An imaginary-time nonuniform mesh method for diagonalizing multidimensional quantum Hamiltonians is proposed and used to find the first 50 eigenstates and energies of up to $D = 5$ strongly interacting spinless quantum Lennard-Jones particles trapped in a one-dimensional harmonic potential. We show that the use of tailored grids allows exploitation of the symmetries of the system (in our case the $D!$ degeneracy derived from all possible permutations of distinguishable particles), reducing drastically the computational effort needed to diagonalize the Hamiltonian. This leads to a favorable scaling with dimensionality, requiring for the five-dimensional system four orders of magnitude fewer grid points than the equivalent regular grid. Solutions to both bosonic and fermionic counterparts of this strongly interacting system are constructed, the bosonic case clustering as a Tonks-Girardeau gas of an ensemble of up to five distinguishable Lennard-Jones (LJ) spinless particles trapped in a one-dimensional harmonic potential in the Tonks-Girardeau regime. Once the states are obtained, we find, via symmetrization and antisymmetrization, that the computed excited states can be used in a thermal average to describe the melting of the LJ clusters at finite temperature.

Theoretical methods and constitute a formidable challenge. A very rich testing ground for such methods was provided by the observation of new quantum phases at ultracold temperatures in finite and homogeneous systems [22–27], and by the development of optical lattices where ultracold atoms are trapped [28]. Due to their fascinating structural and dynamical properties, special attention has been recently devoted to the strongly interacting Tonks-Girardeau regime in one-dimensional traps [29–34].

Rigorous description and explanation of the new physics found in these well-controlled experiments require accurate theoretical methods and constitute a formidable challenge [35], the main technical difficulty being the scaling of numerical algorithms with the number of dimensions $D$. Indeed, standard algorithms scale exponentially with the dimension [36]. Improved methods addressing this difficulty in the case of stationary states include the discrete-variable representation (DVR) [1,7,8], the collocation method [2], the phase-space method based on von Neumann lattices [3,4], variational or diffusion quantum Monte Carlo (MC) methods [5,22], density functional theory (DFT) [6,22], mean-field or pseudopotential interaction models [9–11], and many others. Some of these methods find only the ground state of the time-independent MDSE, using different efficient techniques such as imaginary-time (IT) propagation [37] or the variational principle [38]. Methods for real-time quantum dynamics include the time-dependent DVR [12], DFT [13], mean-field approaches [14], trajectory-based methods such as Bohmian dynamics [15,16], and the time-dependent density-matrix renormalization-group method [17], which has proven to be very efficient in one-dimensional geometries. Despite many accomplishments in special cases, finding excited states of a general high-dimensional Hamiltonian remains a difficult computational challenge.

In this paper we propose a general method, scaling favorably with the dimension, which is able to solve the time-independent MDSE numerically exactly for ground and excited states. Obviously, the proposed IT nonuniform mesh method (ITNUMM) is not intended to replace other well-established approaches; instead we expect it to have a domain of applicability where other methods present more technical difficulties, such as in finding excited states of many-dimensional systems and where efficiency is more important than high accuracy. To show that the ITNUMM achieves these goals, we apply it to find the wave functions of the first 50 states of an ensemble of up to five distinguishable Lennard-Jones (LJ) spinless particles trapped in a one-dimensional harmonic potential in the Tonks-Girardeau regime. Once the states are obtained, we find, via symmetrization and antisymmetrization, the solutions for Bose-Einstein and Fermi-Dirac statistics, respectively, and observe fermionization in the bosonic case. We demonstrate that the ITNUMM belongs to the category of methods, such as the DVR or von Neumann lattices, that can exploit symmetries of the system to reduce computational cost. For $D$ indistinguishable LJ particles confined to a one-dimensional (1D) geometry, the computational cost is reduced enormously by taking advantage of the $D!$ degeneracy due to permutations of identical particles. We also show that the computed excited states can be used in a thermal average to describe the melting of the LJ clusters at finite temperature.

The derivation of our method starts by rewriting the time-dependent MDSE [38]

$$i\hbar \frac{d}{dt}\psi(t) = \mathcal{H}\psi(t),$$

where $|\psi(t)\rangle$ is the quantum state at time $t$ of the $D$-dimensional system described by the Hamiltonian $\mathcal{H}$, in terms of the quantum propagator $K(q,q';t-t')$.
corresponds to the classical Boltzmann distribution of we show that Eq. (3) simplifies in the IT scheme if this density \( H \) is known, while \( H_\mathrm{t} = H_0 + H_\mathrm{t} \) is any many-body potential depending only on \( q \). For very short time intervals \( t - t' = \Delta t \), the time evolution operator can be split to first order as [39]

\[
e^{-i\Delta t\hat{H}_0/\hbar} = e^{-i\Delta t\hat{H}_0/\hbar} e^{-i\Delta t\hat{H}_\mathrm{t}/h} + O(\Delta t^2),
\]

and one can write

\[
\psi(q, t') = \int dq' K_0(q, q'; t - t')\psi(q', t'),
\]

where \( K_0 \) is the propagator of \( \hat{H}_0 \), which is assumed to be known explicitly.

