Uniqueness of the solution of the contracted Schrödinger equation

Koji Yasuda

Graduate School of Human Informatics, Nagoya University, Chikusa-ku, Nagoya 464-8601, Japan

In this paper two fundamental questions in the contracted Schrödinger equation (CSE) approach are considered by using Lipkin's quasispin model: 1-1 mapping between the second-order reduced density matrix (2-RDM) and the wave function of an excited state, and the uniqueness of the solution of CSE under incomplete N-representability conditions. We present some examples of the wave functions that give the same 2-RDM as the excited state. Thus 2-RDM of an excited state does not determine the wave function uniquely, and it alone cannot be used as basic variable for excited states of the density-matrix theory. Under the incomplete representability constraints the solution of the second-order CSE contains all the exact 4-RDMs together with the spurious ones. We examined the distribution of the solutions as a function of energy, and found that the solutions are well separated from each other under the P- and G-representability conditions of 4-RDM in the low-energy region, but with moderate interaction, or in the higher-energy region, there exist spurious solutions for almost all energies. Thus the G condition of 4-RDM is not sufficient to solve the excited states, although it gives accurate results for the ground state of Lipkin's model.

I. INTRODUCTION

In this paper we consider the uniqueness of the solution of the many-body theory based on the density equation or the contracted Schrödinger equation, which uses the reduced density matrices (RDMs) as the basic variable instead of the wave function. Although the wave function contains all the accessible information of a many-body quantum system, it is a complicated function that is available only for a simple system. Since all the relevant physical information is contained in the RDM, a many-body theory that avoids such complexity has been searched for a long time [1–10]. The present density equation theory, which is an example of such a theory, tries to determine the nth-order RDM (n-RDM) that is a simpler function than the wave function. The n-RDM is defined as [11]

$$\Gamma^{(n)}(x_1'\cdots x_n', x_1\cdots x_n)$$

$$= \frac{1}{n!}\langle \Psi | \phi^\dagger(x_1)\cdots\phi^\dagger(x_n)\phi(x_n')\cdots\phi(x_1')|\Psi\rangle, \quad (1.1)$$

where $\phi^\dagger$ (ϕ) and $x_i \equiv r_i\sigma_i$ denote the creation (annihilation) field operator and the set of the spatial and spin coordinates of an electron, respectively.

The basic equation in our theory is the Schrödinger equation in RDM form, which is called the density equation or the contracted Schrödinger equation (CSE) [12,13],

$$\left\{ \sum_i^n v(r_i) + \sum_{i>j}^n 1/r_{i,j} - E \right\} \Gamma^{(n)}(x_1'\cdots x_n'; x_1\cdots x_n)$$

$$+ (n+1)\int dx_{n+1}\left\{ v(r_{n+1}) + \sum_i^n 1/r_{i,n+1} \right\}$$

$$\times \Gamma^{(n+1)}(x_1'\cdots x_n', x_{n+1}; x_1\cdots x_n, x_{n+1})$$

$$+ \frac{(n+1)(n+2)}{2}\int dx_{n+1}dx_{n+2}/r_{n+1,n+2}$$

$$\times \Gamma^{(n+2)}(x_1'\cdots x_n', x_{n+1}, x_{n+2}; x_1\cdots x_{n+2}) = 0, \quad (1.2)$$

where v is the one-body operator in the Hamiltonian. Clearly the exact n-RDMs satisfy this equation. More importantly as proved by Nakatsuji, the second-order CSE [Eq. (1.2) with $n=2$] is still equivalent to the original Schrödinger equation in the representable density-matrix space [13], though the domain of the ensemble-representable density matrices is much larger than the antisymmetric wave functions. Hence we can obtain RDMs without using the wave function by solving this equation. Unfortunately the number of unknowns in the 4-RDM is greater than the number of conditions in the 2-CSE, and the equation itself is underdetermined [14]. This prevents us from solving the equation directly. Other conditions that make the solution unique are provided by the representability conditions of the 4-RDM, but these conditions are still unknown today [11]. This is in sharp contrast to the traditional wave-function approach: wave functions contain much redundant information but satisfy simple boundary conditions (symmetry or antisymmetry upon particle permutations), while density matrices eliminate the redundant information but satisfy complicated and unknown boundary conditions (N-representability conditions).

There are two different approaches to solve this equation [3]. The first one takes the 2-RDM as the basic variable and reconstructs 3- and 4-RDMs from the 2-RDM to eliminate the indeterminacy. This approach was first demonstrated by Valdemoro and co-workers. The fundamental question in this functional approach is the existence of the reconstruction functional. As pointed by Mazziotti [5], Rosina's theorem [15] rationalizes the reconstruction of the higher-order RDMs from the 2-RDM of the ground state.

Valdemoro approximated the functional by a heuristic approach based on the particle-hole duality [2], and we derived approximated functionals systematically by the Green's-

function method [3]. Mazziotti transcribed the product writable term in these functionals concisely with cumulants [6,7]. He also determined the connected n-RDM by requiring that the $(n+1)$-RDM approximated by his cumulant expansion reduces to the original n-RDM by contraction [6,7].

The Green's-function method we employed is so powerful that one can derive the approximation by Valdemoro and co-workers. In addition, Mazziotti's functional based on the cumulant expansion and the contraction requirement is essentially equivalent to, or the subset of, the functional by the Green's-function method [3] and the infinite sum of the selected diagrams called the Parque sum [4]. In the Green's-function method, the formula of the connected n-RDM is derived with the $(n-1)$th-order perturbation theory. Hence the contraction relation is satisfied within the accuracy of the $(n-1)$th-order of the perturbation. On the other hand a certain portion of $(n+1)$-RDM always vanishes by the contraction. Such terms first appear in the $(n-1)$th-order of the perturbation. Hence the missing terms in Mazziotti's approximation are of the same order as those of the Green's-function method.

Compared with the ground state, there remain many unresolved problems in the functional method of excited states. Mazziotti claimed that Rosina's theorem can be extended to excited states, and proposed the ensemble representability method (ERM) to reconstruct the higher-order RDMs from the 2-RDMs of the ground and the excited states [5]. However a question is raised about the validity of this theorem [16]. The first aim of this paper is to examine the 1-1 mapping theorem between the 2-RDM and the wave function of the excited state. In Sec. II, we examine this relationship for Lipkin's quasispin model [17]. We found that there is no such mapping between the 2-RDM of the excited state and the wave function, and that Mazziotti's theorem is not correct. That is, the functional method that uses only the 2-RDM as a basic variable cannot be an exact theory for excited states.

The second approach to solve the 2-CSE, which was originally proposed by Nakatsuji, uses the 4-RDM as a basic variable and imposes some known representability constraints. This procedure yields all the exact solutions together with the spurious ones. One must then distinguish the unphysical ones [13]. However Harriman pointed out that this procedure may be impossible. Since the 2-CSE in terms of the 4-RDM is a system of linear equations and the known representability conditions can be regarded as linear ones, there are always innumerable number of spurious solutions near the exact ones. The CSE cannot yield discrete solutions, and it may be impossible to distinguish the correct one [14].

