Computation of quantum phase transitions by reduced-density-matrix mechanics

Gergely Gidofalvi and David A. Mazziotti*
Department of Chemistry and the James Franck Institute, The University of Chicago, Chicago, Illinois 60637, USA
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Quantum phase transitions are explored with reduced-density-matrix (RDM) mechanics. While in wave mechanics the quantum phase transition is identified by a crossing or avoided crossing between ground- and excited-state energies, in RDM mechanics the transition is characterized by movement of the ground-state two-electron RDM (2-RDM) along the boundary of the convex set of 2-RDMs between regions with dramatically different expectation values (order parameters) of one or more operators. With recent advances the ground-state 2-RDM can be directly computed without the many-particle wave function by variational optimization of the energy with the 2-RDM [D. A. Mazziotti, Phys. Rev. Lett. **93**, 213001 (2004)]. Because the variational calculation of the 2-RDM does not depend on a reference wave function, it can accurately predict the energies and properties of a system both near and far from the quantum phase transition.

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I. INTRODUCTION

Quantum mechanical systems can undergo significant changes in properties with a small change in a system parameter such as the doping ratio of a superconductor. These dramatic changes, known as *quantum phase transitions*, are signatures of the quantum world, and the value of the parameter at which the change occurs is called the *critical point* [1]. In wave mechanics the transformation at the critical point is associated with either an actual or an avoided level crossing of the energies from ground- and excited-state wave functions where one or more properties, described as *order parameters*, exhibit an abrupt change. In this paper a different approach to computing and identifying quantum phase transitions using two-particle reduced density matrices (2-RDMs) [2] is developed and applied to investigating the phase transition in the Lipkin quasispin model [3]. The *two-particle density matrix* (2-RDM) is defined by integrating the N-particle density matrix over coordinates 3 to N,

$$^2D(1,2;1',2') = \int {}^ND(1,2,\ldots,N;1',2',\ldots,N)d3\cdots dN, \tag{1}$$

where, when the quantum system is in a pure state, the N-particle density matrix is expressible in terms of the wave function $\psi(1,2,\ldots,N)$

$$^ND(1,2,\ldots,N;1',2',\ldots,N') = \psi(1,2,\ldots,N)\psi^*(1',2',\ldots,N'). \tag{2}$$

Because Rosina's theorem [4,5] demonstrates that for Hamiltonians with pairwise interactions there is a one-to-one mapping between the ground-state 2-RDM and its many-particle wave function, the 2-RDM contains sufficient information to describe ground-state energies and properties including the location of critical points and the behavior of order parameters within quantum phase transitions [6].

The goal of the paper is twofold: (i) to present a general 2-RDM perspective on the process of quantum phase transitions with the Lipkin model as a specific example and (ii) to demonstrate the accuracy of the variational 2-RDM method [7–12] *without* construction of the wave function for modeling the energies and properties of a quantum system in the vicinity of the quantum phase transition. In RDM mechanics the transition is characterized by movement of the ground-state 2-RDM along the boundary of the convex set of 2-RDMs between regions with dramatically different expectation values (order parameters) of one or more operators. The change in the location of the 2-RDM on the boundary of its convex set collectively describes the changes in *all* of the order parameters. With recent advances the ground-state 2-RDM can be directly computed without the many-particle wave function by variational optimization of the energy with respect to the elements of the 2-RDM [7–12]. Because the variational calculation of the 2-RDM does not depend on a reference wavefunction, it can accurately predict the energies and properties of a system both near and far from the quantum phase transition. The formulation of RDM mechanics for quantum phase transitions has relevance to optimization theory, particularly semidefinite programming [13], as well as quantum entanglement [14] and electronic structure [7–12].

II. THEORY

The Lipkin quasispin model consists of N fermions distributed over two N-fold degenerate levels separated by an energy ϵ where the particles have a pairwise "monopole-monopole" interaction of strength V that scatters pairs of particles between the two levels [3,5,7,15–20]. Because N fermions can occupy each level, the Lipkin model may also be interpreted as a two-level bosonic model [15,16] with the Hamiltonian

$$\hat{H} = \frac{\epsilon}{N}\sum_{m=\pm1} m\hat{a}_m^\dagger\hat{a}_m + \frac{V}{N(N-1)}\sum_{m=\pm1}\hat{a}_{+m}^\dagger\hat{a}_{+m}^\dagger\hat{a}_{-m}\hat{a}_{-m}, \tag{3}$$

where $\hat{a}^\dagger(\hat{a})$ are bosonic creation (annihilation) operators, $m(\pm1)$ denotes the two levels, and ϵ and V are parameters. The expectation value of Eq. (3) yields the energy