The \( \{q\} \) basis is discretized as

\[
\int dq \psi(q) \psi(q') = \lim_{N \to \infty} \sum_{j=1}^{N} w(q_j) \langle q_j | q \rangle,
\]

where \( w(q_j) \) is a weight function defined as \( w(q) = [N p(q)]^{-1} \), \( p(q) \) being the density distribution of the \( q \). Since our main interest is in finding the stationary states of \( \hat{H}_\mathrm{t} \), in the following we will assume that (i) \( \psi(q, t) = e^{-i t \hat{H}_t/\hbar} \phi_n(q) \) where \( \phi_n(q) \) and \( E_n \) are the \( n \)th eigenstate and eigenenergy of the Hamiltonian \( \hat{H}_\mathrm{t} \), and (ii) the evolution is performed in IT \( (\Delta t \to -i \Delta t) \). Although the density \( p(q) \) is arbitrary, below we show that Eq. (3) simplifies in the IT scheme if this density corresponds to the classical Boltzmann distribution of \( \hat{H}_\mathrm{t} \), namely, if

\[
p(q) = Z_{\hat{H}_\mathrm{t}}^{-1} e^{-\Delta t \hat{H}_\mathrm{t}(q)/\hbar},
\]

where \( Z_{\hat{H}_\mathrm{t}} = \text{Tr} e^{-\Delta t \hat{H}_\mathrm{t}/\hbar} \) is a normalization constant. Under these conditions, Eq. (3) reads

\[
e^{-\Delta t E_n/\hbar} \phi_n(q_j) = Z_{\hat{H}_\mathrm{t}} \frac{1}{N} \sum_{k=1}^{N} K_0(q_j, q_k; -i \Delta t) \phi_n(q_k).
\]

By defining the vector \( \Phi_n := \{\phi_n(q_j)\}_{j=1}^{N} \), whose \( j \)th component is the wave function evaluated at position \( q_j \), and the matrix \( \tilde{K}_j := K_0(q_j, q_k; -i \Delta t) Z_{\hat{H}_\mathrm{t}} / N \), whose elements are proportional to the propagator \( K_0 \) from \( q_j \) to \( q_k \), one can rewrite Eq. (6) as a matrix eigenvalue equation

\[
e^{-\Delta t E_n/\hbar} \Phi_n = \tilde{K} \Phi_n.
\]

This equation, central to the ITNUMM, exhibits the main advantage of our method—the problem of finding the spectrum and eigenfunctions of the original Hamiltonian \( \hat{H}_\mathrm{t} \) is reduced to sampling the classical Boltzmann distribution and diagonalizing \( \tilde{K} \) evaluated at those points. Instead of the Hamiltonian, we diagonalize the imaginary-time propagator, i.e., a matrix with analytically known and real-valued elements. Evaluation of, e.g., derivatives or Fourier transforms is not needed. Indeed, the implementation of the algorithm is rather simple since it requires only standard methods for sampling from arbitrary probability distributions and diagonalizing sparse real-valued matrices. The computational effort is also reduced by constructing a nonuniform grid in which more grid points are placed in areas where the wave functions exhibit more detailed features. In the special case of \( \hat{H}_0 = T \), \( \hat{H}_\mathrm{t}(q) \) equals the classical potential energy, \( K_0 \) is a free-particle propagator in \( D \) dimensions [38], and the matrix elements \( \tilde{K}_j \) assume the Gaussian form

\[
\tilde{K}_j = \frac{Z_{\hat{H}_\mathrm{t}} \left( \frac{m}{2\pi \hbar \Delta \tau} \right)^{D/2}}{N} \exp \left[ -\frac{m}{2\hbar \Delta \tau} (q_j - q_k)^2 \right] .
\]

Any type of grid is compatible with the ITNUMM: uniform or nonuniform; structured or unstructured; deterministic, pseudorandom, or quasi-random (such as, e.g., Sobol sequences); etc. Thus direct-product grids are not required, which ensures favorable scaling with dimensionality. Moreover, it is straightforward to test different grids, since the form of the matrix elements is independent of the grid type.

As the first application of the ITNUMM, we solved (i) the 1D harmonic oscillator [38] \( H_\mathrm{t}(q) = m \omega^2 q^2 / 2 \), using natural units for energy and position (defined by \( \hbar, m \), and \( \sqrt{\hbar / m \omega} \), respectively), and (ii) two particles of equal mass \( m \) interacting via a LJ potential \( \hat{H}_\mathrm{t}(q) = V_\mathrm{LJ}(q) \equiv \epsilon [(r_j / \sigma_j)^{12} - 2 (r_j / \sigma_j)^6] \). For the latter, we used a de Boer quantum delocalization length [40] of \( \lambda = 2^{1/6} \hbar / (\epsilon r_j \sqrt{\epsilon}) = 0.16 \), corresponding to hypothetical particles with properties between those of para-hydrogen—where quantum effects dominate—and neon—where quantum effects are present but classical behavior dominates. In both cases we have employed a structured nonuniform grid, choosing the \( q_j \) points with a deterministic algorithm. In the Supplemental Material [41], we show the grid points, eigenvalues, and several eigenstates obtained in a notebook executable in Wolfram Research’s Mathematica software.
high-frequency oscillations of these wave functions. Yet the agreement with exact results is very good for the first 150 states using \( N = 500 \) grid points, as shown in Fig. 1 for the first 50 states (the whole spectrum is shown in [41]).

