

## Excited states of $^4\text{He}$ droplets

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We study low-lying excited states of  $^4\text{He}$  clusters up to a cluster size of 40 atoms in a variational framework. The ansatz wave function combines two- and three-body correlations, coming from a translationally invariant configuration interaction description, and Jastrow-type short-range correlation. We have previously used this scheme to determine the ground-state energies of  $^4\text{He}$  and  $^3\text{He}$  clusters. Here we present an extension of this ansatz wave function having a good quantum angular momentum  $L$ . The variational procedure is applied independently to the cases with  $L=0,2,4$ , and upper bounds for the corresponding energies are thus obtained. Moreover, centroid energies for  $L$  excitations are calculated through the use of sum rules. A comparison with previous calculations is also made.

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### I. INTRODUCTION

The study of liquid helium clusters is an active area of both experimental and theoretical research.<sup>1,2</sup> Since the atom-atom interaction is well known, a detailed knowledge of some ground-state properties has been obtained using several microscopic methods.<sup>3-10</sup> In particular, diffusion Monte Carlo (DMC), Green function Monte Carlo, or path integral techniques provide essentially exact the ground-state energies of  $^4\text{He}$  droplets at zero temperature. These calculations are thus a useful benchmark to test other many-body methods.

Collective states have also been studied within several theoretical approaches.<sup>7,11-15</sup> The excitation spectrum of quantum liquids is expected to be experimentally more easily accessible than the ground-state energetics. Moreover, study of the excitation spectrum varying the number of constituents has been used<sup>11</sup> to establish the onset of superfluidity in finite  $^4\text{He}$  clusters. Bulk helium becomes superfluid at very low temperatures; recently,<sup>16</sup> evidence for the occurrence of this phenomenon has been observed in finite droplets consisting of 60  $^4\text{He}$  atoms doped with the OCS molecule. Helium clusters can sustain collective oscillations of different multipolarities. Theoretical descriptions of the excited states usually follow the Feynman<sup>17</sup> treatment of compressional excitations in liquid  $^4\text{He}$ . The trial wave function of the excited state is written as the product  $F(\mathbf{R})\Psi_0(\mathbf{R})$ , where  $\Psi_0$  is the ground-state wave function,  $F$  is an excitation operator to be determined, and  $\mathbf{R}$  refers to the translationally invariant set of coordinates. The excitation operator is determined by minimizing the excitation energy; if  $\Psi_0$  is the exact wave function, a rigorous upper bound is then obtained for the excitation energy.

In this work we follow a different approach, although it could be related formally to Feynman's approach. Recently we reported<sup>10</sup> accurate variational calculations for the ground

state of  $^4\text{He}$  droplets based on trial functions which combine two-body Jastrow-type short-range correlations as well as two- and three-body correlations, coming from a translationally invariant configuration interaction (CI) description, with the role of incorporating fine details to the wave function at medium and long range. We have shown that these so-called J-CI3 wave functions represent a considerable improvement in the ground-state energy. Typically the calculated upper bounds are within 2% of the diffusion Monte Carlo ground-state energies. In this work we apply the same scheme to calculate variational upper bounds to the energy of the low-lying excited states with angular momentum different from zero.

In Sec. II we present an extension of the J-CI3 scheme to describe excited states characterized by an orbital angular momentum  $L$ . Our results for the excitation spectra of states with  $L=2$  and 4 are discussed in Sec. III. We also present a closely related approach based on sum rules, as well as a comparison with other calculations, based on the Feynman prescription. In Sec. IV we show our results for the dispersion relation and the structure factor. Finally some concluding remarks are given in Sec. V.

### II. TRIAL WAVE FUNCTION

We use a generalization of our previous variational treatment of the ground-state wave function, which has proved to be very efficient in describing the ground state of both bosonic<sup>10</sup> and fermionic<sup>18</sup> helium clusters. The trial function is based on a special linear version of the coupled cluster method (CCM),<sup>19</sup> supplemented by Jastrow correlations to control the strong atom-atom repulsion at short distances. We shall refer to this as the J-CI3 approximation, meaning the presence of two-body Jastrow correlations (J) and additive configuration interaction correlations involving up to three particles (CI3).