*Electronic address: damazz@uchicago.edu

$$E = \frac{\epsilon}{N} \sum_{m=\pm 1} m {}^1D^m_m + \frac{V}{N(N-1)} \sum_{m=\pm 1} {}^2D^{+m,+m}_{-m,-m}, \tag{4}$$

where the elements of the 1- and 2-RDMs are defined in second quantization as

$${}^1D^i_k = \langle \psi | \hat{a}^\dagger_i \hat{a}_k | \psi \rangle, \tag{5}$$

$${}^2D^{i,j}_{k,l} = \langle \psi | \hat{a}^\dagger_i \hat{a}^\dagger_j \hat{a}_l \hat{a}_k | \psi \rangle. \tag{6}$$

The 1-RDM can be obtained from integration (or contraction) of the 2-RDM. With the generators of the SU(2) group the Hamiltonian can be expressed in terms of the operators $\hat{J}_z$, $\hat{J}_+$, and $\hat{J}_-$ which obey the usual angular-momentum commutation relations [3],

$$\hat{H} = \frac{2\epsilon}{N} \hat{J}_z + \frac{V}{N(N-1)} (\hat{J}^2_+ + \hat{J}^2_-), \tag{7}$$

and the elements of the 2-RDM can be fully parametrized by the expectation values of these three operators. Because of symmetry the N-particle wave function has at most $(N+1)$ degrees of freedom, and exact solutions of the Schrödinger equation are available from diagonalization of the Hamiltonian in the basis $|J, J_z\rangle$ with $J=N/2$ and J_z ranging from $-J$ to J. Two properties of the Lipkin model will be especially important in this study. First, sets of approximately N-representable 2-RDMs can be visually compared with the set of N-representable 2-RDMs because the model's 2-RDM is fully described by three expectation values [17], namely $\langle \hat{J}_z \rangle$, $\langle \hat{J}^2_z \rangle$, and $\langle \hat{J}^2_+ + \hat{J}^2_- \rangle$. Second, the model exhibits a quantum phase transition[18–20] at $V_c = |\epsilon|$ from an ordered phase with most particles in the lower level to a deformed phase.

Equation (4) suggests that the ground-state energy may be minimized with respect to the 2-RDM matrix elements *without* the many-particle wave function. (In general, for any quantum system of indistinguishable particles with pairwise interactions the energy may be expressed as a linear functional of the 2-RDM.) However, direct minimization of Eq. (4) yields an energy which is significantly below the correct ground-state energy. A *reduced* density matrix requires nontrivial constraints, known as *N-representability conditions* [21,22], to guarantee that it derives from the integration of an N-particle density matrix (or wave function). The N-representability problem of the 2-RDM stymied the variational calculation of the energy with respect to elements of the 2-RDM for 50 years [2,12]. Recent calculations [7–10], however, show that N-representability of the 2-RDM can be controlled by systematic necessary constraints on the p-RDM, known as p-positivity conditions [7].

The p-positivity conditions can be defined by restricting the metric (or overlap) matrix M with elements

$$M^i_j = \langle \psi | \hat{C}_i \hat{C}^\dagger_j | \psi \rangle \tag{8}$$

to be positive semidefinite, where the operators $\hat{C}_i$ are products of p second-quantized operators. The metric matrix M is block diagonal with $p+1$ distinct blocks corresponding to the number of creation operators in $\hat{C}_i$. For $p=2$ we obtain the three metric matrices [7,22]

$${}^2D^{i,j}_{k,l} = \langle \psi | \hat{a}^\dagger_i \hat{a}^\dagger_j \hat{a}_l \hat{a}_k | \psi \rangle,$$

$${}^2G^{i,j}_{k,l} = \langle \psi | \hat{a}^\dagger_i \hat{a}_j \hat{a}^\dagger_l \hat{a}_k | \psi \rangle,$$

$${}^2Q^{i,j}_{k,l} = \langle \psi | \hat{a}_i \hat{a}_j \hat{a}^\dagger_l \hat{a}^\dagger_k | \psi \rangle, \tag{9}$$

which are three representations of the 2-RDM corresponding to the probability distributions for two particles, one particle and one hole, and two holes, respectively, where a hole is the absence of a particle. Although each matrix can be computed from the other matrices by linear mappings from rearranging the second-quantized operators by commutation relations, constraining each matrix to be positive semidefinite is necessary to prevent each probability distribution from becoming negative.

