Application of the Sakurai-Sugiura Projection Method to Core-Excited-State Calculation by Time-Dependent Density Functional Theory

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Abstract: The Sakurai-Sugiura projection (SS) method was implemented and numerically assessed for diagonalization of the Hamiltonian in time-dependent density functional theory (TDDFT). Since the SS method can be used to specify the range in which the eigenvalues are computed, it may be an efficient tool for use with eigenvalues in a particular range. In this article, the SS method is applied to core excited calculations for which the eigenvalues are located within a particular range, since the eigenvalues are unique to atomic species in molecules. The numerical assessment of formaldehyde molecule by TDDFT with core-valence Becke’s three-parameter exchange (B3) plus Lee-Yang-Parr (LYP) correlation (CV-B3LYP) functional demonstrates that the SS method can be used to selectively obtain highly accurate eigenvalues and eigenvectors. Thus, the SS method is a new and powerful alternative for calculating core-excitation energies without high computation costs.

Key words: Sakurai-Sugiura projection method; diagonalization; time-dependent density functional theory; core-valence B3LYP; core-excitation energy

Introduction
Core excitations can provide valuable information regarding a specific part of a molecular system, which is not easily obtained by other means. Therefore, a wide range of core excitation studies have been carried out to elucidate local electronic structures experimentally1–7; for example, the hydrogen-bonded network structure in liquid water as investigated by X-ray absorption spectroscopy and X-ray Raman scattering has been reported to be essentially different from structures that have been proposed based on the results of conventional molecular dynamics simulations. A wide variety of studies on the hydrogen-bonded network structure have been performed since then. Considerable information has also been obtained through the application of X-ray technologies to biomolecules.5,6

Within this decade, time-dependent density functional theory (TDDFT)17–20 has become one of the most popular tools for calculating valence excited states with high accuracy and low computational cost. However, our assessment reveals that TDDFT with conventional functionals fails to give accurate excitation energies for core-excited states11; for example, Becke’s three-
parameter hybrid functional using Lee-Yang-Parr (LYP) correlation functional (B3LYP)\textsuperscript{21} significantly underestimates core-excitation energies of more than 10 eV. Based on the assessment\textsuperscript{11} and analysis of this underestimation,\textsuperscript{12} we have proposed two different kinds of functionals for core excitations: pure\textsuperscript{13} and hybrid functionals.\textsuperscript{14–16} The new hybrid functional core-valence B3LYP (CV-B3LYP)\textsuperscript{14} is useful for accurately reproducing not only core- but also valence-excitation energies. CV-B3LYP permits us to describe core-excited states at a lower cost.

Although the computational cost of TDDFT is significantly lower than those of \textit{ab initio} methods, diagonalization of the TDDFT Hamiltonian matrix for core excitations still requires high computational costs for the following reason. The conventional Davidson method\textsuperscript{22} fails to efficiently reduce computational cost for high-lying core-excited states, although it is a suitable tool for obtaining low-lying excited states, e.g., valence-valence excited ones. This has made it difficult to efficiently compute core-excited states of a large system.

To overcome this difficulty, we introduce a projection method that was recently developed by Sakurai and Sugiura (SS)\textsuperscript{23} to calculate core-excited states. The new SS method in principle permits us to obtain eigenvalues in a particular range by solving linear simultaneous equations given by the TDDFT Hamiltonian matrix. However, it has been reported that the SS method leads to unrealistic results in some occasions. Therefore, numerical applications are essential for ensuring its reliability and feasibility. Recently, application of the SS method to Hartree-Fock (HF) ground-state calculations as reported by Sakurai et al.\textsuperscript{24,25} demonstrated that the CPU time can be dramatically decreased. This article is organized as follows: first, the theories underlying the SS method and CV-B3LYP are briefly reviewed. Next, a numerical assessment of the SS method as applied to TDDFT is shown. The concluding remarks are given in the last section.

