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How to cite this publication:

J. Chem. Phys. **150**, 194105 (2019); <https://doi.org/10.1063/1.5095991>

Publication metadata

Title:	Excitation spectra of systems of indistinguishable particles by the autocorrelation function technique: Circumventing the exponential scaling for bosons
Author(s):	Camille Lévêque and Lars Bojer Madsen
Journal:	Journal of Chemical Physics
DOI/Link:	https://doi.org/10.1063/1.5095991
Document version:	Publisher's PDF (Version of Record)

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Submitted: 14 March 2019 . Accepted: 30 April 2019 . Published Online: 20 May 2019

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Submitted: 14 March 2019 • Accepted: 30 April 2019 •

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ABSTRACT

We consider the autocorrelation function technique for obtaining excitation spectra for indistinguishable particles. The interacting particles are described by coherent superpositions of configurations built from time-dependent spin-orbitals. The fermionic or bosonic character of the particles is taken into account by considering Slater determinants or permanents, respectively. The approach involves the calculation of overlaps between nonorthonormal Slater determinants for fermions and permanents for bosons. Efficient methods already exist for fermions. In the case of bosons, the evaluation of permanents generally scales exponentially with system size. We present an efficient approach for bosons for calculating the excitation spectrum, which circumvents this scaling. The approach is illustrated and validated by comparison with an analytical model for interacting bosons, for a system with a number of bosons so large that the autocorrelation technique could not be applied without the present development.

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I. INTRODUCTION

The role of particle-particle correlations in out-of-equilibrium few- and many-body systems has attracted considerable interest across different areas of physics and chemistry for decades. Examples of recent investigations of the role of the electron-electron and electron-nuclei correlation include single-photon double ionization dynamics in diatomic molecules^{1,2} and in photodissociation of dimers³ and organic compounds.⁴ In intense laser pulses, strong electromagnetic fields drive atomic and molecular many-electron wavefunctions and electronic dynamics and correlation effects are investigated on the attosecond time scale.⁵ In condensed-matter systems, experiments have investigated correlation times in nuclear spin baths with quantum dots.⁶ In cold-atom physics, quenching of the interaction between particles and temporal modification of trapping potentials have been used.^{7–12}

Common to the investigations listed above is the need for fully time-dependent theory that accounts for correlations, i.e., for

effects not captured by mean-field approaches. If the system Hamiltonian H_0 could be diagonalized, such a time-dependent propagation would be straightforward in the eigenstate basis of H_0 . More often than not, the dimensionality of the system under consideration is, however, so large that a diagonalization of H_0 is impossible. Alternative ways to perform the time-dependent propagation and to retrieve information about the energies and states involved are therefore needed. Thus, as a viable alternative, wavepacket dynamics approaches, with optimized time-dependent orbitals minimizing the number of configurations, are used.¹³ In the multiconfigurational time-dependent Hartree (MCTDH) setting, the autocorrelation method, discussed in Sec. II, has been used for decades to access energies of molecular nuclear degrees of freedom treating the nuclei as distinguishable particles, i.e., not accounting for the particle statistics. For instance, it was used to compute absorption spectra of molecules with wavepacket propagation methods.^{14–17} Once the autocorrelation function is evaluated, the energies and relative populations of the states involved in a particular

dynamics can be obtained from the Fourier transform of the autocorrelation function following a single time-propagation. Nonetheless, for fermions or bosons described by coherent superpositions of time-dependent Slater determinants or time-dependent permanents, i.e., when accounting for the spin-symmetry of the indistinguishable particles, the practical application of the autocorrelation technique is to some extent challenged by the need to evaluate time-dependent overlaps between nonorthogonal Slater determinants or permanents as a function of time. The evaluation of overlaps between nonorthogonal Slater determinants has been considered,^{18,19} and the computational cost remains tractable with the number of correlated fermions; see, for instance, Refs. 20–22. The autocorrelation function for electronic wavefunctions has been computed in the framework of trajectory-guided random grids of coupled wavepackets for H₂²³ and electronic energies with chemical accuracy were obtained. More recently, the autocorrelation function of a Hartree-Fock based electronic wavefunction was used to interpret proton dynamics in water complexes.²⁴ For bosons, the exponential scaling of the computational cost with respect to the number of particles has so far prevented the evaluation of the autocorrelation function for a many-body wavefunction (see Sec. II C). Alternative approaches, such as linear response theory within the Gross-Pitaevskii approximation^{25,26} or equivalently the Bogoliubov approximation,²⁷ were considered to access excitation spectra of weakly interacting bosons. More recently, linear response theory has been used with a multiconfigurational ansatz to take into account correlation effects and fragmentation^{28–31} but remains challenging computationally for many-body systems. For this reason, we present in this work a recipe for obtaining the spectrum for bosons using the autocorrelation function, with a significant reduction in the computational cost with respect to the number of particles.