Mazziotti was the first person to apply the representability method to a simple many-body system. He solved the 2-CSE for Lipkin's model [5–8]. The 4-RDMs of the excited states were calculated by imposing non-negativity, which is the most important representability condition known today [5]. Unfortunately since he did not analyze the uniqueness of the solution, it is not clear whether his result is solely determined by the equation or it is also affected by the artifacts of initial guess and the iteration procedure. Recently Nakata and Nakatsuji solved the 1-CSE to calculate the ground-state

3-RDM of atoms and molecules under some representability conditions of the 3-RDM [10]. They tried to eliminate the nonuniqueness of the solution by minimizing the energy expectation value. The accuracy and the variety of their result stimulate us to extend the representability approach to excited states.

The second aim of this paper is to examine the distribution of the solutions of CSE as a function of energy under some incomplete representability conditions. In Sec. III we analyze the properties of the 4-RDM of Lipkin's model and give the explicit formula of it. Then we examine numerically the distribution of the solutions under the P- and G-representability conditions. We found that the distribution heavily depends on the interaction strength and the representability conditions imposed. Solutions are well separated from each other in the lower-energy region near the ground state if we impose the P or G condition. However many spurious solutions appear for almost all energies in the higher-energy region under the moderate interaction. P and G conditions are not sufficient for the excited states of Lipkin's model.

The third aim of this paper is to analyze the RDMs of Lipkin's model in detail. While many numerical results were presented in a series of papers [5–8], analytical properties of these RDMs were never reported. In this paper we will present analytical expressions of these RDMs and show how simple their structures are compared with those of atoms and molecules.

II. DOES THE 2-RDM OF AN EXCITED STATE DETERMINE THE WAVE FUNCTION?

In this section we consider the question of whether the second-order reduced density matrix (2-RDM) of an excited state contains enough information to determine the wave function. This question is of essential importance for a recent attempt to establish a quantum-mechanical approach based on 2-RDM instead of the wave function [2–9]. However the answer of this question is not yet known today.

About 30 years ago, Rosina proved that the ground-state 2-RDM uniquely determines the wave function using the variational principle of the ground-state energy [15]. Recently Mazziotti claimed that the 2-RDM of an excited state uniquely determines the wave function (Theorem 2 in Ref. [5]). Based on this theorem he proposed the ERM, which determines the higher-order RDMs from the 2-RDM of an excited state. He also applied ERM to Lipkin's quasispin model [17]. This ERM searches for a higher-order RDM that satisfies some representability conditions [11] and at the same time reduces to the given 2-RDM by contraction. His Theorem 2 ensures that ERM yields the exact higher-order RDMs when the complete N-representability conditions are imposed.

Mazziotti's proof of the theorem for a nondegenerate system reads that eigenfunctions that belong to different energy levels do not give the same 2-RDM, but this statement is almost trivial. The problem of this proof is that it could not exclude the possibility that the two wave functions, an excited state of a two-body Hamiltonian and another antisym-

TABLE I. Classification of the elements of n-RDM in Lipkin's quasispin model. Expectation values of operators in the first $(n+1)$ rows in this table cover all the elements of n-RDM. Due to the symmetry of this model, independent elements are those in the first and the second column.

Independent operators	Removable by CSE	Redundant due to symmetry	Constants of motion	Identically zero
			1	
J_z				J_+
J_z^2, H			J^2	$J_z J_+$
	$J_z^3, J_z H$	$J_z J^2$		$J_z^2 J_+, J_+ H, J_+ J^2$
J_z^4	$J_z^2 H, H^2$	$J_z^2 J^2, J^2 H$	J^4	$J_z^3 J_+, J_z J_+ H, J_z J_+ J^2$

metric wave function, give the same 2-RDM. If this is possible, the ERM for excited states does not yield the exact higher-order RDMs even if we impose the complete N-representability conditions. In this section we present some counterexamples of this theorem using Lipkin's model. Our conclusion is that other antisymmetric wave functions also give the same 2-RDM as the excited state, and hence it does not determine the wave function uniquely.

A. Lipkin's quasispin model

N spinless fermions that obey the Hamiltonian of Eq. (2.1) constitute Lipkin's model,

$$H = \frac{1}{2} \sum_{\sigma} \sum_{p=1}^{N} \sigma a_{p\sigma}^{\dagger} a_{p\sigma} + \frac{V}{2} \sum_{\sigma} \sum_{p,p'=1}^{N} a_{p\sigma}^{\dagger} a_{p'\sigma}^{\dagger} a_{p'-\sigma} a_{p-\sigma}. \quad (2.1)$$

Here σ takes the values of ± 1. Using the angular-momentum operators

$$J_z = \frac{1}{2} \sum_{p\sigma} \sigma a_{p\sigma}^{\dagger} a_{p\sigma}, \quad J_+ = \sum_{p} a_{p+1}^{\dagger} a_{p-1}, \quad J_- = J_+^{\dagger},$$

this Hamiltonian can be written as

$$H = J_z + \frac{V}{2}(J_+^2 + J_-^2). \quad (2.2)$$

Lipkin's model has three kinds of conserved quantities: the total angular momentum J^2, parity π, and the number of particles in the pth site n_p,

$$J^2 = J_+ J_- + J_z^2 - J_z, \quad (2.3a)$$

$$\pi = (-1)^{J_z + J}, \quad (2.3b)$$

$$n_p = \sum_{\sigma} a_{p\sigma}^{\dagger} a_{p\sigma}. \quad (2.3c)$$

Thus the N-particle wave function can be expressed by the angular-momentum eigenfunctions $|J,J_z\rangle$ of $J = N/2$ and $J_z = -J,\ldots,J$ [17]. Only even and odd values of J_z are neces-

sary for $\pi = +1$ and -1, respectively. Because of this special symmetry, the degree of freedom in the N-particle wave function [that is, the full-CI (configuration-interaction) dimension] is at most $(N+1)$.

Next, let us consider the independent 2-RDM elements of an eigenstate of the Lipkin's model. Due to the symmetry of this model, the condensed 2-RDM defined by Eq. (2.4) covers all the independent elements [5],

$$D_{\sigma_1'\sigma_2'}^{\sigma_1\sigma_2} = \frac{1}{2} \sum_{p_1 p_2} \langle \Psi | a_{p_1\sigma_1}^{\dagger} a_{p_2\sigma_2}^{\dagger} a_{p_2\sigma_2'} a_{p_1\sigma_1'} | \Psi \rangle. \quad (2.4)$$

Since this condensed 2-RDM has the symmetry $D_{ij}^{kl} = D_{kl}^{ij} = D_{ji}^{lk}$, one finds that the number of independent 2-RDM elements is at most seven, *irrespective of the number of the particles involved*. This number is the same as that in the simplest model in quantum chemistry: the H_2 molecule with a minimal basis set. The remarkable feature of this model is that the degrees of freedom in the wave function and 2-RDM are at most $(N+1)$ and 7, respectively. This simplicity enables us to obtain the exact solution.