**Theory**

**Sakurai-Sugiura Method for TDDFT Calculation**

In the TDDFT scheme, the excitation energies are solutions of the pseudoeigenvalue equation:

\[
HC = \varepsilon^2 C
\]  

(1)

where \(H\) is the TDDFT Hamiltonian matrix, \(C\) is the coefficient vector, and \(\varepsilon\) is the excitation energy. The SS method requires that the domain be set first. Since the domain is arbitrary, we choose a circle with center and radius \(\gamma\) and \(R_{PS}\), respectively. Since \(\varepsilon^2\) is real because \(H\) is Hermitian, it is natural to set \(\gamma\) on a real axis. To calculate core-excited states, \(\gamma\) and \(R_{PS}\) are set to specify the domain so as to obtain desired core-excitation energies.

Let us consider a matrix pencil, \(\varepsilon^2 I - H\), where \(H \in \mathbb{C}^{n \times n}\), which is obviously regular on the complex plane. A function is defined by:

\[
f(\varepsilon) = u^H(\varepsilon I - H)^{-1}v,
\]

(2)

where \(u^H\) and \(v\) are row and column random vectors of \(\mathbb{C}^{n}\), respectively. When \(\varepsilon = \varepsilon^2\), \(f(\varepsilon)\) has poles, which are singular points of \(f(\varepsilon)\). Considering the Laurent series for \(f(\varepsilon)\) about \(\gamma\), the coefficients, \(\mu_j\), are defined by a line integral which is a generalization of Cauchy’s integral formula,

\[
\mu_j = \frac{1}{2\pi i} \int_{\Gamma} (\varepsilon - \gamma)^j f(\varepsilon) d\varepsilon = \frac{1}{N} \sum_{k=0}^{N-1} (\omega_k - \gamma)^{j+1} f(\omega_k)
\]

(3)

where \(\Gamma\) represents the domain. In eq. (3), a trapezoidal rule is used, where \(\omega_k\) are \(N\) points at an even interval on \(\Gamma\):

\[
\omega_k = \gamma + R_{PS} \exp \left(\frac{2\pi i k}{N}\right) \quad k = 0, 1, 2, \ldots, N - 1.
\]

(4)

Since \(f(\varepsilon) = \overline{f(\varepsilon)}\) for a circular domain with \(\gamma\) on a real axis, we need to calculate \(f(\omega_k)\) only \(N/2\) times instead \(N\) times by turning \(\omega_k\) up to \(\pi/N\), if \(N\) is an even number.

By defining

\[
H_m = \begin{bmatrix}
\mu_0 & \mu_1 & \cdots & \mu_{m-1} \\
\mu_1 & \mu_2 & \cdots & \mu_m \\
\vdots & \vdots & \ddots & \vdots \\
\mu_{m-1} & \mu_m & \cdots & \mu_{2m-2}
\end{bmatrix}
\]

(5)

and

\[
H_m^{*} = \begin{bmatrix}
\mu_1 & \mu_2 & \cdots & \mu_m \\
\mu_2 & \mu_3 & \cdots & \mu_{m+1} \\
\vdots & \vdots & \ddots & \vdots \\
\mu_m & \mu_{m+1} & \cdots & \mu_{2m-1}
\end{bmatrix}
\]

(6)

the following equation can be derived:

\[
\det(H_m^{*} - \lambda H_m) = 0,
\]

(7)

where \(\lambda = \varepsilon^2 - \gamma\). The detailed derivation of Eq. (7) is given in ref. 23. Equation (7) implies that the desired eigenvalues of the matrix pencil \(\varepsilon^2 I - H\) within domain \(\Gamma\) are determined by solving a generalized eigenvalue problem,

\[
H_m^{*}x = \lambda H_m x.
\]

(8)

In most cases, eq. (8) is easy to solve because \(m\), the expected number of the eigensolutions in the selected domain, is remarkably less than \(n\) in general.

Solving the generalized eigenvalue problem, eq. (8), yields approximate excitation energies \(\varepsilon^2_l = \lambda_l + \gamma\) \((l = 1, 2, \ldots, m)\). Note that when one of the excitation energies exists at the points of the circle, \(\omega_k\), the entire theory fails since \(f(\varepsilon)\) is not regular.