The paper is organized as follows. For completeness, Sec. II A briefly reviews the autocorrelation technique. Following the work of Löwdin,¹⁸ we provide in Sec. II B the formulas for the overlaps involved in the evaluation of the autocorrelation function. Section II C presents a discussion of the scaling of the computational cost involved in the evaluation of the overlaps and underlines the need for a new approach to handle the case of bosons. Section II D presents a derivation of such an approach by evaluating the autocorrelation function using a mean-field Gross-Pitaevskii solution as reference. This method removes the exponential scaling, and excitation spectra can be calculated if the many-boson wavefunction can be propagated. Section III gives results validating the approach in an application that cannot be easily treated without the present approach. Section IV concludes and provides an outlook.

II. THEORY AND DISCUSSION

A. Autocorrelation technique

Let us briefly recapitulate the autocorrelation function technique. To this end, consider the case where an external interaction induces dynamics in a many-body system and is switched off at a time, t_0 , which we take as zero for convenience, $t_0 = 0$. At times $t > t_0$, the system evolves under the action of the system Hamiltonian, H_0 , alone and there are no further transitions between the

eigenstates. The time-dependent wavefunction of the system can be formally, but, as mentioned in the Introduction, often not practically, expanded on the eigenstates of H_0 , satisfying $H_0|\psi_j\rangle = E_j|\psi_j\rangle$. Accordingly, at time $t = 0$, the wavefunction can be expressed as

$$|\Psi(0)\rangle = \sum_j c_j(0)|\psi_j\rangle. \quad (1)$$

At a later time t , the exact wavefunction can be expressed as

$$|\Psi(t)\rangle = \sum_j c_j(0)e^{-iE_j t/\hbar}|\psi_j\rangle, \quad (2)$$

with $c_j(0)$ being the expansion coefficients, which are determined by the external interaction. Equation (2) shows how the knowledge of E_j and $|\psi_j\rangle$ allows the prediction of the time-evolution. The autocorrelation function at a time t is defined as

$$a(t) = \langle\Psi(0)|\Psi(t)\rangle. \quad (3)$$

Inserting Eq. (2) in Eq. (3), we see that $a(t)$ only depends on the expansion coefficients and the energies, as $a(t) = \sum_j |c_j(0)|^2 \exp(-iE_j t/\hbar)$. The Fourier transform of $a(t)$ reads

$$\tilde{a}(E) = \sum_j |c_j(0)|^2 \delta(E - E_j) \quad (4)$$

and is a sum of Dirac delta functions nonvanishing at the eigenvalues of the Hamiltonian and weighted by the norm squared of the coefficients in Eq. (2). The accuracy of the energies is subject to the Fourier constraint between time and energy variables. The evaluation of the Fourier transform over a finite time-interval of the autocorrelation function gives the eigenenergies as peaks in the spectrum as a function of the Fourier variable E . Moreover, the population $|c_j(0)|^2$ in a given state $|\psi_j\rangle$ induced by the external interaction is given by the height of that peak. The evaluation of $a(t)$ requires (i) a description of $|\Psi(t)\rangle$ and (ii) the evaluation of its overlap with $|\Psi(0)\rangle$, as expressed in Eq. (3). The evaluation of $a(t)$ does not require the expansion (2) be at hand, and this is important for practical applications.³²