The condensed 1-RDM has three independent elements D_+^+, D_-^+, and D_-^-, which can alternatively be expressed as $\langle \hat{1} \rangle = (D_+^+ + D_-^-)/N$, $\langle \hat{J}_z \rangle = (D_+^+ - D_-^-)/2$, and $\langle \hat{J}_+ \rangle = D_-^+$. Similarly we express the independent 2-RDM elements as the expectation values of the seven operators listed in the first three rows of Table I: 1, J_z, J_+, J_z^2, $J_+ J_z$, H, and J^2. The expectation values calculated from an eigenfunction of Lipkin's model are $\langle J_+ \rangle = \langle J_+ J_z \rangle = 0$ and $\langle J^2 \rangle = J(J+1)$ due to the conservation of the total angular momentum and the parity, and $\langle 1 \rangle = 1$ due to the normalization condition. Thus the number of independent 2-RDM elements is at most three irrespective of the number of the particles involved. This spin representation of RDMs provides us with a very concise way to analyze Lipkin's model.

B. 2-RDM of the excited states of Lipkin's model

Let us calculate the eigenvalues and normalized eigenfunctions of Lipkin's model with $N = 4$ and parity $+1$. Using the angular-momentum eigenfunctions of $|2, 2\rangle$, $|2, 0\rangle$, and $|2, -2\rangle$ as bases, we obtain the following eigenvalues and eigenfunctions:

$$\Psi_0 = \frac{1}{\sqrt{2\alpha}}(-\sqrt{3}V, \sqrt{2}, \sqrt{3}V), \quad E_0 = 0, \qquad (2.5)$$

$$\Psi_{\pm 2} = \frac{1}{2\sqrt{\alpha}}(\sqrt{\alpha}\pm 1, \pm\sqrt{6}V, \sqrt{\alpha}\mp 1),$$

$$E_{\pm 2} = \pm 2\sqrt{\alpha}, \qquad (2.6)$$

$$\alpha = 1 + 3V^2.$$

Since $\Psi_{\pm 2}$ correspond to the largest and the smallest eigenvalues, respectively, their 2-RDMs uniquely determine the wave function due to Rosina's theorem. Now consider the 2-RDMs of Ψ_0 and the trial function χ defined by Eq. (2.7),

$$\chi = \sqrt{1-a^2}\Psi_0 + \frac{a}{\sqrt{2}}(\Psi_{-2} - \Psi_{+2})$$

$$= (-\sqrt{3[1-a^2]}V - a, \sqrt{2[1-a^2]} - \sqrt{6}Va,$$

$$\sqrt{3[1-a^2]}V + a)/\sqrt{2a}. \qquad (2.7)$$

Two vectors Ψ_0 and χ are not the same for $a \neq 0$, because the eigenfunctions Ψ_k are linearly independent. Both Ψ_0 and χ give the same expectation values of $\langle J_z \rangle = 0$ and $\langle H \rangle = 0$. The expectation values of J_z^2 are

$$\langle \Psi_0 | J_z^2 | \Psi_0 \rangle = \frac{12V^2}{1+3V^2}, \qquad (2.8)$$

$$\langle \chi | J_z^2 | \chi \rangle = \frac{4\{\sqrt{3(1-a^2)}V + a\}^2}{1+3V^2}. \qquad (2.9)$$

If they have the same value, Ψ_0 and χ give the same 2-RDM. This occurs when

$$a^{-1} = \pm\sqrt{\frac{1}{12}(3V - V^{-1})^2 + 1}. \qquad (2.10)$$

Positive and negative solutions should be taken for $V > 1/\sqrt{3}$ and $V < 1/\sqrt{3}$, respectively. Thus the 2-RDM of the excited state does not determine the wave function uniquely.

Now it is clear why Rosina's one-to-one mapping exists for ground states but not for excited states. In the case of excited states (such as Ψ_0 here), one can always add a higher- and a lower-energy eigenstate (Ψ_{+2} and Ψ_{-2}) to obtain a new function χ with the same energy as Ψ_0. This is necessary for these wave functions to give the same 2-RDM. For the ground state, however, this is not possible, because there are no lower-energy eigenfunctions of the Hamiltonian.

There is no need for three eigenfunctions in our trial function χ to belong to the same Hamiltonian as the target state Ψ_0. This trial function can be constructed from the eigenfunctions of another Hamiltonian. For example, the trial function $\chi' = 1/\sqrt{2}(\Psi'_{-2} - \Psi'_{+2})$, where $\Psi'_{\pm 2}$ are the eigenfunctions of Lipkin's model with interaction strength $1/(3V)$, gives the same 2-RDM as that of Ψ_0. Moreover, there exist innumerable wave functions giving the same

2-RDM. The expectation value $\langle \Psi_0 | J_z^2 | \Psi_0 \rangle$ of Eq. (2.8) is in the range 0–4 and is a monotonically increasing function of V. On the other hand, the function $f(a,U)$,

$$f(a,U) = \frac{4\{\sqrt{3(1-a^2)}U + a\}^2}{1+3U^2}, \qquad (2.11)$$

has a maximum $f = 4$ at $a_{max} = (1+3U^2)^{-1/2}$, minimum $f = 0$ at $a_{min} = -\sqrt{3}(1+3U^2)^{-1/2}$. Since $0 < f < 4$ for $a \neq a_{max}$ or a_{min}, we can always select V for Ψ_0 to give the same 2-RDM as χ. That is, there exist innumerable wave functions that give the same 2-RDM.

C. Ensemble representability method for excited states

Having established this result, one may have a doubt about the meaning of the ERM for excited states. The 2-RDM of an excited state does not have enough information to specify the wave function uniquely, and there are many wave functions that give the same 2-RDM. These wave functions generally give different 4-RDMs from that of the excited state under consideration. This is because these trial functions differ from the target state and hence their 4-RDMs do not satisfy the 2-CSE [13]. Our Ψ_0 and χ provide an example, since the 4-RDM is equivalent to the wave function in the four-particle system. Therefore it is hopeless to reconstruct higher-order RDMs including 4-RDM from the 2-RDM of the excited state alone, even if we know the complete representability conditions.

Mazziotti applied the ERM to the excited states of Lipkin's model. The results are summarized in Tables II and IV of Ref. [5]. We first point out that the states of 4_2, 6_2, 8_2, 10_2, 15_2, and 25_2 in these tables are not excited states but ground states with different parity. Notation N_k indicates the kth eigenfunction of Lipkin's model with N particles. The ERM could be applied to these states because Rosina's theorem ensures the uniqueness of the reconstruction. As shown in Table II, ERM gives the exact result for the 4_2 state. This is because the 4_2 state is the ground state of parity -1, and the 4-RDM is equivalent to the wave function for this four-particle system. It does not mean that the ERM gives exact results even for excited states if we impose the complete N-representability conditions. The results of the true excited states, 10_3 and 25_3 in Table II, depend on the artifacts of initial guess and the iterative solution method.

The ERM for excited states should be understood as a method to realize Nakatsuji's original proposal [13] to solve the CSE under known representability conditions using the RDM as a basic variable. It is interesting that the ERM+CSE gives good results shown in Tables II and IV. However, as Harriman pointed out [14], the solution of this method may not be even unique. In the following section we examine the uniqueness of these solutions.