Corresponding eigenvectors \(q\) are determined as follows. If

\[
s_j := \int_{\Gamma} (\varepsilon - \gamma)^j (\varepsilon I - H)^{-1} v d\varepsilon = \frac{1}{N} \sum_{k=0}^{N-1} (\omega_k - \gamma)^{j+1} (\omega_k I - H)^{-1} v,
\]

(9)

then

\[
[q_1, \ldots, q_m] = [s_0, s_1, \ldots, s_{m-1}] (V_m^{*})^{-1}
\]

(10)
Application of the variational principle to Eq. (12) leads to two kinds of Fock operators:

\[ F_c = h + 2J - (a_c K_c + a_v K_{ov}) + (b_{cc} - b_{cv}) V_{xc} [\rho_c] + b_{cv} V_{xc} [\rho_v], \]

(14)

\[ F_{ov} = h + 2J - (a_{cv} K_c + a_{ov} K_{ov}) + (b_{cv} - b_{cc}) V_{xc} [\rho_{ov}] + b_{cv} V_{xc} [\rho_v]. \]

(15)

where \( \rho \) and \( J \) are 1-electron and Coulomb operators, HF-exchange operators and the first derivatives of \( E_{xc} \) are defined as:

\[ K_c = \sum_k^{c} K_k, \quad K_{ov} = \sum_m^{ov} K_m, \]

\[ V_{xc} [\rho] = \delta E_{xc} [\rho] / \delta \rho, \quad V_{xc} [\rho_c] = \delta E_{xc} [\rho_c] / \delta \rho_c, \quad V_{xc} [\rho_{ov}] = \delta E_{xc} [\rho_{ov}] / \delta \rho_{ov}. \]

To guarantee invariance under unitary transformation between core and occupied valence orbitals, we used Roothaan’s coupling-operator technique,\(^{11,13-15}\) which is often adopted in restricted open-shell HF calculations. Further details of CV-B3LYP and TDDFT procedures with CV-B3LYP are given in ref. 14.

Numerical Assessments

We coded TDDFT and the SS method into the GAMESS program.\(^{34}\) For numerical stability in the SS method, a given problem was scaled so that the radius of the domain, \( R_D = |\alpha_k - \gamma| \), becomes one.

Formaldehyde

First, numerical assessments for a formaldehyde molecule were performed while changing the domain. The geometry optimized by B3LYP with the correlation-consistent polarized valence double-\( \zeta \) (cc-pVDZ) basis set\(^ {35} \) was used. We performed TDDFT calculations at the CV-B3LYP/cc-pVDZ level, in which the dimension of the TDDFT Hamiltonian is 256.

As mentioned earlier, because of its strongly localized nature, excitation energies from core orbitals are always observed within a particular range, which strongly depends on the element. Figure 1 shows the excitation energies of formaldehyde calculated by the usual direct diagonalization scheme.

With the SS method, valence- and core-excitation energies from C1s and O1s orbitals can be obtained selectively if three domains are set. We chose the ranges to be 0–10, 285–295, and 530–540 eV for valence-, C1s-, and O1s-excitation energies, respectively, which are presented by longer black lines in Figure 1. \( N \) and \( m \) were set to 128 and 5.

The excitation energies obtained by the SS method are shown in Table 1. The real parts of each of the four eigenvalues obtained by the SS method agree with the results from direct
diagonalization are less than 1.0. These correct eigenvalues have significantly small imaginary parts and thus can be considered true excitation energies, since excitation energies should be real. On the other hand, eigenvalues with large imaginary parts and those outside the chosen range are not only physically meaningless but also mathematically incorrect, i.e., eigenvalues of Eq. (7) are real in principle since \( \{ \mu_i \} \) defined by Eq. (4) are real and their imaginary parts come from integration errors. Since, as mentioned, the errors between real parts of core eigenvalues and excitation energies obtained from direct diagonalization are less than 1.0 \( \times \) 10\(^{-3} \) eV, eigenvalues with imaginary parts greater than 1.0 \( \times \) 10\(^{-3} \) eV are considered to be incorrect eigenvalues in this work. Hence, the 5th, 10th, and 15th eigenvalues should be excluded from the results. Therefore, imaginary parts of the eigenvalues should become a good indicator to specify incorrect solutions.