In a nutshell, the CCM ansatz for the wave function of an  $N$ -body system is constructed by means of the action of an exponentiated many-body operator on a reference or model state  $|\Psi\rangle = e^{\hat{S}}|\Phi\rangle$ . The operator  $\hat{S}$  is a sum of particle-hole excitations for 1 up to  $N$  bodies. The usual CCM formulation for extended systems must be modified in the case of finite systems to eliminate the spurious center-of-mass coordinate.<sup>20</sup> In the linearized version of the CCM or configuration-interaction formalism, the ansatz wave function contains only the linear terms of the exponential expansion. In coordinate representation, it may be written in the form

$$\Psi(\mathbf{R}) = \Phi_{CI}(\mathbf{R})\Phi_J(\mathbf{R}), \quad (1)$$

where  $\Phi_J(\mathbf{R})$  is the translationally invariant reference state wave function with zero angular momentum. Without loss of generality the configuration interaction operator, up to three-particle and three-hole (3p-3h) excitations, may be assumed to be

$$\Phi_{CI}(\mathbf{R}) = 1 + \sum_{i<j} f_2(r_{ij}) + \sum_{i<j<k} f_3(r_{ij}, r_{ik}, r_{jk}), \quad (2)$$

where  $f_2$  and  $f_3$  are unknown correlation functions to be determined by minimization of the Hamiltonian expectation value. To preserve Bose statistics the function  $f_3(s, t, u)$  must be symmetric in the three relative distances. Interestingly, this term may be interpreted as the linearization of a Feenberg trial wave function containing two- and three-body correlations; we shall show that a short-range term is nevertheless included in the reference state. It is worth noticing that as compared with a CCM description, the CI scheme loses the size extensivity guaranteed by the exponentiated form, and thus is not appropriate for extended systems.

In a theoretical analysis it is frequent to take as reference state the one based on a harmonic oscillator single-particle Hamiltonian, which permits a simple factorization of the center-of-mass coordinate. However, to deal with a system of strongly interacting particles it is advisable to include Jastrow correlations into the reference state. In previous work we have used the explicit parametrization

$$\Phi_J(\mathbf{R}) = \prod_{i<j} \exp\left[-\frac{1}{2}\left(\frac{b}{r_{ij}}\right)^\nu\right] \prod_{i<j} \exp\left(-\frac{\alpha^2}{2N}r_{ij}^2\right), \quad (3)$$

where the first term is a truly short-range correlation and the second term is a convenient form of representing the bosonic harmonic oscillator reference state. The parameters  $\nu$  and  $b$  are fixed according to the short-distance behavior of the two-body interaction potential; for the Aziz HFD-B(HE) potential<sup>21</sup> we have found the optimal values to be  $\nu=5.2$  and  $b=2.95$  Å, independent of the number of atoms in the cluster. The size parameter  $\alpha$  is determined by minimizing the expectation value of the Hamiltonian using a simpler trial wave function with only two-body correlations. This so-called J-CI2 wave function is written as in Eqs. (1)–(3), setting  $f_3=0$ . Depending on the size of the cluster,  $\alpha$  lies between 0.27 and 0.30 Å<sup>-1</sup>. An interesting outcome of our J-CI3 ground-state calculations<sup>14</sup> is that the ground-state en-

ergies are not very sensitive to the precise value of  $\alpha$  in a large interval around that obtained without triplet correlations. Finally, the yet unknown functions  $f_2$  and  $f_3$  can be optimally determined by minimizing the expectation value of the Hamiltonian, as will be shown below. This scheme has been applied with success to the ground-state description of small <sup>4</sup>He droplets and, adequately modified, to small <sup>3</sup>He droplets. The resulting ground-state energies are typically within 2% of those obtained within a diffusion Monte Carlo scheme.

In order to deal with  $L \neq 0$  states two options are immediately apparent. First of all, one may change the harmonic oscillator reference state and promote one particle to the nearest  $l=L$  open shell. In fact, a convenient combination of 1p-1h plus 2p-2h is required to guarantee translational invariance. Alternatively, one may change the CI operator and put there a translationally invariant operator with angular momentum  $L$ . We have found the first approach to be simpler, and thus the reference state  $\Phi^{(L)}$  has been changed to

$$\Phi_J^{(L)}(\mathbf{R}) = \prod_{i<j} \exp\left[-\frac{1}{2}\left(\frac{b}{r_{ij}}\right)^\nu\right] \prod_{i<j} \exp\left(-\frac{\alpha^2}{2N}r_{ij}^2\right) \times \sum_{i<j} r_{ij}^L P_L(\hat{r}_{ij}), \quad (4)$$

where  $P_L$  is a Legendre polynomial, with argument  $\hat{r}_{ij} \equiv (z_i - z_j)/r_{ij}$  given by the relative unit distance along the  $z$  axis of each pair  $ij$  of particles. Note that only even- $L$  states may be generated in this way. This is similar to what happens in nuclei, where the  $L=1$  resonance must have isospin  $T=1$ ; otherwise, it describes a translation of the system as a whole.