Before closing this section we will pay attention to the "complete reconstruction method" [7] proposed by Mazziotti for ground states. This method tries to determine the exact higher-order RDMs from the exact 2-RDM of the ground state. Rosina's theorem indicates that there exists only one representable N-RDM that gives the 2-RDM by contraction and that there exist such functionals for the

TABLE II. Energies of the ground-, first-, and second-excited states of Lipkin's quasispin model calculated by the second-order contracted Schrödinger equation compared with the coupled cluster and the exact ones. The results of the P and G conditions show the upper- and the lower-energy regions where solutions are found. NA denotes data not available.

Number of particles	Potential	Correlation energy (%)	P condition	G condition	Variational CCSD	CCSDTQ [21]	Exact
14	0.3571	59.46	−17.8134, 17.8134	−17.2778, −16.9855	−16.7136	−17.2660	−17.2684
				−11.5852, 11.5852		−11.0760	−17.2638
						−6.2530	−11.1850
	0.07143	3.71	−7.3554, 7.3554	−7.2700, −7.2699	−7.2696	−7.2533	−7.2700
				−6.6220, −6.5073		−6.6023	−6.6219
				−5.7729, 5.7729		−5.6843	−5.7167
	0.05714	2.27	−7.1994, −7.0598	−7.1631, −7.1631	−7.1631	−7.1706	−7.1631
			−6.7380, 6.7380	−6.3933, −6.3303		−6.4006	−6.3933
				−5.5040, 5.5040		−5.4806	−5.4745
	0.04615	1.45	−7.1177, −7.0757	−7.1029, −7.1029	−7.1029	NA	−7.1029
			−6.4169, 6.4169	−6.2542, −6.2230		NA	−6.2542
				−5.3300, 5.3300		NA	−5.3153
30	0.02667	1.18	−15.2795, 15.2795	−15.1794, −15.1794	−15.1794	−15.1790	−15.1794
				−14.4804, −14.3452		−14.4799	−14.4804
				−13.7671, 13.7671		−13.6830	−13.6844
50	0.016	0.741	−25.2954, 25.2954	−25.1866, −25.1866	−25.1865	−25.1870	−25.1866
				−24.5202, −24.3237		−24.5206	−24.5202
				−23.9210, 23.9210		−23.7847	−23.7848

higher-order RDMs, in principle. Mazziotti expressed the product writable term in these functionals with cumulants [6,7], and determined the rest term called the connected n-RDM $\Delta^{(n)}$ by requiring that the $(n+1)$-RDM approximated by his cumulant expansion reduces to the original n-RDM by contraction [6,7].

Mazziotti claimed that it is possible to get the exact higher-order RDMs from the exact 2-RDM if one reconstructs all the orders of the connected n-RDMs by his method [7]. Let us examine how this method works for a four-electron system. Neglecting the connected 4-RDM $\Delta^{(4)}$ we first approximate $\Delta^{(3)}$ by requiring that the approximated 4-RDM in terms of the 1-RDM and $\Delta^{(k)}$ ($k \leq 3$) contracts to the 3-RDM. In order to determine $\Delta^{(4)}$ we must use the contraction relation between 4- and 5-RDMs even for a *four-electron system*, but 5-RDM defined by Eq. (1.1) becomes identically zero. In principle, even in this case we can solve Eq. (12) of Ref. [7] to determine $\Delta^{(4)}$ by neglecting $\Delta^{(5)}$. This $\Delta^{(5)}$ should be determined from the higher-order contraction relation, showing that this is a nonterminating procedure. Note that $\Delta^{(k)}$ for $k>4$ is not zero to ensure k-RDM is zero. Rosina's theorem says nothing about the exactness of these approximated 4-RDMs, since generally they are not ensemble-representable RDMs. Approximated 4-RDMs are antisymmetric with respect to the permutation of the indices, but generally they are not non-negative operators. In other words, we must explicitly impose the representability conditions on the 4-RDM to get exact RDMs. The similarity of the formulas for the connected 3-RDM derived by the Green's-function method and his method indicates that his complete reconstruction would yield results of the same accuracy as the Parquet sum reported previously.

III. UNIQUENESS OF THE SOLUTION OF THE CONTRACTED SCHRÖDINGER EQUATION

In this section we focus on the distribution of the solutions of the 2-CSE as a function of energy under incomplete representability conditions. First we discuss properties of the 4-RDM of Lipkin's model in detail, and present the explicit formula of the 4-RDM. We then explain our basic analysis method to examine the existence of the solution. Finally we apply this analysis method to Lipkin's model to investigate the distribution of the solutions of CSE under the nonnegativity conditions of the 4-RDM and G matrix.

A. 4-RDM of Lipkin's model

The 4-RDM of Lipkin's model is discussed in detail and the explicit formula is presented, which will be used later. We examine the condensed 4-RDM defined as

$$D^{\sigma_1 \cdots \sigma_4}_{\sigma_1' \cdots \sigma_4'} = \frac{1}{4!} \sum_{P_i} \langle \Psi | a^\dagger_{p_1 \sigma_1} \cdots a^\dagger_{p_4 \sigma_4} a_{p_4 \sigma_4'} \cdots a_{p_1 \sigma_1'} | \Psi \rangle. \tag{3.1}$$

This 4-RDM has 256 elements in total. Using the Hermiticity and the permutational symmetry of the 4-RDM, $D^{klmn}_{abcd} = D^{abcd}_{klmn} = D^{lkmn}_{bacd} = \cdots$, the number of independent elements is reduced to 22. We express these elements as the expectation values of 22 spin operators shown in Table I. Using the conservation of parity, the number of particles, and the total angular momentum J^2, eight of them vanish, three are constants of motion, and the other three are just the multipliers of other variables. Thus the number of independent elements is at most eight, *irrespective of the number of the particles*

involved or the rank of the spin-orbital basis. Note that the rank of the spin-orbital basis primarily affects the accuracy and the difficulty of electronic structure calculations for real atoms and molecules.

Due to the conservation of the total angular momentum J^2, condensed RDMs of Lipkin's model have an additional symmetry,

$$D_{ab}^{kl} = D_{ba}^{kl}, \tag{3.2a}$$

$$D_{abc}^{klm} = D_{bac}^{klm}, \tag{3.2b}$$

$$D_{abcd}^{klmn} = D_{bacd}^{klmn}. \tag{3.2c}$$

Note that the exchange of indices does not accompany the negative sign. Due to this special symmetry, the following nine elements cover all the distinguishable 4-RDM elements: D_{++++}^{++++}, D_{++++}^{+++-}, D_{++--}^{++--}, D_{+---}^{+---}, D_{----}^{----}, D_{++++}^{+++-}, D_{++--}^{++--}, D_{+---}^{+---}, D_{----}^{++++}. Equations (3.2) are proved using Valdemoro's identities of RDMs [19], which express the condensed RDMs as

$$2! D_{ab}^{kl} = \langle J_a^k J_b^l - J_b^k \delta_a^l \rangle, \tag{3.3a}$$

$$3! D_{abc}^{klm} = \langle J_c^m (J_a^k J_b^l - J_b^k \delta_a^l) \rangle - 2(D_{ab}^{km} \delta_c^l + D_{ba}^{lm} \delta_c^k), \tag{3.3b}$$

$$4! D_{abcd}^{klmn} = \langle (J_c^m J_d^n - J_d^m \delta_c^n)(J_a^k J_b^l - J_b^k \delta_a^l) \rangle - 3! (D_{abd}^{mln} \delta_c^k + D_{abd}^{kmn} \delta_c^l + D_{abc}^{nlm} \delta_d^k + D_{abc}^{knm} \delta_d^l) - 2(D_{ab}^{mn} \delta_c^k \delta_d^l + D_{ab}^{nm} \delta_c^l \delta_d^k). \tag{3.3c}$$