Table 1 summarizes the valence-, C1s-, and O1s-excitation energies obtained by calculations with the SS method. This table also shows experimental excitation energies. Only desired excitation energies are obtained with the SS method. The results agree well with experimental values because of the good description of the CV-B3LYP functional for both valence and core excitations.

### Nucleobases

Next, N1s excitations of cytosine and uracil molecules (shown in Fig. 2), which are bases of deoxyribonucleic acid (DNA) and ribonucleic acid (RNA), respectively, were assessed. Since X-ray absorption spectra of DNA or nucleobases are related to the breaking of bonds in DNA or damage to DNA by X-rays, they have attracted considerable attention. However, these spectra often give complex peaks which are difficult to assign and analyze. Hence, it would be practical if assignment could be done by quantum chemical calculation.

The geometries of cytosine and uracil molecules are optimized at the B3LYP/6-311G** level. We performed TDDFT calculations with the SS method at the CV-B3LYP/cc-pVDZ level and chose 398–403 eV as the range for excitation energies, which corresponds to the N1s excitations. \( N \) and \( m \) were set to 128 and 10, respectively. The dimensions of the TDDFT Hamiltonians for cytosine and uracil molecules are 3364 and 3219, respectively.

### Table 1. Excitation Energies of Formaldehyde Obtained by the Combination of CV-B3LYP and the SS Method in eV. The ranges chosen are 0–10, 285–295, and 530–540 eV, which are of valence-, C1s-, and O1s-excitation energies, respectively.

<table>
<thead>
<tr>
<th>Range</th>
<th>Excitation energy</th>
<th>Direct</th>
<th>SS method</th>
</tr>
</thead>
<tbody>
<tr>
<td>0–10</td>
<td></td>
<td>4.0513</td>
<td>4.0513 + 3.5003 ( \times 10^{-11} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>7.9656</td>
<td>7.9656 + 2.3470 ( \times 10^{-9} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>9.1989</td>
<td>9.1989 + 2.6504 ( \times 10^{-8} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>9.4426</td>
<td>9.4426 + 1.2046 ( \times 10^{-8} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>–</td>
<td>10.2005 – 1.1495 ( \times 10^{-4} ) i</td>
</tr>
<tr>
<td>285–295</td>
<td></td>
<td>286.6949</td>
<td>286.6950 + 2.1475 ( \times 10^{-15} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>290.9542</td>
<td>290.9543 – 1.5584 ( \times 10^{-13} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>292.3777</td>
<td>292.3779 – 1.2585 ( \times 10^{-12} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>294.3121</td>
<td>294.3122 – 1.5296 ( \times 10^{-13} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>–</td>
<td>294.3310 + 6.8903 i</td>
</tr>
<tr>
<td>530–540</td>
<td></td>
<td>532.0208</td>
<td>532.0210 + 3.9732 ( \times 10^{-13} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>536.6519</td>
<td>536.6521 + 2.4493 ( \times 10^{-13} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>538.3412</td>
<td>538.3415 + 7.5124 ( \times 10^{-13} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>540.0163</td>
<td>540.0165 – 3.3240 ( \times 10^{-14} ) i</td>
</tr>
<tr>
<td></td>
<td></td>
<td>–</td>
<td>537.5492 + 2.9505 i</td>
</tr>
</tbody>
</table>

### Table 2. Summarized Results of Valence-, C1s-, and O1s-Excitation Energies of Formaldehyde Calculated by the Combination of CV-B3LYP and the SS Method in eV.

<table>
<thead>
<tr>
<th>Main configuration</th>
<th>SS method</th>
<th>(Deviation)</th>
<th>Exptl.</th>
</tr>
</thead>
<tbody>
<tr>
<td>( n \rightarrow \pi^* )</td>
<td>4.0513</td>
<td>(+0.11)</td>
<td>3.94*</td>
</tr>
<tr>
<td>( \sigma \rightarrow \pi^* )</td>
<td>7.9656</td>
<td>(–)</td>
<td></td>
</tr>
<tr>
<td>( n \rightarrow \sigma^<em>, \pi \rightarrow \pi^</em> )</td>
<td>9.1989</td>
<td>(+0.52)</td>
<td>8.68*</td>
</tr>
<tr>
<td>( C1s \rightarrow \pi^* )</td>
<td>286.6950</td>
<td>(+0.69)</td>
<td>286.01*</td>
</tr>
<tr>
<td>( O1s \rightarrow \pi^* )</td>
<td>532.0210</td>
<td>(+1.22)</td>
<td>530.80*</td>
</tr>
</tbody>
</table>

Deviations from the experimental excitation energies are shown in parentheses.