On the other hand, for the CI correlation term we take the same structure as in the  $L=0$  ground state, i.e.,

$$\Phi_{CI}^{(L)}(\mathbf{R}) = 1 + \sum_{i<j} f_2^{(L)}(r_{ij}) + \sum_{i<j<k} f_3^{(L)}(r_{ij}, r_{ik}, r_{jk}). \quad (5)$$

The index  $L$  in functions  $f_i^{(L)}$  reflects the fact that they are determined by minimizing the expectation value of the Hamiltonian with respect to the new trial state with angular momentum  $L$ , i.e.,  $\Psi^{(L)} = \Phi_{CI}^{(L)}\Phi_J^{(L)}$ . It is worth noting that even if there is a close similarity between our approach and the Feynman treatment for excited states, both methods differ in the sense that the latter uses as trial wave function the product of the excitation operator  $F(\mathbf{R})$  times the (hopefully) exact  $L=0$  ground-state wave function  $\Psi_0(\mathbf{R})$ .

Similarly to the ground-state case, the optimal  $f_2^{(L)}$  and  $f_3^{(L)}$  correlation functions could be determined by minimizing the expectation value of the Hamiltonian, with an appropriate normalization constraint, resulting in a system of coupled integrodifferential equations for the  $f_i^{(L)}$ . As a practical alternative to determine the correlation functions  $f_i^{(L)}$  we have expanded this term in a set of  $N_\beta$  Gaussians  $\{e^{-\beta r^2}\}$  in the following form:

$$\Phi_{CI}^{(L)}(\mathbf{R}) = \sum_{\mu} C_{\mu}^{(L)} F_{\mu}^{(L)}(\mathbf{R}), \quad (6)$$

TABLE I. Energies of the lowest states with angular momentum  $L=0,2,4$ , excitation energies, and absolute value of chemical potential, given in K, for different cluster sizes. The ground state corresponds to  $L=0$ .

$N$	$E_0$	$E_2$	$E_4$	$E_2 - E_0$	$E_4 - E_0$	$ \mu $
6	-2.316(7)	-0.979(13)	-0.030(17)	1.38(2)	2.29(2)	1.02(1)
8	-5.022(9)	-3.535(11)	-2.28(4)	1.49(1)	2.74(4)	1.45(1)
10	-8.488(15)	-6.906(12)	-5.412(17)	1.58(2)	3.08(2)	1.85(2)
12	-12.426(18)	-10.843(14)	-9.311(17)	1.58(2)	3.12(2)	2.09(2)
14	-16.83(2)	-15.32(2)	-13.66(2)	1.51(3)	2.97(3)	2.26(3)
16	-21.62(2)	-20.15(2)	-18.47(2)	1.47(3)	3.15(3)	2.47(3)
18	-26.86(3)	-25.31(2)	-23.56(2)	1.55(4)	3.30(4)	2.64(4)
20	-32.73(2)	-31.15(3)	-29.40(4)	1.58(4)	3.33(4)	2.87(3)
30	-63.60(6)	-62.23(7)	-60.33(7)	1.37(9)	3.27(9)	3.54(11)
40	-98.17(16)	-96.70(16)	-95.07(17)	1.5(2)	3.1(2)	3.7(2)

with

$$F_\mu = \mathcal{S} \left\{ \sum_{i < j < k} \exp(-\beta_p r_{ij}^2 - \beta_q r_{ik}^2 - \beta_r r_{jk}^2) \right\}, \quad (7)$$

where the subindex  $\mu$  refers to the ordered set of Gaussian labels  $p \leq q \leq r$ , and the symbol  $\mathcal{S}$  indicates symmetrization with respect to them.

