf{z}) = \int_{V} \mathrm{d}\mathbf{x} f(\mathbf{x}+\mathbf{y},\mathbf{x},\mathbf{z}),$$

using change of variable $\mathbf{x} \to \mathbf{x}+\mathbf{y}$ and integrand periodicity with respect to $\mathbf{x} \in V$. With this reformulation, the following proof can be applied to $\widetilde{f}(\mathbf{x}_1,\mathbf{x}_2,\mathbf{z}) = f(\mathbf{x}_2,\mathbf{x}_1,\mathbf{z})$ with $F(\mathbf{y},\mathbf{z}) = \int_V \mathrm{d}\mathbf{x}\widetilde{f}(\mathbf{x},\mathbf{x}+\mathbf{y},\mathbf{z})$, where the order for the second variable of $\widetilde{f}$ equals γ_1 and is the minimum order.

First we study the smoothness property of $F(\mathbf{y},\mathbf{z})$ with respect to $\mathbf{y} \in V$ while fixing $\mathbf{z}$. For notation brevity, we denote $F(\mathbf{y},\mathbf{z})$ and $f(\mathbf{x}_1,\mathbf{x}_2,\mathbf{z})$ by $F_{\mathbf{z}}(\mathbf{y})$ and $f_{\mathbf{z}}(\mathbf{x}_1,\mathbf{x}_2)$, respectively, when $\mathbf{z}$ is assumed to be a fixed point in V_Z. Consider an arbitrary open ball domain B_σ of radius σ in V that does not contain $\mathbf{0}$. We can split $F_{\mathbf{z}}(\mathbf{y})$ for any $\mathbf{y} \in B_\sigma$ into two parts,

$$F_{\mathbf{z}}(\mathbf{y}) = \int_{V\setminus B_\sigma} \mathrm{d}\mathbf{x} f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}) + \int_{B_\sigma} \mathrm{d}\mathbf{x} f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}), \quad \forall \mathbf{y} \in B_\sigma. \tag{61}$$

For the first term, its integrand $f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y})$ is smooth at $(\mathbf{x},\mathbf{y}) \in (V \setminus (B_\sigma \cup \{\mathbf{0}\})) \times B_\sigma$ and its partial derivatives over $\mathbf{y} \in B_\sigma$ are integrable over $\mathbf{x} \in V \setminus B_\sigma$ based on the nonsmoothness characterization in Eq. (32). We thus can use the Leibniz integral rule to prove that the first term is smooth at $\mathbf{y} \in B_\sigma$ and

$$\frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{y}^{\boldsymbol{\alpha}}} \int_{V\setminus B_\sigma} \mathrm{d}\mathbf{x} f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}) = \int_{V\setminus B_\sigma} \mathrm{d}\mathbf{x} \frac{\partial^{\boldsymbol{\alpha}}}{\partial \mathbf{y}^{\boldsymbol{\alpha}}} f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}), \quad \forall \mathbf{y} \in B_\sigma.$$

For the second term with any $\mathbf{y} \in B_\sigma$, we introduce a small perturbation $\delta\mathbf{y}$ to $\mathbf{y}$ such that $\mathbf{y}+\delta\mathbf{y} \in B_\sigma$ and $B_\sigma + \delta\mathbf{y}$ does not contain $\mathbf{0}$, and consider the difference between the second term evaluated at $\mathbf{y}+\delta\mathbf{y}$ and $\mathbf{y}$ as

$$\begin{aligned}
&\int_{B_\sigma} \mathrm{d}\mathbf{x} f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}-\delta\mathbf{y}) - \int_{B_\sigma} \mathrm{d}\mathbf{x} f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}) \\
=&\int_{B_\sigma} \mathrm{d}\mathbf{x}\Big(f_{\mathbf{z}}(\mathbf{x}-\delta\mathbf{y},\mathbf{x}-\mathbf{y}-\delta\mathbf{y}) - f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}) + f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}-\delta\mathbf{y}) - f_{\mathbf{z}}(\mathbf{x}-\delta\mathbf{y},\mathbf{x}-\mathbf{y}-\delta\mathbf{y})\Big) \\
=&\left(\int_{B_\sigma-\delta\mathbf{y}} \mathrm{d}\mathbf{x} - \int_{B_\sigma} \mathrm{d}\mathbf{x}\right) f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}) + \int_{B_\sigma-\delta\mathbf{y}} \mathrm{d}\mathbf{x}\left(f_{\mathbf{z}}(\mathbf{x}+\delta\mathbf{y},\mathbf{x}-\mathbf{y}) - f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y})\right) \\
=&\int_{\partial B_\sigma} \mathrm{d}S f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y})(-\delta\mathbf{y})\cdot\mathbf{n}(\mathbf{x}) + \mathcal{O}(|\delta\mathbf{y}|^2) + \int_{B_\sigma-\delta\mathbf{y}} \mathrm{d}\mathbf{x}\delta\mathbf{y}\cdot\nabla_1 f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}) + \mathcal{O}(|\delta\mathbf{y}|^2) \\
=&\delta\mathbf{y}\cdot\left(-\int_{\partial B_\sigma} \mathrm{d}S f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y})\mathbf{n}(\mathbf{x}) + \int_{B_\sigma} \mathrm{d}\mathbf{x}\nabla_1 f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y})\right) + \mathcal{O}(|\delta\mathbf{y}|^2),
\end{aligned}$$