The spin operator $J_{\sigma'}^\sigma$ is defined as

$$J_{\sigma'}^\sigma = \sum_p a_{p\sigma}^\dagger a_{p\sigma'} = \left(\frac{N}{2} + \sigma J_z \right) \delta_{\sigma'}^\sigma + \delta_{\sigma'+2}^\sigma J_+ + \delta_{\sigma'}^{\sigma+2} J_-. \tag{3.4}$$

Equation (3.3a) is precisely Eq. (B5) of Ref. [5]. To prove Eq. (3.2) we can assume that $a \neq b$ and $k \neq l$ without loss of generality. This assumption simplifies the equality to be proved for the 2-RDM as $D_{+-}^{+-} = D_{+-}^{-+}$, but one can easily show it by substituting $J_{\sigma'}^\sigma$ of Eq. (3.4) in Eq. (3.3a) and using the conservation of the angular momentum. Equalities for 3- and 4-RDMs are proved in the same way.

Next, consider the 2-CSE expressed by the condensed RDMs, which is

$$\frac{1}{2} \sum_{p_i} \langle \Psi | a_{p_1\sigma_1}^\dagger a_{p_2\sigma_2}^\dagger a_{p_2\sigma_2'} a_{p_1\sigma_1'} (H-E) | \Psi \rangle$$

$$= \{ v_{\sigma_1'}^{n_1} \delta_{\sigma_2'}^{n_2} + v_{\sigma_2'}^{n_2} \delta_{\sigma_1'}^{n_1} + w_{\sigma_1'\sigma_2'}^{n_1 n_2} \} D_{n_1 n_2}^{\sigma_1 \sigma_2} + 3 \{ v_{k_3}^{n_3} D_{\sigma_1'\sigma_2' n_3}^{\sigma_1 \sigma_2 k_3}$$

$$+ w_{\sigma_1' k_3}^{n_1 n_3} D_{n_1 \sigma_2' n_3}^{\sigma_1 \sigma_2 k_3} + w_{\sigma_2' k_3}^{n_2 n_3} D_{\sigma_1' n_2 n_3}^{\sigma_1 \sigma_2 k_3} \} + 6 w_{n_3 n_4}^{k_3 k_4} D_{\sigma_1' \sigma_2' k_3 k_4}^{\sigma_1 \sigma_2 n_3 n_4}$$

$$- E D_{\sigma_1'\sigma_2'}^{\sigma_1 \sigma_2} = 0. \tag{3.5}$$

Einstein's summation convention has been used here. The one- and two-particle potentials have the following two nonzero elements:

$$v_+^+ = -v_-^- = 1/2,$$

$$w_{--}^{++} = w_{++}^{--} = V.$$

Due to the symmetry of the model, the nontrivial relations are the following five:

$$\langle H \rangle = E, \tag{3.6a}$$

$$\langle J_z H \rangle = \langle J_z \rangle E, \tag{3.6b}$$

$$\langle J_z^2 H \rangle = \langle J_z^2 \rangle E, \tag{3.6c}$$

$$\langle H^2 \rangle = E^2, \tag{3.6d}$$

$$\langle (J_+^2 - J_-^2) H \rangle = 0. \tag{3.6e}$$

These equalities eliminate four free parameters in Table I, $\langle J_z H \rangle$, $\langle J_z^2 H \rangle$, $\langle H^2 \rangle$, and $\langle J_z^3 \rangle$. Thus the 4-RDM of Lipkin's model is expressed with the four parameters E, $z_1 = \langle J_z \rangle$, $z_2 = \langle J_z^2 \rangle$, and $z_3 = \langle J_z^4 \rangle$.

The actual formula of the 4-RDM is derived as follows. Using the commutation relation of the angular-momentum operators, we expressed the expectation values of the operators $J_{\sigma_1'}^{\sigma_1} J_{\sigma_2'}^{\sigma_2}$, $J_{\sigma_1'}^{\sigma_1} J_{\sigma_2'}^{\sigma_2} J_{\sigma_3'}^{\sigma_3}$, and $J_{\sigma_1'}^{\sigma_1} J_{\sigma_2'}^{\sigma_2} J_{\sigma_3'}^{\sigma_3} J_{\sigma_4'}^{\sigma_4}$ in terms of the independent parameters z_i and E. Valdemoro's identities of Eqs. (3.3) relate them to the condensed RDMs. After some complicated algebra, we obtain the distinguishable elements of the 4-RDM that satisfies the 2-CSE,

$$4! D_{++++}^{++++} = z_3 + z_2(6J^2 - 18J + 11) + z_1 A_2 + J(J-1) \times (J-2)(J-3) - F, \tag{3.7a}$$

$$4! D_{+++-}^{+++-} = -z_3 + z_2(3J-2) - z_1 A_1 + J^2(J-1)(J-2) + F/2, \tag{3.7b}$$

$$4! D_{++--}^{++--} = z_3 - z_2(2J^2 - 2J + 1) + J^2(J-1)^2, \tag{3.7c}$$

$$4! D_{+---}^{+---} = -z_3 + z_2(3J-2) + z_1 A_1 + J^2(J-1)(J-2) - F/2, \tag{3.7d}$$

$$4! D_{----}^{----} = z_3 + z_2(6J^2 - 18J + 11) - z_1 A_2 + J(J-1) \times (J-2)(J-3) + F, \tag{3.7e}$$

$$4! V D_{++++}^{++--} = -z_2(2J-3-E) - z_1 B_- + E C_-, \tag{3.7f}$$

$$4! V D_{+++-}^{+---} = -z_2 E + z_1(6J - 5 + V^{-2})/2 + E C_+, \tag{3.7g}$$

$$4! V D_{++--}^{----} = z_2(2J-3+E) - z_1 B_+ + E C_-, \tag{3.7h}$$

$$4!D_{++++}^{----} = -z_3 + 2z_2(J^2 + J - 5/2 + V^{-2}) - 4z_1 E V^{-2}$$
$$+ 2E^2 V^{-2} - (J+2)(J+1)J(J-1). \quad (3.7i)$$