Let us now see how the autocorrelation function is expressed in terms of quantities that enter a practical calculation. We consider time-dependent wavefunction-based methods that use a set of single-particle spin-orbitals to express an approximation, $|\tilde{\Psi}(t)\rangle$, to the true wavefunction as a linear combination of configurations, $|\Phi_J(t)\rangle$,

$$|\tilde{\Psi}(t)\rangle = \sum_J C_J(t)|\Phi_J(t)\rangle, \quad (5)$$

where each $|\Phi_J(t)\rangle$ describes a certain arrangement of the particles in a given number of spin-orbitals. The bosonic (fermionic) symmetry of the wavefunction is ensured by expressing the configurations as permanents (Slater determinants). The configurations are time-dependent (time-independent) if time-dependent (time-independent) spin-orbitals are used. For a sufficiently large number of configurations and spin-orbitals, the wavefunction $|\tilde{\Psi}(t)\rangle$ converges to the exact wavefunction $|\Psi(t)\rangle$. At this point, it is useful to recall a few characteristics of time-dependent wavefunction-based theories. In the mean-field case, the time-dependent wavefunction is expressed by a single configuration.^{25,26,33} Beyond mean-field, often

configuration-interaction expansions are used. The configuration-interaction coefficients are time-dependent, while the configurations are built from either time-independent or time-dependent spin-orbitals. In the case of time-independent orbitals, the exponential growth of the number of configurations with an increasing number of spin-orbitals is a major bottleneck, even when constraints on the allowed excitations are introduced.^{34–36} In the case of time-dependent orbitals, the evolution of the system can be described with a much smaller number of time-dependent spin-orbitals, optimized in a self-consistent manner in each time step.¹³ This approach was successfully applied to electrons in atoms and molecules,^{37–40} cold bosons⁴¹ and fermions,⁴² and mixtures of fermions and bosons.^{43–46} Further reduction of the number of configurations is possible by specification of restricted-active-space schemes.^{47–55} Due to the favorable number of spin-orbitals and configurations, theories based on time-dependent spin-orbitals are promising among time-dependent wavefunction-based theories for a description of correlation in few- and many-body systems. Therefore, it is particularly relevant to develop methods for such theories to extract eigenenergies and populations of the eigenstates involved in a specific dynamics.

With the wavefunction (5), $a(t)$ reads

$$a(t) = \sum_I \sum_J C_I^*(0) C_J(t) \langle \Phi_I(0) | \Phi_J(t) \rangle, \quad (6)$$

where the overlap between two configurations at different times needs to be evaluated. For time-independent spin-orbitals, this overlap is evaluated only once, and if the spin-orbitals are orthonormal, the evaluation is trivial. Also in the case of distinguishable particles, i.e., when the configurations are not permanents or Slater determinants and do not account explicitly for the symmetry of the considered particles, the overlaps entering Eq. (6) have been successfully evaluated in applications of the MCTDH approach mentioned in the Introduction. The usage of the autocorrelation technique, when taking the bosonic or fermionic character of indistinguishable particles into account, seems to be more limited. In this latter case, the required overlaps can, as discussed in Sec. II B, be formally treated following the work of Löwdin.¹⁸

B. General formulas for overlaps between configurations

The overlaps between two nonorthogonal Slater determinants were considered many years ago.¹⁸ In this section, we follow Löwdin's approach for the fermionic case and suitably modify it to permanents in the case of bosons. For the evaluation of the overlaps, we consider two general time-dependent or time-independent configurations of an N -particle system, $|\Phi_I\rangle$ and $|\Phi_J\rangle$, built from two distinct sets of M spin-orbitals, $\{|\phi_{I_i}\rangle\}_{i=1}^M$ and $\{|\phi_{J_i}\rangle\}_{i=1}^M$. For time-dependent spin-orbitals, the same configuration at two different times is not orthogonal with itself and the following results cover also that case. For ease of notation, a possible dependence on time t of the configurations and orbitals is suppressed in the rest of this section. In space and spin coordinates, $z = (\mathbf{r}, \sigma)$, the two configurations are expressed as