The variational determination of the energy reduces to the solution of a generalized eigenvalue problem, which can be stated as follows:

$$\sum_{\mu_2} (K_{\mu_1, \mu_2}^{(L)} + V_{\mu_1, \mu_2}^{(L)}) C_{\mu_2}^{(L)} = E^{(L)} \sum_{\mu_2} N_{\mu_1, \mu_2}^{(L)} C_{\mu_2}^{(L)}, \quad (8)$$

The matrix elements of the norm and the potential energy are given, respectively, by the integrals

$$N_{\mu_1, \mu_2}^{(L)} = \int d\mathbf{R} |\Phi_J^{(L)}|^2 F_{\mu_1}^*(\mathbf{R}) F_{\mu_2}(\mathbf{R}) \quad (9)$$

and

$$V_{\mu_1, \mu_2}^{(L)} = \int d\mathbf{R} |\Phi_J^{(L)}|^2 F_{\mu_1}^*(\mathbf{R}) \sum_{i < j} V(r_{ij}) F_{\mu_2}(\mathbf{R}), \quad (10)$$

where  $V(r)$  is the atom-atom interaction potential. We write the matrix elements of the kinetic energy operator as

$$K_{\mu_1, \mu_2}^{(L)} = \int d\mathbf{R} |\Phi_J^{(L)}|^2 F_{\mu_1}^*(\mathbf{R}) \frac{1}{\Phi_J^{(L)}(\mathbf{R})} \left( -\frac{\hbar^2}{2m} \sum_i \Delta_i \right) \times F_{\mu_2}(\mathbf{R}) \Phi_J^{(L)}(\mathbf{R}), \quad (11)$$

so that we may use the positive definite function  $|\Phi_J^{(L)}(\mathbf{R})|^2$  as the guide for a Metropolis random walk. As we are using translationally invariant wave functions no subtraction of the center-of-mass motion is necessary.

The lowest eigenvalue solution provides a rigorous upper bound to the exact lowest energy for states with angular momentum  $L$ , and it also gives the values of the  $N_\beta(N_\beta+1) \times (N_\beta+2)/6$  unknown amplitudes  $C_\mu^{(L)}$  of the corresponding optimal wave function. The Gaussian expansion has proved

to be a very accurate representation of the correlation function provided both negative and positive values of  $\beta$  are included, with the only restriction of having a square integrable wave function. We have chosen a set of six ranges, with values  $\beta/\alpha^2 = 0, -0.05, 0.5, 1, 2, 4$ . Note that the null value gives rise to the first and second terms on the right-hand side (RHS) Eq. (5), up to a normalization constant.

### III. EXCITATION SPECTRA FOR $L=2$ AND $4$

We have solved the generalized eigenvalue problem (8) to obtain an upper bound  $E^{(L)}$  for the energies of the lowest state with angular momentum  $L=2$  and  $L=4$ . The calculations have been done for the Aziz interaction HFD-H(HE).<sup>21</sup> For each cluster size considered, the parameter  $\alpha$  has been fixed to the ground-state ( $L=0$ ) value, so that the determination of the J-CI3 correlation functions  $f_2^{(L)}$  and  $f_3^{(L)}$  has no adjustable parameter.

Our results are indicated in Table I, together with the ground-state energies previously given in Ref. 10. Excitation energies and absolute values of the chemical potential are shown in the last three columns. For a drop with  $N$  atoms the chemical potential has been explicitly calculated as the difference between the ground-state energies  $E^{(0)}(N) - E^{(0)}(N-1)$ . Our results indicate that in the considered interval of cluster sizes  $N$  all these energies follow a rough quadratic dependence on  $N$ . Bound excitations with angular momentum 2 appear for systems with ten or more atoms, whereas the threshold for bound excitations with  $L=4$  lies in  $N \approx 30$ . Moreover, the excitation energies are approximately constant in the considered interval of  $N$ . States with  $L=2$  have an excitation energy of around 1.5 K, whereas those with  $L=4$  have a value of around 3.1 K.

Useful information about the excitation spectrum may be also obtained by using sum rules, which only require knowledge of the ground-state wave function. Suppose the ground state is probed with an operator  $Q$ . Denoting by  $\Psi_n$  the kinematically accessible excited states having energy  $E_n$ , the sum rule of order  $p$  is defined as

$$M_p(Q) = \sum_{n \neq 0} (E_n - E_0)^p |\langle \Psi_n | Q | \Psi_0 \rangle|^2. \quad (12)$$

TABLE II. Centroids  $\hbar\omega_L$  (in K) calculated by means of sum rules, Eq. (17).

$N$	$\hbar\omega_0$	$\hbar\omega_2$	$\hbar\omega_4$
6	1.699(11)	1.434(8)	2.70(2)
8	2.156(16)	1.569(11)	3.03(3)
10	2.63(2)	1.695(12)	3.40(3)
12	2.78(2)	1.646(14)	3.20(3)
14	3.00(3)	1.640(16)	3.21(3)
16	3.19(4)	1.62(2)	3.21(4)
18	3.32(5)	1.68(3)	3.33(5)
20	3.57(7)	1.78(4)	3.62(6)
30	3.92(9)	1.85(5)	3.55(6)
40	3.93(10)	1.79(6)	3.38(9)

The centroid of the excitation energies is defined as the ratio between the energy-weighted  $M_1(Q)$  and the non-energy-weighted  $M_0(Q)$  sum rules. For an arbitrary operator it follows that

$$E_1 - E_0 \leq \frac{M_1(Q)}{M_0(Q)}; \quad (13)$$

i.e., the energy of the first excited state is bounded by the centroid related to  $Q$ .