Parameters A_k, $B_\pm$, $C_\pm$, and F are defined as

$$A_k = k(2J-3)\{(2J-1)^k + V^{-2}\}/2,$$

$$B_\pm = \{(2J-1)^2 + V^{-2}\}/2 \pm (2J-3)E,$$

$$C_\pm = \{(2J-3)(J-1) \pm (J+1-V^{-2})\}/2,$$

$$F = (2J-3)EV^{-2}.$$

This 4-RDM is normalized and gives an energy expectation value of E. The lower-order RDMs are obtained by contraction. Clearly parameters z_i should satisfy the following inequalities:

$$-J \leq z_1 \leq +J, \quad (3.8a)$$

$$0 \leq z_2 \leq J^2, \quad (3.8b)$$

$$0 \leq z_3 \leq J^4. \quad (3.8c)$$

Similar to the ordinary 4-RDM, a representable condensed 4-RDM should be non-negative, which is shown as follows. Due to the conservation of the number of particles in each spatial orbital, Eq. (2.3c), the n-RDM is nonzero only when the lower indices $(p'_1 \cdots p'_4)$ are a permutation of the upper ones $(p_1 \cdots p_4)$. Using the equivalence of the sites p, the 4-RDM is factorized into p and σ parts,

$$N(N-1)(N-2)(N-3)\Gamma^{(4)}{}^{p_1\sigma_1 \cdots p_4\sigma_4}_{p'_1\sigma'_1 \cdots p'_4\sigma'_4}$$

$$= D^{\sigma_1 \cdots \sigma_4}_{\sigma'_1 \cdots \sigma'_4} \mathrm{Det} \begin{pmatrix} \delta^{p_1}_{p'_1} & \cdots & \delta^{p_4}_{p'_1} \\ \vdots & & \vdots \\ \delta^{p_1}_{p'_4} & \cdots & \delta^{p_4}_{p'_4} \end{pmatrix}. \quad (3.9)$$

Thus the eigenvalue problem of the 4-RDM is split into those of p and σ parts, and the non-negativity of the 4-RDM is equivalent to that of the condensed 4-RDM D. Similar relations hold for other orders of RDMs.

The condensed 4-RDM could be expressed as a 16×16 matrix. Using conservation of parity, the eigenvalue problems of this matrix are split into those of 8×8 matrices. The additional symmetry of Lipkin's model of Eq. (3.2) further simplifies them. All the nonzero eigenvalues of the 4-RDM are the same as those of the following 2×2 and 3×3 matrices, and the non-negativity of the 4-RDM is equivalent to their non-negativities:

$$\begin{pmatrix} D^{+++-}_{+++-} & D^{+---}_{+++-} \\ D^{+++-}_{+---} & D^{+---}_{+---} \end{pmatrix}, \quad (3.10a)$$

$$\begin{pmatrix} D^{++++}_{++++} & \sqrt{6}D^{++--}_{++++} & D^{----}_{++++} \\ \sqrt{6}D^{++++}_{++--} & 6D^{++--}_{++--} & \sqrt{6}D^{----}_{++--} \\ D^{++++}_{----} & \sqrt{6}D^{++--}_{----} & D^{----}_{----} \end{pmatrix}. \quad (3.10b)$$

The G condition we employed is the non-negativity of the expectation value $\langle G^\dagger G \rangle$ for an arbitrary scalar $x_{k_1 \cdots k_4}$, where G is defined as

$$G = \frac{1}{2} \sum_{p_1 < p_2} \sum_{p_3 < p_4} \sum_{\sigma_i} x_{p_1\sigma_1 \cdots p_4\sigma_4}(a^\dagger_{p_1\sigma_1}a^\dagger_{p_2\sigma_2}$$
$$+ a^\dagger_{p_1\sigma_2}a^\dagger_{p_2\sigma_1})a_{p_3\sigma_3}a_{p_4\sigma_4}. \quad (3.11)$$

Thus the G condition is the non-negativity of the matrix,

$$G^{p_1\sigma_1 \cdots p_4\sigma_4}_{p'_1\sigma'_1 \cdots p'_4\sigma'_4} = \frac{1}{4}\langle\Psi|a^\dagger_{p'_4\sigma'_4}a^\dagger_{p'_3\sigma'_3}(a_{p'_2\sigma'_2}a_{p'_1\sigma'_1} + a_{p'_2\sigma'_1}a_{p'_1\sigma'_2})$$
$$\times (a^\dagger_{p_1\sigma_1}a^\dagger_{p_2\sigma_2} + a^\dagger_{p_1\sigma_2}a^\dagger_{p_2\sigma_1})a_{p_3\sigma_3}a_{p_4\sigma_4}|\Psi\rangle. \quad (3.12)$$

$G^{p_1\sigma_1 \cdots p_4\sigma_4}_{p'_1\sigma'_1 \cdots p'_4\sigma'_4}$ is nonzero only when $p_1 \neq p_2$, $p_3 \neq p_4$, $p'_1 \neq p'_2$, and $p'_3 \neq p'_4$, because the RDMs are factorizable as Eq. (3.9). Thus the sum in Eq. (3.11) can be regarded as all the distinct pairs of spin orbitals.

This G matrix is a block-diagonal matrix due to the symmetry of Eq. (2.3), which is easily verified. Suppose, for example, all the four indices p_i are different. Then the matrix element is zero unless $p'_k = p_k$. Using the equivalence of the site p_k, the eigenvalue problems of this G matrix become those of three 16×16 matrices,

$$G_1 = C_4 - C_3 + C_2, \quad (3.13a)$$

$$G_2 = -(2J-3)C_4 + (2J-4)C_3 + C_2, \quad (3.13b)$$

$$G_3 = (2J-3)(J-1)C_4 + (2J-2)C_3 + C_2. \quad (3.13c)$$

C_i are defined as

$$C_2 = \frac{1}{2}\binom{2J}{4}^{-1} D^{\sigma'_3\sigma'_4}_{\sigma_3\sigma_4}(\delta^{\sigma_1}_{\sigma'_1}\delta^{\sigma_2}_{\sigma'_2} + \delta^{\sigma_2}_{\sigma'_1}\delta^{\sigma_1}_{\sigma'_2}), \quad (3.14a)$$

$$C_3 = \frac{1}{2}\binom{2J}{3}^{-1}(D^{\sigma_1\sigma'_3\sigma'_4}_{\sigma'_1\sigma_3\sigma_4}\delta^{\sigma_2}_{\sigma'_2} + D^{\sigma_1\sigma'_3\sigma'_4}_{\sigma'_2\sigma_3\sigma_4}\delta^{\sigma_2}_{\sigma'_1} + D^{\sigma_2\sigma'_3\sigma'_4}_{\sigma'_1\sigma_3\sigma_4}\delta^{\sigma_1}_{\sigma'_2}$$
$$+ D^{\sigma_2\sigma'_3\sigma'_4}_{\sigma'_2\sigma_3\sigma_4}\delta^{\sigma_1}_{\sigma'_1}), \quad (3.14b)$$

$$C_4 = \binom{2J}{4}^{-1} D^{\sigma_1\sigma_2\sigma'_3\sigma'_4}_{\sigma'_1\sigma'_2\sigma_3\sigma_4}, \quad (3.14c)$$

where $\binom{n}{k}$ is the binomial coefficient. The eigenvalues of G_1 are completely the same as those of the 4-RDM. That is, the G condition contains the P condition in Lipkin's model. The symmetry of the 4-RDM, Eq. (3.2), could be used to further
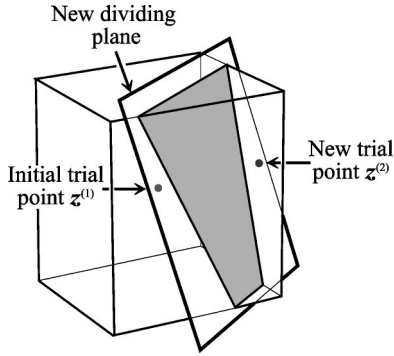
FIG. 1. Iterative reduction of the convex set. Initial trial convex set is the rectangle of Eq. (3.8), and the center-of-mass coordinate $z^{(1)}$ gives the trial 4-RDM through Eq. (3.7). The eigenvector of the negative eigenvalue of the P or G matrix gives an additional constraint of Eq. (3.16), which divides the trial convex set by a plane. The rightmost triangular prism contains the representable convex set, which is used as a new trial one.

simplify the G_i. We do not have to impose the non-negativity of hole-RDM separately because of the particle-hole equivalence in Lipkin's model.