As a more stringent test, we now apply the method to \( D \) LJ particles in a one-dimensional harmonic trap. Potentials \( V_1 \) and \( V_2 \) are defined by

\[
V_1(q) = \sum_{\lambda=1}^{D} \frac{1}{2} m \omega^2 q_{\lambda}^2, \quad V_2(q) = \sum_{\lambda<\mu} V_{1\mu}(|q_{\lambda} - q_{\mu}|). \tag{10}
\]

Note the enormous reduction of \( N \) in comparison with the equivalent uniform grid of \( N_0^D \) points and a further reduction (\( N_k \) vs \( N^2 \)) due to the sparsity of the matrix \( \hat{K} \), which depends on a particular value of \( \Delta \tau \). These reductions in computational cost do not compromise the accuracy, since at least three significant digits are obtained in each case with a general estimated error of \( \sigma(N) \sim N^{-\alpha} \), where \( \alpha > 1 \) (in all cases studied \( \alpha \approx 2 \) or even 3.5; details about the convergence are in [41]). Figure 2 shows the ground and 19th states for \( D = 2 \) and for the three statistics: distinguishable particles (in the above-mentioned subspace), bosons, and fermions (in the full space). The spectrum of \( \mathcal{H} \) as a function of \( D \) is shown in the left panel of Fig. 1. Remarkably, even for \( D > 1 \) the energies of the lowest excited states depend linearly on the
show the same one-body densities in position space (Fig. 1, right panel). In all three cases the densities show a well-defined structure, forming a quantum crystal. The displayed one-body density (top), the two-body correlation function (bottom), and the thermal average of the wavefunction. Left: distinguishable particles in the subspace $q_1 > q_2$; center: indistinguishable bosons; right: indistinguishable fermions.

quantum number $n$, indicating vibrations of the cluster in the harmonic trap. This linear dependence disappears at the energy of the first excited state of the LJ well, which is an unbound state reflecting the disintegration of the cluster into smaller components vibrating in the trap. The three statistics yield the same spectrum, which is a consequence of the fermionization mechanism. Indeed, the bosonic and fermionic systems have the same spectrum, which is a consequence of the fermionization scheme.

FIG. 2. (Color online) Wavefunctions $\psi_n(q_1, q_2)$ of the ground and 19th states for $D = 2$ LJ particles in a 1D harmonic trap (see text for details). Lighter (darker) color indicates positive (negative) values of the wavefunction. Left: distinguishable particles in the subspace $q_1 > q_2$; center: indistinguishable bosons; right: indistinguishable fermions.

The (unnormalized) probability distribution of the system, $p_\beta(q)$, at finite inverse temperature $\beta$ is defined as the thermal average

$$p_\beta(q) = \sum_{n=1}^{\infty} e^{-\beta E_n} |\psi_n(q)|^2, \quad (13)$$

and the corresponding one-body density $\rho_\beta(q)$ is obtained in the same way as for pure states. Figure 3 shows the one-body density (top), the two-body correlation function (bottom), and the concomitant Boltzmann factors (inset) for $D = 4$ at three different temperatures. Extrapolation was used to estimate that the combined population of states not included in the calculation is less than 1% even at the highest temperature.

The (unnormalized) two-body correlation function $g_\beta(q)$ was obtained as the inverse Fourier transform of the static structure factor

$$S(k) = \sum_{\lambda, \nu} p_\beta(q) e^{-ik(q_\lambda - q_\nu)} \prod_{\mu=1}^{D} dq_\mu. \quad (14)$$

The peaks of $g_\beta(q)$ are very localized at the lowest temperature, indicating that the particles are still bound to each other. However, at higher temperatures the peaks become broader and overlap with one another, which is a consequence of the population of unbound states and concomitant disintegration of the cluster. Together with the lack of structure of the one-body density, this can be thermodynamically understood as the beginning of melting of the quantum crystal into a liquidlike state.

To summarize, we have presented compelling evidence that the proposed method achieves the original goals. Indeed, the ITNUMM is very easy to implement and execute since the Gaussian form of the matrix elements (8) is independent of the grid type and also ensures a rapid decay with distance.
increasing the sparsity of the matrix to be diagonalized. As we have not found any a priori limitation to the applicability of the method, other systems described by the MDSE will be studied in the future.

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