$$\Phi_I(z_1, z_2, \dots, z_N) = \mathcal{N}_I \sum_{p \in S_N} \xi^p \phi_{I_1}(z_{p(1)}) \dots \phi_{I_M}(z_{p(N)}) \quad (7)$$

and

$$\Phi_J(z_1, z_2, \dots, z_N) = \mathcal{N}_J \sum_{q \in S_N} \xi^q \phi_{J_1}(z_{q(1)}) \dots \phi_{J_M}(z_{q(N)}), \quad (8)$$

where S_N denotes the symmetric group of degree N and $\mathcal{N}_L = 1/\sqrt{N! \prod_{i=1}^M n_{L_i}!}$, with $L = I, J$, ensures normalization. The number of particles that occupies the spin-orbital $|\phi_{L_i}\rangle$ is denoted by n_{L_i} such that $\sum_{i=1}^M n_{L_i} = N$. The indices p, q run over all permutations of the indices of the coordinates z_i , and $\xi = +1$ for bosons and $\xi = -1$ for fermions. Consequently, for bosons, ξ^p is always equal to $+1$. For fermions, ξ^p is equal to $+1(-1)$ when p is an even (odd) permutation. Evaluation of the overlap in Eq. (6) yields

$$\langle \Phi_J | \Phi_I \rangle = \mathcal{N}_I \mathcal{N}_J \sum_{p \in S_N} \xi^p Ov(p), \quad (9)$$

where

$$Ov(p) = \sum_{q \in S_N} \xi^q \langle \phi_{J_1}(z_{q(1)}) | \phi_{I_1}(z_{p(1)}) \rangle \times \dots \times \langle \phi_{J_M}(z_{q(N)}) | \phi_{I_M}(z_{p(N)}) \rangle \quad (10)$$

denotes a determinant (fermions, $\xi = -1$) or a permanent (bosons, $\xi = +1$) of the overlaps between the spin-orbitals $\{|\phi_{J_i}\rangle\}$ and $\{|\phi_{I_i}\rangle\}$.

The dependence on the permutation p can be simplified since it acts as a pairwise exchange of the columns of the determinant or permanent such that in either case $Ov(p) = \xi^p Ov$, with Ov being the result obtained for an arbitrary permutation taken as reference. Accordingly, the expression (9) for the overlap between the two configurations simplifies to

$$\langle \Phi_J | \Phi_I \rangle = \mathcal{N}_I \mathcal{N}_J \sum_{p \in S_N} \xi^{2p} Ov = \mathcal{N}_I \mathcal{N}_J N! Ov. \quad (11)$$

Thus, the overlap between two configurations built from nonorthonormal spin-orbitals is proportional to the permanent (determinant) of the overlap of the spin-orbitals of the two configurations for bosons (fermions). With these overlaps at hand, Eq. (6) and hence the Fourier transform of the autocorrelation function $\bar{a}(E)$ can be evaluated, and as is standard with the autocorrelation technique, access to all eigenstates and their relative contribution to a considered dynamics can be obtained.

C. Remarks on computational costs for fermions and bosons

While the overlaps entering the evaluation of the autocorrelation function are well-defined through Eq. (11), there exists a challenge in evaluating these overlaps efficiently. In this section, we discuss the scaling of the computational cost with the number of particles, N , and configurations, N_c . In particular, we point out the difference in scaling for fermions (determinants) and bosons (permanents).

The evaluation of determinants is computationally more efficient than the evaluation of permanents. For determinants, efficient methods exist.^{20–22} For example, LU decomposition can be used and then the evaluation of a determinant requires $O(N^3)$ operations and can be simplified further using the orthogonality between the spin functions. The number of determinants that needs to be evaluated scales as the square of the number of configurations, N_c , entering the expansion in Eq. (5), and the evaluation of $a(t)$

requires $N_c^2 \times O(N^3)$ operations. The number N_c can be reduced by inspection of amplitudes and by using restrictions on the active orbital space for fermions.^{47–51,53} Therefore, atomic and molecular systems with many electrons can be investigated with the present approach.