The advantage of Lipkin's model is that the inherent symmetry dramatically simplifies RDMs, and the detailed analysis could be carried out easily. However there may also be shortcomings. The 4-RDM of this model is much simpler than those of atoms and molecules, which are our main concerns. It has only eight free parameters, while the simplest model of a molecule, H_2 with a minimal basis set, has much complicated RDMs. This model is useful to verify a theory if we keep in mind such shortcomings.

B. Method to examine the existence of solutions

We want to know whether a normalized 4-RDM exists that satisfies the 2-CSE [Eq. (1.2) with $n=2$], P-, and G-representable conditions for a given energy E. Since the 2-CSE in terms of the 4-RDM is an underdetermined system of linear equations, the general solution D is expressed as a linear combination of the nontrivial special solutions D_i,

$$D = \sum z_i D_i. \tag{3.15}$$

Hence for a given energy E we examine the existence of parameters z_i that yield a representable 4-RDM. Specific forms of D_i for the 4-RDM of Lipkin's model are given by Eq. (3.7). Since the ensemble-representable 4-RDMs form a convex set, the associated vectors $z=(z_1,z_2,z_3)$ also form another convex set. Our analysis method is similar to that in Ref. [18], which is as follows.

(1) We first take an initial trial convex set, and choose an arbitrary point $z^{(1)}=(z_1^{(1)},z_2^{(1)},z_3^{(1)})$ in it. The center-of-mass coordinate of the vertices of the convex polytope was used as this trial point (see Fig. 1). These vertices were calculated by the same procedure as the phase I problem of linear programming [22]. Such vertices cover all the extreme elements of the trial convex set.

(2) Then we calculate the 4-RDM of Eq. (3.7) or (3.15) for this trial point $z^{(i)}$, and the eigenvalues of the 4-RDM and G matrix, or equivalently those of Eqs. (3.10) and (3.13). All the eigenvalues should be non-negative, but some eigenvalues may become negative. Denoting one of the corresponding eigenvectors as v_k, the following inequality is necessary for the 4-RDM to satisfy the P condition:

$$\sum z_i \langle v_k | D_i | v_k \rangle \geq 0. \tag{3.16}$$

This inequality divides the trial convex set by a plane, as shown in Fig. 1. One of the divided convex sets without the trial point $z^{(1)}$ contains N-representable 4-RDMs, and we take it as a new trial convex set. The G condition also gives the same kind of inequalities.

(3) By repeating this procedure we obtain several necessary inequalities for the 4-RDM to be representable. Finally we arrive at one of the following two results. (i) The 4-RDM associated with the point $z^{(k)}$ does not have any negative eigenvalues. This indicates that there exists the normalized 4-RDM that satisfies the 2-CSE, P, and G conditions with energy E. (ii) No region of the convex set remains that satisfies the necessary inequalities for the 4-RDM to be representable. That is, no physically acceptable solution exists. In this way we can decide whether the solution of the 2-CSE with a given energy exists under some approximated representability conditions.

C. Numerical result

The method described so far was applied to the excited states of Lipkin's model to investigate the distribution of the solutions of CSE under the incomplete representability conditions. The results are summarized in Tables II and III. Table II compared the energies with the exact ones and those of coupled cluster methods, including variational single and double substituted coupled cluster method and the coupled cluster method up to the quadruple excitations (CCSDTQ) [21]. The percentage of the ground-state correlation energy with respect to the total energy is also shown for convenience. Note that these ratios are about 1.45% for the He atom and 3.5% for the H_2 molecule, respectively. Table III shows the calculated 4-RDMs for some selected energies. These energies correspond to the upper or lower end of the regions where solutions exist.

Our finding is that the distribution of the solutions changes dramatically with the interaction parameter V and the representability conditions imposed. As shown in Table II, the exact energies of the first and second excited states can be estimated accurately from the results of the G condition if the interaction is smaller than $1/N$. Spurious solutions are localized near the true one. On the contrary, the P condition does not give any valuable results for the second excited state, or even for the ground state, under the moderate interaction $V=0.071\approx 1/14$. Note that in this model exact energies appear as pairs of $\pm E$. The G condition is stronger than the P condition in this model because the former contains the latter. However the G condition is not yet sufficient

TABLE III. Condensed 4-RDMs of the Lipkin's quasispin model calculated by the second order contracted Schrödinger equation and the P- and G-representable conditions. Energies in this table correspond to the upper or the lower end of the region, where solutions exist. The convex polytope of solutions shrinks to a single point at these points. Numbers in the parentheses indicate the errors to the exact values, while those in the square brackets indicate powers of 10.

State	Potential V	Energy E		$\langle J_z \rangle$		$\langle J_z^2 \rangle$		$\langle J_z^4 \rangle$	
					G condition				
14_1	5	-17.2778	$(-9.4022[-3])$	-1.6122	$(-3.3832[-2])$	7.7839	(0.2987)	1.5593[2]	(1.5285[1])
14_2		-16.9855	(0.2782)	-1.3877	(0.1747)	5.4450	(-1.9245)	5.3998[1]	$(-7.9444[1])$
14_2	1	-6.6219	$(-8.5078[-5])$	-5.3420	$(-3.2683[-4])$	2.9857[1]	$(4.5571[-3])$	9.9518[2]	(0.3514)
14_2		-6.5073	(0.1146)	-5.1660	(0.1756)	2.7948[1]	(-1.9095)	8.7652[2]	$(-1.1831[2])$
					P condition				
14_1	0.8	-7.1995	$(-3.6418[-2])$	-6.8655	$(-6.1063[-2])$	4.7477[1]	(0.7522)	2.2999[3]	(6.2027[1])
14_1		-7.0596	(0.1035)	-6.6285	(0.1760)	4.4547[1]	(-2.1782)	2.0575[3]	$(-1.8045[2])$

for higher excited states: a spurious solution appears at every energy above the second excited state. It is impossible to obtain a discrete energy region that approximates the energy of the true excited state.

The energy width containing the spurious solutions increases as the interaction parameter increases. As a result, the estimated energy and 4-RDM become worse as the interaction increases. This is especially true for the excited states. The calculated ground-state energy becomes much lower than the exact one and the error will not be negligible if we impose only the P condition. The G condition works well for the ground state. It gives energies as accurate as or even more accurate than the CCSDTQ method. The calculated 4-RDMs at the lowest end of the region were in good agreement with the exact values. They were always better than those of the higher end, and the additional variational minimization of energy to eliminate the nonuniqueness would work well for the ground state.