where $\nabla_1 f_{\mathbf{z}}(\cdot,\cdot)$ denotes the gradient of f over its first variable and similarly for notation $\partial_1^{\boldsymbol{\alpha}} f(\cdot,\cdot)$ in later use. This calculation shows that the second term in Eq. (61) is continuous at any $\mathbf{y} \in B_\sigma$ up to first order derivatives and its gradient equals to the term in the parenthesis above.

Putting the above analysis for the two terms in Eq. (61) together, we have

$$\nabla F_{\mathbf{z}}(\mathbf{y}) = \int_{V\setminus B_\sigma} \mathrm{d}\mathbf{x}\nabla_{\mathbf{y}} f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}) - \int_{\partial B_\sigma} \mathrm{d}S f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y})\mathbf{n}(\mathbf{x}) + \int_{B_\sigma} \mathrm{d}\mathbf{x}\nabla_1 f_{\mathbf{z}}(\mathbf{x},\mathbf{x}-\mathbf{y}), \quad \mathbf{y} \in B_\sigma. \tag{62}$$

It is worth noting that the above smoothness analysis and the gradient calculation in Eq. (62) work for any open domain $B_\sigma \subset V$ that does not contain $\mathbf{0}$. Thus, the analysis above shows the $F_{\mathbf{z}}(\mathbf{y})$ is continuous up to first order derivative at any $\mathbf{y} \in V \setminus \{\mathbf{0}\}$.

In Eq. (62), the integrands of the first two terms are smooth at any $\mathbf{y} \in B_\sigma$ as $\mathbf{x} \notin B_\sigma$. These two integrals are thus smooth with respect to $\mathbf{y} \in B_\sigma$ according to the Leibniz integral rule. For the third term, each entry in $\nabla_1 f_{\mathbf{z}}(\mathbf{x}_1, \mathbf{x}_2)$ shares similar nonsmooth behavior as $f_{\mathbf{z}}(\mathbf{x}_1, \mathbf{x}_2)$ described in Eq. (32) only with γ_1 changed to $\gamma_1 - 1$. Due to this similarity, we can use the same analysis for $\int_V \mathrm{d}\mathbf{x} f_{\mathbf{z}}(\mathbf{x}, \mathbf{x}-\mathbf{y})$ above to prove the continuity up to first order derivatives for the third term at any $\mathbf{y} \in V \setminus \{\mathbf{0}\}$. Recursively applying this analysis, we then prove that $F_{\mathbf{z}}(\mathbf{y})$ is smooth in B_σ and thus in $V \setminus \{\mathbf{0}\}$.

Now taking $\mathbf{z}$ back into account, the above analysis shows that $F(\mathbf{y}, \mathbf{z})$ is smooth with respect to $\mathbf{y} \in V \setminus \{\mathbf{0}\}$ for any fixed $\mathbf{z} \in V_Z$. On the other hand, with any fixed $\mathbf{y}$, $f(\mathbf{x}, \mathbf{x}-\mathbf{y}, \mathbf{z})$ is smooth with respect to $\mathbf{z} \in V_Z$, has two nonsmooth points $\mathbf{x} = \mathbf{0}$ and $\mathbf{x} = \mathbf{y}$ with respect to $\mathbf{x}$, and its partial derivatives over $\mathbf{z}$ are integrable over $\mathbf{x} \in V$ according to the assumption in Eq. (32). This meets the condition for the Leibniz integral rule and thus $F(\mathbf{y}, \mathbf{z})$ is smooth with respect to $\mathbf{z} \in V_Z$ for any fixed $\mathbf{y}$. Based on these two partial smoothness properties, we have $F(\mathbf{y}, \mathbf{z})$ to be smooth everywhere in $V \times V_Z$ except at $\mathbf{y} = \mathbf{0}$.