In the bosonic case, the evaluation of $a(t)$ is trivial if the bosons are accurately described by the time-dependent mean-field Gross-Pitaevskii (GP) equation,^{25,26} i.e., when the many-body wavefunction is accurately described by the symmetric product $|\Phi_{GP}(t)\rangle = |\phi_{GP}(t), \dots, \phi_{GP}(t)\rangle$, where all the bosons are in the same single time-dependent GP orbital $|\phi_{GP}(t)\rangle$. In this case, we find the GP autocorrelation function

$$a_{GP}(t) = [\langle \phi_{GP}(t=0) | \phi_{GP}(t) \rangle]^N, \quad (12)$$

and from the point of view of computational scaling, there is no limitation on particle number N in the evaluation of $a_{GP}(t)$. When N becomes large, however, the high power in N of the overlap will induce rapidly oscillating terms that will be challenging to account for accurately in obtaining $\tilde{a}_{GP}(E)$.

In general for bosons, the evaluation of a permanent is computationally expensive; for instance, $O(N \times 2^N)$ operations are required using the Ryser-Nijenhuis-Wilf algorithm.⁵⁶ The number of permanents that needs to be evaluated scales as the square of the number of configurations N_c entering the expansion in Eq. (5); thus, the evaluation of $a(t)$ requires $N_c^2 \times O(N \times 2^N)$ operations. This exponential scaling will in practice limit the approach of Sec. II B to 10–15 bosons. The computational cost can be reduced by discarding overlaps between configurations with small amplitudes $C_I(t)$ or using the time-dependent restricted-active-space (RAS) method for bosons.⁵² Developments relying on screening of the magnitude of the overlaps entering the permanent could also be considered. Nevertheless, the severe problem with the exponential scaling, $\sim \exp(N \ln 2)$, remains. For this reason, we propose an alternative approach to obtain the bosonic ground and excited state spectrum.

D. Efficient evaluation of permanents for spectra of bosons described beyond mean-field theory

As discussed above, the evaluation of the autocorrelation function for bosons is plagued by the numerical scaling $O(N \times 2^N)$ to compute a permanent. To evaluate the autocorrelation function for large bosonic systems, we therefore use the fact that any many-body wavefunction of N particles can be expressed in the basis of the eigenstates of the Hamiltonian of interest [Eq. (2)]. Thus, following Sec. II A, the time-evolution of the wavefunction will lead to the knowledge of the eigenvalues of this Hamiltonian. It turns out that using the GP wavefunction as the initial wavefunction with solely one occupied orbital considerably simplifies the evaluation the overlaps (permanents) needed for $a(t)$. Thus, we take $|\Psi(0)\rangle = |\Phi_{GP}(0)\rangle = |\phi_{GP}(0), \dots, \phi_{GP}(0)\rangle$, with $|\phi_{GP}(0)\rangle$ being the only orbital occupied in the wavefunction at $t = 0$. During the time-evolution, more than one orbital may be occupied; i.e., $|\Psi(t)\rangle$ develops into the form of Eq. (5). The autocorrelation function simplifies to

$$a(t) = \sum_J C_J(t) \langle \Phi_{GP}(0) | \Phi_J(t) \rangle, \quad (13)$$

and a calculation of the overlaps between the permanents leads to the following closed expression for the overlap:

$$\langle \Phi_{GP}(0) | \Phi_J(t) \rangle = \sqrt{\frac{N!}{\prod_{i=1}^M n_i!}} \left(\langle \phi_{GP}(0) | \phi_1(t) \rangle^{n_1} \times \dots \times \langle \phi_{GP}(0) | \phi_M(t) \rangle^{n_M} \right). \quad (14)$$