*Reference 36.

*Reference 37.
The results are listed in Table 3 along with previously reported experimental data. The chemical shifts from the lowest N1s-excitation energy are given in parentheses. The label numbers of N atoms are shown in Figure 2. The calculated excitation energies of both molecules agree well with experimental spectra with an error of at most 1 eV. In a cytosine molecule, however, the experimental assignments in ref. 40 are different from those obtained by the present calculation. Because of the sp² character of the imine nitrogen of N(2) in a cytosine molecule, the calculated excitation energies from N(1), 401.47 eV, and N(3), 401.21 eV, are higher than that from N(2), 399.40 eV. Thus, core-excitation spectra can be assigned correctly by using CV-B3LYP with the SS method.

Finally, we should mention the computational cost of the present method as compared with the direct diagonalization method. To reduce the computational cost to solve linear equation, we have utilized sparse matrix linear equation solver PARDISO. Matrix elements less than the threshold \( \theta = 2.0 \times 10^{-2} \) au were set to be zero. The central processing unit (CPU) times for the diagonalization procedure in the TDDFT calculation of a cytosine molecule, which were measured on an Intel Xeon/2.66 GHz processor, were 372.9 and 9.3 s for direct and SS methods, respectively. Differences of excitation energies were still less than 1.0 \( \times 10^{-3} \) eV. The dependence of the accuracy and CPU time on the threshold \( \theta \) is discussed in the Appendix. We confirmed that the present method can drastically reduce the computational cost for obtaining core-excitation energies with reasonable accuracy and apply to large systems such as biomolecules and nanomaterials.

**Concluding Remarks**

As an alternative to the direct and/or Davidson diagonalization methods, we have implemented and numerically assessed the SS method for diagonalization of the TDDFT Hamiltonian. The SS method was applied to calculations of core-excited states of formaldehyde and successfully gave core-excitation energies in two selective ranges: 285–295 and 530–540 eV for C1s and O1s excitations, respectively. The deviations of the core-excitation energies do not exceed 1.0 \( \times 10^{-3} \) eV, which is accurate enough for analyzing the chemical shifts of core excitations. Thus, the SS method combined with TDDFT using CV-B3LYP allows us to estimate core-excitation energies selectively and accurately without calculating unnecessary excited states. Furthermore, we have demonstrated that the present scheme can be used to satisfactorily assign core-excitation spectra of cytosine and uracil molecules.

We think that core-excited states can be calculated more efficiently and speedily by parallel execution of the SS method, and we should also be able to compute larger systems, which have not been investigated. Calculations for core-excited states of a significantly large system using the SS method are in progress.

**Acknowledgments**

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<table>
<thead>
<tr>
<th>Molecule</th>
<th>Main configuration</th>
<th>Present</th>
<th>Exptl. a</th>
<th>Exptl. b</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cytosine</td>
<td>N(2)1s → π₁*</td>
<td>399.40</td>
<td>(0.00)</td>
<td>400.68</td>
</tr>
<tr>
<td></td>
<td>N(2)1s → π₂*</td>
<td>401.04</td>
<td>(1.65)</td>
<td>402.00</td>
</tr>
<tr>
<td></td>
<td>N(3)1s → π₁*</td>
<td>401.21</td>
<td>(1.81)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>N(1)1s → π₁*</td>
<td>401.47</td>
<td>(2.07)</td>
<td>400.4</td>
</tr>
<tr>
<td></td>
<td>N(2)1s → σ₁*</td>
<td>402.20</td>
<td>(2.81)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>N(2)1s → σ₂*</td>
<td>402.50</td>
<td>(3.11)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>N(3)1s → π₂*</td>
<td>402.85</td>
<td>(3.45)</td>
<td>404.2</td>
</tr>
<tr>
<td>Uracil</td>
<td>N(2)1s → π₁*</td>
<td>401.53</td>
<td>(0.00)</td>
<td>402.49</td>
</tr>
<tr>
<td></td>
<td>N(1)1s → π₁*</td>
<td>401.94</td>
<td>(0.41)</td>
<td>401.9</td>
</tr>
<tr>
<td></td>
<td>N(2)1s → π₂*</td>
<td>402.83</td>
<td>(1.31)</td>
<td>403.5</td>
</tr>
</tbody>
</table>