Lastly, we characterize the algebraic singularity of $F(\mathbf{y}, \mathbf{z})$ at $\mathbf{y} = \mathbf{0}$ by proving that there exists constants $\{C_{\boldsymbol{\alpha},\boldsymbol{\beta}}\}$ such that

$$\left|\partial_{\mathbf{y}}^{\boldsymbol{\alpha}} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} F(\mathbf{y}, \mathbf{z})\right| \leqslant C_{\boldsymbol{\alpha},\boldsymbol{\beta}} |\mathbf{y}|^{\gamma_2 - |\boldsymbol{\alpha}|}, \qquad \forall \mathbf{y} \in V \setminus \{\mathbf{0}\}, \mathbf{z} \in V_Z, \forall \boldsymbol{\alpha}, \boldsymbol{\beta} \geqslant \mathbf{0}. \tag{63}$$

Consider any $\mathbf{y} \in V \setminus \{\mathbf{0}\}$ and choose the ball domain B_σ centered at $\mathbf{y}$ with radius $\sigma = |\mathbf{y}|/2$. Using the Leibniz integral rule over variable $\mathbf{z}$, Eq. (62) can be generalized to provide the first-order partial derivatives of $\partial_{\mathbf{z}}^{\boldsymbol{\beta}} F(\mathbf{y}, \mathbf{z})$ over $\mathbf{y}$ as

$$\begin{aligned} \partial_{\mathbf{y}}^{\boldsymbol{\alpha}} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} F(\mathbf{y}, \mathbf{z}) = & \int_{V \setminus B_\sigma} \mathrm{d}\mathbf{x} \partial_{\mathbf{y}}^{\boldsymbol{\alpha}} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x}-\mathbf{y}, \mathbf{z}) - \int_{\partial B_\sigma} \mathrm{d}S \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x}-\mathbf{y}, \mathbf{z}) (\boldsymbol{\alpha} \cdot \mathbf{n}(\mathbf{x})) \\ & + \int_{B_\sigma} \mathrm{d}\mathbf{x} \partial_1^{\boldsymbol{\alpha}} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x}-\mathbf{y}, \mathbf{z}), \qquad \forall \mathbf{y} \in B_\sigma, \mathbf{z} \in V_Z, \forall |\boldsymbol{\alpha}| = 1, \boldsymbol{\beta} \geqslant \mathbf{0}. \end{aligned} \tag{64}$$

These three terms can be estimated separately based on the assumption in Eq. (32) as

$$\begin{aligned} \left| \int_{V \setminus B_\sigma} \mathrm{d}\mathbf{x} \partial_{\mathbf{y}}^{\boldsymbol{\alpha}} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x}-\mathbf{y}, \mathbf{z}) \right| & \leqslant C \int_{V \setminus B_\sigma} \mathrm{d}\mathbf{x} |\mathbf{x}|^{\gamma_1} |\mathbf{x}-\mathbf{y}|^{\gamma_2 - 1} \\ & \leqslant C \int_{V \setminus B_\sigma} \mathrm{d}\mathbf{x} |\mathbf{x}|^{\gamma_1} |\sigma|^{\gamma_2 - 1} \leqslant C |\mathbf{y}|^{\gamma_2 - 1}, \\ \left| \int_{\partial B_\sigma} \mathrm{d}S \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x}-\mathbf{y}, \mathbf{z}) (\boldsymbol{\alpha} \cdot \mathbf{n}(\mathbf{x})) \right| & \leqslant C \int_{\partial B_\sigma} \mathrm{d}S |\mathbf{x}|^{\gamma_1} |\mathbf{x}-\mathbf{y}|^{\gamma_2} \\ & \leqslant C |\mathbf{y}|^{\gamma_1} |\partial B_\sigma| \sigma^{\gamma_2} \leqslant C |\mathbf{y}|^{\gamma_1 + \gamma_2 + d - 1}, \\ \left| \int_{B_\sigma} \mathrm{d}\mathbf{x} \partial_1^{\boldsymbol{\alpha}} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x}-\mathbf{y}, \mathbf{z}) \right| & \leqslant C \int_{B_\sigma} \mathrm{d}\mathbf{x} |\mathbf{x}|^{\gamma_1 - 1} |\mathbf{x}-\mathbf{y}|^{\gamma_2} \\ & \leqslant C |\sigma|^{\gamma_1 - 1} \int_{B_\sigma} |\mathbf{x}-\mathbf{y}|^{\gamma_2} \leqslant C |\mathbf{y}|^{\gamma_1 + \gamma_2 + d - 1}, \end{aligned}$$

where all constants C's depend on $\boldsymbol{\alpha}, \boldsymbol{\beta}$, and the corresponding prefactors in Eq. (32). Using the assumption $\gamma_1 \geqslant -d + 1$, these estimates together show that the algebraic singularity characterization in Eq. (63) for $F(\mathbf{y}, \mathbf{z})$ holds true for all $|\boldsymbol{\alpha}| = 1$.