The sum rules  $M_0$  and  $M_1$  can be easily calculated through ground-state expectation values of specific operators. Indeed, assuming  $Q$  real, the non-energy-weighted sum rule is written as the following ground-state expectation value:

$$M_0(Q) = \langle \Psi_0 | Q^2 | \Psi_0 \rangle - |\langle \Psi_0 | Q | \Psi_0 \rangle|^2. \quad (14)$$

In the case  $\Psi_0$  is the *exact* ground-state wave function, the energy-weighted sum rule may be written as

$$M_1(Q) = \frac{1}{2} \langle \Psi_0 | [Q, [H, Q]] | \Psi_0 \rangle. \quad (15)$$

We use our ground-state trial wave function to obtain an estimate of the centroid. We first consider excitation operators

$$\begin{aligned} Q_L &= \sum_{i<j} r_{ij}^2 \quad (L=0) \\ &= \sum_{i<j} r_{ij}^L P_L(\hat{r}_{ij}) \quad (L \neq 0), \end{aligned} \quad (16)$$

which somehow are present in the long-wavelength limit of any realistic excitation operator. Given that  $Q_L$  has a good orbital angular momentum it acts like a projector in Eq. (12) in the sense that the sum is restricted to all accessible states  $\Psi_n$  with angular momentum  $L$ . In consequence, Eq. (13) refers to the lowest excited state of angular momentum  $L$ . The calculation of

$$\hbar\omega_L = M_1(Q_L)/M_0(Q_L) \quad (17)$$

is thus an alternative way of evaluating an upper bound to the lowest excited state of angular momentum  $L$ , providing a consistency check of our previous results.

Table II displays the calculated centroid energies  $\hbar\omega_L$  for  $L=0, 2$ , and 4. Comparing with the excitation energies of Table I we see that these centroids always provide an upper bound to the corresponding excitation energies, resulting in a supplementary positive test of our trial wave function. In some cases we have explicitly calculated the contribution of the first excited state for each  $L$  to the moments  $M_0$  and  $M_1$ , obtaining more than 80% of the sum rules, which is an indication of the collectivity of these excitations. This may also be qualitatively inferred from the fact that the excitation energies are rather close to their respective centroids.

As mentioned in Sec. I, the variational determination of excited energies goes usually through the Feynman prescription, writing the trial wave function as

$$\Psi_{\text{exc}}(\mathbf{R}) = F(\mathbf{R})\Psi_0(\mathbf{R}), \quad (18)$$

where  $F$  is an excitation operator to be determined. In the case  $\Psi_0$  is the *exact* ground-state wave function, the energy of the first excited state is given by the centroid energy

$$\hbar\omega(F) = \frac{M_1(F)}{M_0(F)}, \quad (19)$$

corresponding to the operator  $F$ . One is naturally led to determine the excitation operator  $F$  by minimizing the centroid. This scheme was followed by Chin and Krotscheck<sup>7</sup> using a translationally invariant one-body function for  $F(\mathbf{R})$  and restricting their calculations to the monopole and quadrupole components of  $F$ . The ground-state wave function was obtained within a DMC calculation, so that their results provide a rigorous upper bound to the excitation energies. However, even if  $\Psi_0$  is not the exact ground-state wave function, one may still use Eq. (19) to estimate the excitation energy. Chin and Krotscheck used a trial wave function containing a two- and three-body Jastrow factor, and minimized Eq. (19) to estimate the excitation energies with  $L=0$  and 2. Another estimation for the monopolar excitations has been done by Rama Krishna and Whaley,<sup>5</sup> who used a previously determined variational wave function  $\Psi_0$  and parametrized one-body monopolar functions (19) to describe nodal excitations of states with  $L=0$ , through the minimization of the centroid (19).