aEnergies taken from ref. 39.
bEnergies and assignments taken from ref. 40.
Table A1. Threshold Dependence of N1s-Excitation Energies of a Cytosine Molecule Calculated by the Present Method in eV. Numbers of nonzero elements and CPU times are shown together.

<table>
<thead>
<tr>
<th>No.</th>
<th>Main configuration</th>
<th>Direct</th>
<th>( \theta = 5 \times 10^{-3} )</th>
<th>( \theta = 8 \times 10^{-3} )</th>
<th>( \theta = 2 \times 10^{-2} )</th>
<th>( \theta = 5 \times 10^{-2} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2885</td>
<td>N(2)1s ( \rightarrow \pi_1^* )</td>
<td>399.3949</td>
<td>399.3950</td>
<td>399.3950</td>
<td>399.3950</td>
<td>399.7305</td>
</tr>
<tr>
<td>2886</td>
<td>N(2)1s ( \rightarrow \pi_2^* )</td>
<td>401.0401</td>
<td>401.0402</td>
<td>401.0402</td>
<td>401.0403</td>
<td>401.1338</td>
</tr>
<tr>
<td>2887</td>
<td>N(3)1s ( \rightarrow \pi_1^* )</td>
<td>401.2082</td>
<td>401.2083</td>
<td>401.2083</td>
<td>401.2087</td>
<td>401.9934</td>
</tr>
<tr>
<td>2888</td>
<td>N(1)1s ( \rightarrow \pi_1^* )</td>
<td>401.4650</td>
<td>401.4651</td>
<td>401.4651</td>
<td>401.4651</td>
<td>–</td>
</tr>
<tr>
<td>2889</td>
<td>N(2)1s ( \rightarrow \sigma_1^* )</td>
<td>402.2029</td>
<td>402.2030</td>
<td>402.2030</td>
<td>402.2032</td>
<td>–</td>
</tr>
<tr>
<td>2890</td>
<td>N(2)1s ( \rightarrow \sigma_2^* )</td>
<td>402.5038</td>
<td>402.5039</td>
<td>402.5040</td>
<td>402.5047</td>
<td>–</td>
</tr>
<tr>
<td>2891</td>
<td>N(3)1s ( \rightarrow \sigma_2^* )</td>
<td>402.8487</td>
<td>402.8488</td>
<td>402.8487</td>
<td>402.8488</td>
<td>–</td>
</tr>
</tbody>
</table>

No of nonzero elements CPU time (s) 520,934 373 238,218 427 57,853 222 23,201 9 402,5039 401.4651 402.2030 402.2032 – 402.5047 – – – 3

*Only three eigenvalues could be obtained in the range of 398–403 eV and their excitation natures were incorrect.

Appendix A: Dependence of the Accuracy and CPU Time on Threshold \( \theta \)

The computational costs for obtaining N1s excitation energies of a cytosine molecule were examined by varying the threshold \( \theta \). Table A1 shows the computational costs together with obtained excitation energies. For the present method with \( \theta \leq 2 \times 10^{-2} \), the excitation energies agree with those by the direct diagonalization within \( 10^{-3} \) eV errors. However, the usage of a larger threshold leads to an incorrect result. The CPU times become shorter than the direct diagonalization for \( \theta \geq 8 \times 10^{-2} \). Therefore, the choice of the threshold \( \theta \) should be a trade-off between the accuracy and the computational cost.

References