As is clear from this equation, the computation of the permanent simplifies to the product of M overlaps between the occupied orbital at time $t = 0$, $\langle \phi_{GP}(0) |$, and the occupied orbitals in the configurations of $|\Psi(t)\rangle$. This approach improves radically the efficiency with which the autocorrelation function can be calculated and thus the calculation of the excitation spectra of many-boson systems. Indeed, the overlaps needed are simple to evaluate numerically for any number of particles, and the limit of how many bosons can be handled now depends solely on the accuracy of evaluating the factorial entering Eq. (14) and the propagation of $|\Psi(t)\rangle$ —the limiting exponential scaling of the computation of the permanents has been removed. It is important to keep in mind that the choice of the initial wavefunction does not impact the accuracy in obtaining the spectrum; only the accuracy of its time-evolution, i.e., $|\Psi(t)\rangle$, matters. The specific choice of the initial state used above implies that the populations of the different states will only be relevant for a physical situation where the initial state is well-described as an ideal Bose gas. If the populations are not of interest, the use of any symmetric initial state built from a single orbital, say $|\phi_{ref}(0)\rangle$, will lead to the same reduction in the calculation of the overlap as described by Eq. (14). Note to this end that only the eigenenergies of the eigenstates of the Hamiltonian with a nonvanishing overlap with the initial wavefunction built from the reference orbital will appear in the spectrum. Different initial reference orbitals can be used to ensure that all symmetries of the Hamiltonian are explored.

III. RESULTS

To validate the approach discussed above and to illustrate its potential, we consider the harmonic interaction model with N bosons confined in a harmonic trap.^{57–59} This model has been extensively studied^{60–66} and offers analytical solutions for the energies of the ground and excited states⁵⁸ and suffices for our present purpose. The Hamiltonian reads

$$H_0 = \frac{1}{2} \sum_{i=1}^N \left[-\frac{\partial^2}{\partial x_i^2} + \Omega^2 (x_i - x_{\min})^2 \right] + \frac{K}{2} \sum_{i \neq j}^N (x_i - x_j)^2, \quad (15)$$

with Ω and $x_{\min}$ being the trap frequency and minimum, respectively, and K being the interaction strength between the particles. The energy of the system is expressed in units of $E_0 = \hbar\Omega_0$, where Ω_0 is a reference frequency defining the system of units. For instance, a typical value for the trap frequency for ^{85}Rb atoms⁶⁷ is $\Omega_0 = 25 \times 2\pi$ Hz, which then defines the units of energy $E_0 = 2.6 \times 10^{-33}$ J, distance $x_0 = 5.5 \mu\text{m}$, and time $t_0 = 40$ ms. Note that for $K > 0$, considered in the following, the interaction is attractive, as can be realized through a Feshbach resonance^{68,69} with ^{85}Rb atoms,⁷⁰ for instance. The eigenenergies of the Hamiltonian (15) are⁵⁸

FIG. 1. Spectrum of the Hamiltonian of Eq. (15) with $x_{\min} = 0$, $\Omega = 0.5$, and $K = 0.01$ for $N = 50$ bosons. (a) Spectrum obtained from the Gross-Pitaevskii ansatz ($M = 1$ orbital) (b) and using the MCTDHB ansatz with $M = 4$ orbitals. In panels (c) and (d), comparison of the GP and MCTDHB spectra for selected energy range. The vertical dotted lines denote the analytical reference energies of Eq. (16) for values of (n, m) ranging from (2, 12) for low energy to (16, 19) for high energy.

$$E(n, m) = \left(n + \frac{N-1}{2}\right)\delta + \left(m + \frac{1}{2}\right)\Omega, \quad (16)$$

with $\delta = \sqrt{\Omega^2 + 2NK}$. The quantum numbers (n, m) describe the relative motion of the particles with respect to the center-of-mass (*c.o.m.*) and the *c.o.m.* motion, respectively.

As an application, we investigate the spectrum of the Hamiltonian of Eq. (15) with $x_{\min} = 0$, $\Omega = 0.5$, and $K = 0.01$ with $N = 50$ bosons. This number of particles is so large that it impedes the evaluation of the autocorrelation function with the formalism of Sec. II B due the exponential scaling of the computation of the permanents. Therefore, this example serves to illustrate the usefulness of the new approach of Sec. II D. As an initial wavefunction, we use the ground state of noninteracting bosons with $x_{\min} = -1.0$, obtained using a single orbital. The time-evolution is performed until $T_f = 400 t_0$, with both the mean-field Gross-Pitaevskii approximation ($M = 1$) and the MCTDH for bosons (MCTDHB) where $M = 4$ is sufficient to converge the results for the spectrum presented in Fig. 1.