Let us compare the present result with the previous one. Tables III and IV of Ref. [5] summarize the results of the 2-CSE by only imposing the P condition under weak interaction. These results of the ground and the excited states were rather accurate, and even the results of the second excited states of 10_3 and 15_3 were reported. This is in contrast to our results of the P condition. The accurate results of the excited states obtained previously by imposing the P condition is due to the weaker interaction. The interaction parameters used in Ref. [5] are much smaller than in the present study. The ground-state correlation energies in these tables are about 0.1–0.6 % of the total energies. As shown in our Table II, if we increase the interaction to $V = 0.046\,15$, for example, the P condition does not give any discrete energy region for the second excited state. Our present study also indicates the nonuniqueness of the previous results, because every 4-RDM in the allowed convex polytope is equally acceptable as a solution.

Next, let us examine the results of strong interaction. These results are interesting because the ground state almost degenerates with the first excited state. Previous results are summarized in Table III of Ref. [8]. This table compares the ground-state correlation energies of the spin Hamiltonian $H = (2/N)J_z + [V/(N-1)](J_+^2 + J_-^2)$ and unit conversion is necessary for comparison.

The results of the single and double substituted coupled cluster (CCSD) method in that table need some considerations. It is well known that the stable Hartree-Fock (HF) solution of this model is just the unperturbed ground state for weak interaction ($NV < 1$), which is suitable for the zeroth-order state to treat electron correlations. On the other hand, for strong interaction ($NV > 1$), the unperturbed ground state gives an unstable HF solution, and two degenerate, symmetry-breaking solutions give lower energy [20]. These stable solutions must be used as the reference functions of CC or CI, because their results depend on the occupied (or empty) orbital space.

As shown in Table VI of Ref. [21], CCSD based on this symmetry-breaking solution gives 84.0%, 98.6%, and 99.8% of correlation energies for $N = 14$, 30, and 50 systems, respectively. Interaction strength of these calculations corresponds to $V = 1.6/14$, $1.6/30$, and $1.6/50$, respectively. On the other hand, the results of CCSD, single and double substituted configuration interaction, or that up to quadruple excitations in Table III of Ref. [8] are far from exact for $V = 1.6$. This result is due to the improper choice of the reference Hartree-Fock solution. It is interesting that the variational CCSD works well even for the strong interaction, as shown in Table II. CCSD is not so bad for the ground state as is discussed in Ref. [8] if the proper reference function is used.

The ability of the coupled cluster method depends heavily on the target state. As shown in the first three columns of Table II, which correspond to the strongest interaction, CCSDTQ failed to reproduce the second excited state. Calculated energy is rather close to the exact energy of the third excited state. This implies that the multireference CC or CI method should be used for strong interaction, as in the case of bond-breaking processes.

One may expect that the CSE approach works better for these systems, because it does not rely on the choice of the reference state and could be applied to the degenerate states with equal footings. As shown in Table II, the G condition gives the energy range that contains both states, but fails to give the discrete regions. It shows the potential difficulty of the representability method for quasidegenerate systems. Although it may yield exact 4-RDMs of both states, they are

embedded in a sea of the spurious solutions. This makes it difficult to obtain useful information about these states.

IV. CONCLUSIONS

In this paper we consider two fundamental questions in the contracted Schrödinger equation approach: 1-1 mapping between the 2-RDM of an excited state and the wave function, and the uniqueness of the solution of the ensemble-representability method. The goal in our density-matrix theory is to solve the 2-CSE, which is still equivalent to the original Schrödinger equation in the N-representable density-matrix space.

There are two different approaches to solve this equation. The first one, called the functional approach, takes the 2-RDM as a basic variable and reconstructs 3- and 4-RDMs from the 2-RDM to eliminate the indeterminacy. The fundamental question of this approach is the existence of the reconstruction functional. This question is examined for excited states. In contrast to the previous result [5], using Lipkin's quasispin model we found some counterexamples of the wave functions, which give the same 2-RDMs as the excited state. Thus the 2-RDM of an excited state does not determine the wave function uniquely, and the functional method that uses only 2-RDM as a basic variable cannot become an exact theory for excited states.

The second method to solve the 2-CSE is to use the 4-RDM as a basic variable and to impose some known N-representable constraints. The solution of this equation is not unique and it yields all the exact solutions together with the spurious ones. Using Lipkin's quasispin model we examined this nonuniqueness of the solutions as a function of energy under the P- and G-representability conditions of the 4-RDM. We found that the solutions in the low-energy region are well separated from each other, but under moderate interaction or in the higher-energy region, there exist spurious solutions for almost all energies. Thus, although the G condition of the 4-RDM is accurate for the ground state of Lipkin's model, it is not sufficient for the excited states.

We also presented the detailed analysis of the RDMs of Lipkin's model, and found that they have extremely simple structures compared to those of the usual many-body problems because of the special symmetry. The number of independent elements in the 2-RDM and 4-RDM are at most

three and ten, respectively, *irrespective of the number of particles involved, or the rank of the spin-orbital basis*. Their P, Q, and G representable conditions also have simple forms. Lipkin's model is a fine model for giving examples and counterexamples to theorems, as demonstrated in this paper, but probably has a limited usefulness for establishing the validity of any new computational methodology.

APPENDIX

In this appendix we give explicit formulas for the condensed 2- and 3-RDMs of Lipkin's model. Using the permutational symmetry, Hermiticity, and the symmetry of Eq. (3.2), it can be shown that the condensed 2-RDM of Eq. (2.4) has only four nonzero independent elements, D_{++}^{++}, D_{--}^{--}, D_{+-}^{+-}, and D_{--}^{++}. We express them with the expectation values of the operators in the first three rows of Table I as

$$2D_{++}^{++} = J(J-1) + (2J-1)\langle J_z \rangle + \langle J_z^2 \rangle,$$

$$2D_{--}^{--} = J(J-1) - (2J-1)\langle J_z \rangle + \langle J_z^2 \rangle,$$

$$2D_{+-}^{+-} = J^2 - \langle J_z^2 \rangle,$$

$$2D_{--}^{++} = (\langle H \rangle - \langle J_z \rangle)/V.$$

These formulas were derived using the same procedure described in Sec. III A. Similarly the nonzero independent elements of the condensed 3-RDM are

$$6D_{+++}^{+++} = J(J-1)(J-2) + (3J^2 - 6J + 2)\langle J_z \rangle + 3(J-1) \times \langle J_z^2 \rangle + \langle J_z^3 \rangle,$$

$$6D_{---}^{---} = J(J-1)(J-2) - (3J^2 - 6J + 2)\langle J_z \rangle + 3(J-1) \times \langle J_z^2 \rangle - \langle J_z^3 \rangle,$$

$$6D_{++-}^{++-} = J^2(J-1) + J^2\langle J_z \rangle - (J-1)\langle J_z^2 \rangle - \langle J_z^3 \rangle,$$

$$6D_{+--}^{+--} = J^2(J-1) - J^2\langle J_z \rangle - (J-1)\langle J_z^2 \rangle + \langle J_z^3 \rangle,$$

$$6D_{+++}^{+-} = \{(J-1)(\langle H \rangle - \langle J_z \rangle) - \langle J_z^2 \rangle + \langle J_z H \rangle\}/V,$$

$$6D_{---}^{-++} = \{(J-1)(\langle H \rangle - \langle J_z \rangle) + \langle J_z^2 \rangle - \langle J_z H \rangle\}/V.$$

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