The ground-state energy as a functional of a 2-RDM restricted by 2-positivity conditions can be minimized by a special type of optimization known as *semidefinite programming* [7–10,16], which under mild assumptions yields a global minimum [13]. Because 2-positivity conditions are only a subset of the complete N-representability conditions, the set of 2-positive 2-RDMs is larger than the set of N-representable 2-RDMs. Consequently, unlike a Rayleigh-Ritz variational calculation with a wave function, a variational 2-RDM computation with 2-positivity constraints yields a rigorous *lower* bound to the exact energy. The results of 2-positivity may be systematically improved by imposing p-positivity conditions for $p>2$ [7,16]. A 2-RDM is said to be p-positive when it derives from the integration of a p-RDM that satisfies the p-positivity conditions. The 2-positivity constraints have been shown to give accurate ground-state electronic energies and properties of molecules at both equilibrium and nonequilibrium geometries [8–10].

In the Lipkin model the information in the 2-RDM is completely equivalent to the expectation values of the three operators $\langle \hat{J}_z \rangle$, $\langle \hat{J}^2_z \rangle$, and $\langle \hat{J}^2_+ + \hat{J}^2_- \rangle$ [5,17]. The convex set of N-representable 2-RDMs is three dimensional with axes labeled by the expectation values of these operators. Furthermore, the extreme points of the set may be generated by calculating the ground-state 2-RDMs for all Hamiltonians expressible by linear combinations of the three operators. The Lipkin Hamiltonian in Eq. (7), however, does not depend on $\hat{J}^2_z$, and hence, it generates a subset of the extreme N-representable 2-RDMs. While this convex subset is still three dimensional, we can visualize the boundary of the set in two dimensions because each ground-state 2-RDM from a Lipkin Hamiltonian is extreme in the pair of expectation values $\langle \hat{J}_z \rangle$ and $\langle \hat{J}^2_+ + \hat{J}^2_- \rangle$. For the remainder of the paper we will use the phrase "the convex set of N-representable 2-RDMs" to refer to this subset of 2-RDMs characterized by the two expectation values $\langle \hat{J}_z \rangle$ and $\langle \hat{J}^2_+ + \hat{J}^2_- \rangle$. The above discussion applies equally well to "the convex set of p-positive 2-RDMs" whose extreme points are generated by minimizing each Lipkin Hamiltonian with respect to a p-positive 2-RDM.

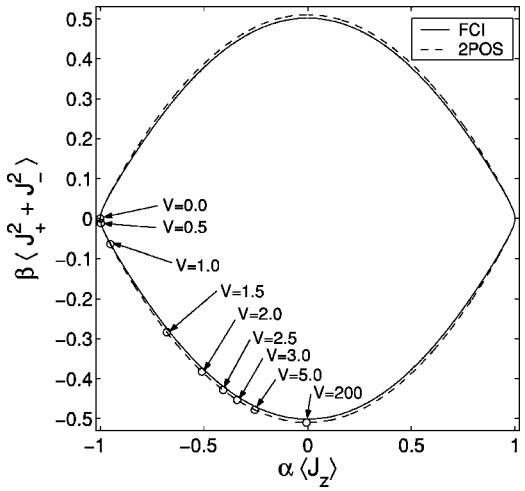

FIG. 1. The convex set of 2-positive 2-RDMs (2POS) is compared to the convex set of *N*-representable 2-RDMs (FCI) for the Lipkin model. The 2-positive set contains the *N*-representable set. The circles show the movement of the 2-RDM along the boundary of the set of 2-positive 2-RDMs as a function of the interaction *V*. The significant increase in the "speed" of the 2-RDM around the critical point ($V \approx 1$) is a signature of the quantum phase transition.