The excitation protocol provides a superposition of highly excited states, ranging from 33 to 55 E_0 , for both mean-field [Fig. 1(a)] and MCTDHB [Fig. 1(b)] calculations. By looking at a portion of the spectrum, panels (c) and (d), we clearly notice that the mean-field eigenvalues are systematically shifted with respect to the analytical results (dotted vertical lines) and the MCTDHB eigenvalues, which are in good agreement with the exact analytical results. In addition to the shift in energy, we also observe additional features in the mean-field results in comparison with MCTDHB. These additional excited state energies are artifacts introduced by the GP approximation, and their presence shows that the dynamics of the system cannot be accurately described with only one orbital. We note in passing that we have conducted calculations also for smaller numbers of bosons for which the present approach and the conventional autocorrelation technique, Sec. II B, can be

compared, and again excellent agreement with the analytical results was obtained.

This application shows that a systematic study of the excitation spectra of many-boson systems can reveal the limitations of the mean-field approach in comparison with more accurate approaches. Moreover, the artificial eigenstates and the shift in energy of the eigenstates would in this case lead to a time-evolution of the system different from the exact one.

IV. CONCLUSIONS AND OUTLOOK

In this work, we discussed the use of the autocorrelation function for obtaining spectra for *indistinguishable* particles considering explicitly their bosonic or fermionic character using time-dependent permanents or Slater determinants, respectively. The numerical cost associated with the evaluation of the overlap of nonorthogonal Slater determinants, entering the evaluation of the autocorrelation function, is tractable. Efficient and accurate numerical methods exist to approximate the time-evolution of the fermionic wavefunctions, and the autocorrelation function could be used to analyze, for instance, electron dynamics. The main result of this work is the calculation of excitation spectra for bosons. The method proposed here is free of the exponential computational scaling with respect to the number of bosons that is inherent to the numerical evaluation of general permanents. The accuracy of the method depends solely on the accuracy of the time-evolved wavefunction, here described within a multiconfigurational time-dependent framework, and can be generalized to another ansatz for the wavefunction. The approach therefore allows an investigation of the impact of correlation effects on the excited state part of the spectrum, as we validated by comparing with an exactly solvable model for bosons. The illustrative example involved 50 bosons, and the permanents involved in the evaluation of the autocorrelation function could not have been computed directly within the conventional approach of Sec. II B. The drawback of the

method is the lack of information on the population of the excited states in a specific dynamics. Therefore, further investigation, for instance, considering the Fourier transform of the time-evolution of observables,⁷¹ could be considered.

Finally, let us remark that one of the advantages of the autocorrelation approach is that it can be extended to mixtures of particles of different symmetry,⁵⁴ for which no systematic approach to compute excitation spectra presently exists. For instance, the wavefunction of a system consisting of N_A particles of type A (fermions or bosons) and N_B particles of type B can be written as $|\Psi(t)\rangle = \sum_{I_A, I_B} C_{I_A, I_B}(t) [|\Phi_{I_A}^A(t)\rangle \otimes |\Phi_{I_B}^B(t)\rangle]$. The evaluation of the autocorrelation function involves products of the type $[\langle \Phi_{I_A}^A(0) | \Phi_{I_A}^A(t) \rangle \times \langle \Phi_{I_B}^B(0) | \Phi_{I_B}^B(t) \rangle]$, which can be evaluated individually by Eq. (11) and in the case of bosons by Eq. (14). A generalization to mixtures of more species is therefore straightforward.

ACKNOWLEDGMENTS

This work was supported by the Villum Kann Rasmussen (VKR) center of excellence, QUSCOPE-Quantum Scale Optical Processes. We thank Jan Arlt, Oriol Vendrell, and Elke Fasshauer for stimulating discussions. The computations were performed at the Centre for Scientific Computing, Aarhus.

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