Next we prove Eq. (63) for all $|\boldsymbol{\alpha}| = 2$. For any $|\boldsymbol{\alpha}| = 2$, decompose $\boldsymbol{\alpha} = \boldsymbol{\alpha}_1 + \boldsymbol{\alpha}_2$ with $|\boldsymbol{\alpha}_1| = |\boldsymbol{\alpha}_2| = 1$ and accordingly $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} F(\mathbf{y}, \mathbf{z})$ into $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1} \left(\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_2} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} F(\mathbf{y}, \mathbf{z}) \right)$. Noting that $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_2} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} F(\mathbf{y}, \mathbf{z})$ can be expanded into three terms in Eq. (64) at any $\mathbf{y} \neq \mathbf{0}$, we consider the outer partial derivative $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1}$ applied to each of the three terms. The partial derivatives $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1}$ over the first two terms in Eq. (64) can be estimated directly as

$$
\begin{aligned}
\left| \partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1} \int_{V \setminus B_\sigma} \mathrm{d}\mathbf{x} \partial_{\mathbf{y}}^{\boldsymbol{\alpha}_2} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x} - \mathbf{y}, \mathbf{z}) \right| &= \left| \int_{V \setminus B_\sigma} \mathrm{d}\mathbf{x} \partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1 + \boldsymbol{\alpha}_2} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x} - \mathbf{y}, \mathbf{z}) \right| \\
&\leqslant C \int_{V \setminus B_\sigma} \mathrm{d}\mathbf{x} |\mathbf{x}|^{\gamma_1} |\mathbf{x} - \mathbf{y}|^{\gamma_2 - 2} \leqslant C |\mathbf{y}|^{\gamma_2 - 2}, \\
\left| \partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1} \int_{\partial B_\sigma} \mathrm{d}S \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x} - \mathbf{y}, \mathbf{z}) (\boldsymbol{\alpha}_2 \cdot \mathbf{n}(\mathbf{x})) \right| &= \left| \int_{\partial B_\sigma} \mathrm{d}S \partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} f(\mathbf{x}, \mathbf{x} - \mathbf{y}, \mathbf{z}) (\boldsymbol{\alpha}_2 \cdot \mathbf{n}(\mathbf{x})) \right| \\
&\leqslant C \int_{\partial B_\sigma} \mathrm{d}S |\mathbf{x}|^{\gamma_1} |\mathbf{x} - \mathbf{y}|^{\gamma_2 - |\boldsymbol{\alpha}_1|} \leqslant C |\mathbf{y}|^{\gamma_1 + \gamma_2 + d - 2}.
\end{aligned}
$$

For the partial derivative $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1}$ over the third term in Eq. (64), note that $\partial_1^{\boldsymbol{\alpha}_2} f(\mathbf{x}_1, \mathbf{x}_2, \mathbf{z})$ has similar non-smooth behavior as $f(\mathbf{x}_1, \mathbf{x}_2, \mathbf{z})$ described in Eq. (32) only with γ_1 changed to $\gamma_1 - 1$. We thus can apply the above derivative estimate in Eq. (63) with $|\boldsymbol{\alpha}| = 1$ for $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} \int_V \mathrm{d}\mathbf{x} f(\mathbf{x}, \mathbf{x} - \mathbf{y}, \mathbf{z})$ to estimate $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} \int_{B_\sigma} \mathrm{d}\mathbf{x} \partial_1^{\boldsymbol{\alpha}_2} f(\mathbf{x}, \mathbf{x} - \mathbf{y}, \mathbf{z})$ and obtain that

$$
\left| \partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1} \partial_{\mathbf{z}}^{\boldsymbol{\beta}} \int_{B_\sigma} \mathrm{d}\mathbf{x} \partial_1^{\boldsymbol{\alpha}_2} f(\mathbf{x}, \mathbf{x} - \mathbf{y}, \mathbf{z}) \right| \leqslant C |\mathbf{y}|^{\gamma_1 + \gamma_2 + d - 2}.
$$

Combining the above estimates of the partial derivatives $\partial_{\mathbf{y}}^{\boldsymbol{\alpha}_1}$ over the three terms in Eq. (64) together then proves the algebraic singularity characterization in Eq. (63) with $|\boldsymbol{\alpha}| = 2$. Recursively applying the above estimates, we can validate Eq. (63) for all derivative orders $\boldsymbol{\alpha} \geqslant \mathbf{0}$ and thus prove that $F(\mathbf{y}, \mathbf{z})$ is smooth everywhere in $V \times V_Z$ except at $\mathbf{y} = \mathbf{0}$ with order γ_2. This finishes the proof.