III. CALCULATIONS

For a Lipkin model with 50 fermions the convex set of *N*-representable 2-RDMs is compared in Fig. 1 with the convex set of 2-positive 2-RDMs. The boundaries of the 2-positive and *N*-representable sets are computed by evaluating the expectation values $\langle \hat{J}_z \rangle$ and $\langle \hat{J}_+^2+\hat{J}_-^2 \rangle$ with respect to the 2-positive and *N*-representable ground-state 2-RDMs of the Hamiltonians in Eq. (3) with $\epsilon=1$ and $V \in [-200,200]$. Pairs of expectation values $\langle \hat{J}_z \rangle$ and $\langle \hat{J}_+^2+\hat{J}_-^2 \rangle$ can be obtained from an *N*-representable 2-RDM if and only if they lie inside the solid curve. Similarly, pairs of expectation values can be obtained from a 2-positive 2-RDM if and only if they lie inside the dashed curve. Because the 2-positivity conditions are necessary but not sufficient *N*-representability conditions, the convex set of 2-positive 2-RDMs contains the set of *N*-representable 2-RDMs. At the two points where the expectation value of the two-body interaction $\langle \hat{J}_+^2+\hat{J}_-^2 \rangle$ vanishes, all of the fermions are in one level ($|\alpha\langle \hat{J}_z \rangle| = 1$), and the 2-positive and *N*-representable sets coincide. The scaling factors α and β are $2/N$ and $2/[N(N-1)]$, respectively. The difference between the two sets peaks at $\alpha\langle \hat{J}_z \rangle \approx 0.6$. Plotting the convex set of 3-positive 2-RDMs in Fig. 1 would yield a curve indistinguishable from the *N*-representable curve.

Because the positivity conditions are neither perturbative nor reference dependent, they have been observed to yield accurate energies and properties for multireferenced systems [7–10,16]. For $N=50$ in the vicinity of the quantum phase transition Table I compares the accuracy of ground-state energies from variational RDM theory with *p*-positivity conditions (*p*POS) with those from coupled-cluster theory [20] with *p*-particle excitations (CC*p*). Both 2-POS and CC2 are similar in accuracy, but the 2-RDM method exhibits much faster convergence with *p*. The 4POS (6POS) energies are 2 to 3 (4 to 6) orders of magnitude more accurate than the CC4 (CC6) energies. Furthermore, the coupled-cluster equations do not have a solution for $V>1$ without a change in the reference wave function [20], but the variational 2-RDM method, which selects the scaled identity matrix as an initial guess for the 2-RDM, does not depend upon a reference 2-RDM or wave function.

TABLE I. For $N=50$ and $V=0.9$, 0.99, and 0.999 the errors in the ground-state energy from variational 2-RDM theory with 2-, 4-, and 6-positivity (2POS, 4POS, and 6POS) are compared with errors from coupled cluster methods with 2-, 4-, and 6-particle excitations (CC2, CC4, and CC6).

Method[a]	0.9	0.99	0.999
CC2	−2.3(−4)	−8.7(−4)	−1.0(−3)
2POS	−3.4(−4)	−8.8(−4)	−9.8(−4)
CC4	2.1(−5)	1.4(−4)	1.8(−4)
4POS	−3.7(−8)	−9.9(−7)	−1.5(−6)
CC6	−2.5(−6)	−3.6(−5)	−4.7(−5)
6POS	8.8(−10)	−5.8(−10)	−1.1(−9)

[a]Errors with parentheses denoting powers of 10 are determined with respect to following FCI energies of −1.010 638 664, −1.014 011 958, and −1.014 412 186 at $V=0.9$, 0.99, and 0.999.

A quantum phase transition is usually characterized by an actual or avoided level crossing between the energies of the ground- and excited-state wave functions with an accompanying significant change in one or more expectation values called order parameters. With 2-RDM mechanics quantum phase transitions can be characterized by a complementary approach. The information contained in the 2-RDM is equivalent to the expectation values of all one- and two-body operators. At the critical point of a quantum phase transition one or more of these expectation values displays dramatic changes. In the language of convex sets a change in a Hamiltonian parameter across a critical point corresponds to a dramatic movement of the ground-state 2-RDM along the boundary of the convex set where the initial and final points on the boundary of the set are distinguished by significant changes in each of the order parameters. In a second-order phase transition from an avoided level crossing, the ground-state 2-RDM for all *V* is a unique extreme point on the boundary of the set. The signature of a second-order quantum phase transition is rapid movement of the 2-RDM as a function of *V* along the boundary of the set. In a first-order phase transition from a level crossing the ground-state 2-RDM is degenerate at the critical point, meaning that it can be any convex combination of two extreme points on the boundary that differ significantly in one or more order parameters. The first-order phase transition, therefore, can be visualized as a movement of the 2-RDM through the interior of the convex set between extreme points with concurrent changes in the order parameters.

In the thermodynamic limit, a quantum phase transition occurs in the Lipkin model around $V=1$. Circles on the dashed curve for the 2-positive set in Fig. 1 denote the position of the 2-RDM as a function of the parameter *V* in the

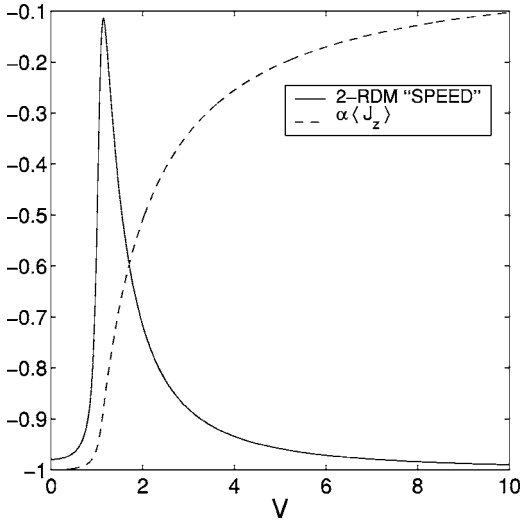

FIG. 2. Both the order parameter $\alpha\langle\hat{J}_z\rangle$ and the "speed" of the 2-RDM, measuring the movement of the 2-RDM on the boundary of the convex set of 2-positive 2-RDMs, show significant changes around the critical point $V\approx 1$ of the phase transition.

Lipkin Hamiltonian. Between $V=0$ and $V=1.0$ the position of the 2-RDM on the boundary of the convex set does not change significantly, but from $V=1.0$ to $V=1.5$, where there is a second-order quantum phase transition, the 2-RDM markedly alters its position on the boundary of the set. After the point $V=1.5$ the derivative in the 2-RDM's position with V decreases. Between $V=5$ and $V=200$ the position of the 2-RDM changes less than between $V=1.0$ and $V=1.5$. In general, the movement of the 2-RDM in the convex set collectively represents the changes in all of the order parameters. Figure 2 shows the order parameter $\alpha\langle\hat{J}_z\rangle$ and the "speed" of the 2-RDM defined as $(\vec{v}\cdot\vec{v})^{1/2}$ where $\vec{v}=(\partial\langle\hat{J}_z\rangle/\partial V,\partial\langle\hat{J}_+^2+\hat{J}_-^2\rangle/\partial V)$. The *peak* in the 2-RDM "speed" defines the location of the critical point. For the Lipkin model with 50 fermions the maximum speed occurs at $V=1.14$. As $N\rightarrow\infty$, calculations show, the peak approaches $V=1$ which is the location of the critical point in the thermodynamic limit for the model. In the $N\rightarrow\infty$ limit the derivative of the 2-RDM "speed" becomes discontinuous which is consistent with a second-order phase transition. Although we illustrate RDM mechanics for quantum phase transitions with the Lipkin model, the concept of 2-RDM "speed" is generally applicable to quantum systems with an arbitrary number of degrees of freedom. For a 2-RDM with n degrees of freedom the velocity vector generalizes to an n-component vector where each component is the derivative of a degree of freedom with respect to a Hamiltonian parameter like V.

IV. DISCUSSION

A complementary, alternative approach to quantum phase transitions has been developed through 2-RDM mechanics. In wave mechanics the quantum phase transition is characterized by a crossing or avoided crossing between the energies of the ground- and excited-state wave functions. In 2-RDM mechanics the quantum phase transition is characterized by the movement of the ground-state 2-RDM within the convex set of N-representable or p-positive 2-RDMs. For a second-order phase transition, as in the Lipkin model, the 2-RDM moves rapidly along the boundary of the convex set within the vicinity of the phase transition where the movement reflects significant changes in one or more order parameters. The "speed" of the 2-RDM movement in the set peaks at or nearby the critical point. In a first-order phase transition the ground-state 2-RDM at the critical point can move through the convex set between two extreme points with markedly different order parameters.

The ground-state energy and properties of a quantum system, such as the position of the critical point and order parameters of a quantum phase transition, can be determined from a variational calculation of the 2-RDM where the variational set of 2-RDMs is constrained by approximate N-representability conditions, known as p-positivity constraints [7]. The variational 2-RDM method has recently been applied to computing the ground-state electronic 2-RDM for a variety of atoms and molecules [8–11]. For the Lipkin model the convex set of 2-RDMs is computed with a series of p-positivity conditions in which p ranges from 2 to 6. Rapid convergence of the energy with p is observed for all interaction strengths. The 4-positivity energies are 2 to 3 orders of magnitude more accurate than the energies from renormalized perturbation theories like coupled-cluster with four-particle excitations. All variational energies are rigorous *lower bounds* to the exact ground-state energies. The 5- and 6-positivity conditions have not been previously implemented within a variational 2-RDM calculation. Because the p-positivity conditions are neither perturbative nor dependent upon a mean-field reference determinant, the 2-RDM method accurately captures the high correlation in the vicinity of the quantum phase transition.

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