It is worth noticing that the J-CI3 approach bears some similarity with Feynman's treatment for the excited state. Inspecting Eqs. (1)–(5) we realize that the J-CI3 trial function for the excited state with angular momentum  $L$  may be formally written as

$$\Psi^{(L)}(\mathbf{R}) = F(\mathbf{R})\Psi^{(L=0)}(\mathbf{R}), \quad (20)$$

where the Feynman-like excitation function is

$$F(\mathbf{R}) = \frac{\Phi_{CI}^{(L)}(\mathbf{R})}{\Phi_{CI}^{(0)}(\mathbf{R})} \sum_{i<j} r_{ij}^L P_L(\hat{r}_{ij}); \quad (21)$$

i.e., it is a many-body function which is formally determined in two steps. First are obtained the correlation functions

TABLE III. Comparison of the variational (VMC) and diffusion (DMC) Monte Carlo results of Ref. 7 with our J-CI3 results, for droplets with 20 and 40  $^4\text{He}$  atoms.

	$N=20$			$N=40$		
	$L=0$	$L=2$	$-\mu$	$L=0$	$L=2$	$-\mu$
VMC	2.72	2.26		3.53	2.04	
DMC	2.80	1.77	2.91	3.68	1.50	3.67
J-CI3		1.58	2.87		1.5	3.72
$\hbar\omega_L$	3.57	1.78		3.93	1.79	

$f_2^{(L=0)}$  and  $f_3^{(L=0)}$  of the ground state, and afterwards are obtained those of the excited state with  $L \neq 0$ . This function does not have the structure of a translationally invariant one-body function, and so the comparison with the above-mentioned previous calculations is not straightforward. We can say that, in principle, this J-CI3 wave function for the excited states spans a larger variational space.

In Table III our J-CI3 results are compared with those obtained by Chin and Krotscheck<sup>7</sup> for clusters with  $N=20$  and 40 atoms, and excitations with angular momentum  $L=0$  and 2. The row labeled VMC contains their results using a trial wave function with a harmonic oscillator factor and a Jastrow factor with two- and three-body correlation functions. It is similar to Eq. (1), apart from the three-body function. This function was used for the importance sampling of their DMC calculations, whose results are displayed in the row labeled DMC. Also the chemical potentials are displayed, and it can be seen that our chemical potentials  $\mu$  are very close to the DMC ones. This was to be expected, since the ground-state energies of the clusters in this interval of values of  $N$  are also very close, as showed in Ref. 10. It is interesting to compare the structure of the J-CI3 wave function with the VMC trial wave function used in Ref. 7. Instead of a three-body Jastrow factor, the J-CI3 wave function contains a CI factor, with two- and three-body functions which are determined by minimizing the expectation value of the energy. The J-CI3 variational space is larger than this VMC one, and this is reflected in the fact that our excitation energies are lower than the VMC ones. This statement is confirmed by the fact that for the cluster with  $N=20$  our  $L=2$  excitation energy is lower than the DMC value.

#### IV. STATIC STRUCTURE FACTOR AND DISPERSION RELATION

Excitations of a many-body system are usually studied by means of scattering experiments. In the case of helium clusters, inelastic neutron scattering could in principle be used to obtain excitation energies and transition densities. It is then useful to consider the response of the system to a probe represented by a plane-wave operator

$$Q(q) = \sum_i e^{i\vec{q} \cdot (\vec{r}_i - \vec{R})}. \quad (22)$$

The use of sum rules is useful to analyze collective properties of the system. As done before, we shall use here both the

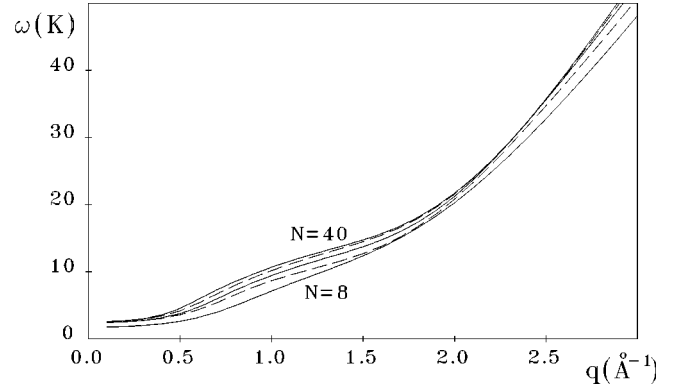


FIG. 1. Dispersion relation  $\hbar\omega(q)$  (in K) for drops with  $N=8, 12, 20, 30,$  and  $40,$  respectively, from bottom to top.

energy-weighted and the non-energy-weighted sum rules, which can be written as

$$M_1(q) = \frac{\hbar^2}{2m} q^2 \left\{ N-1 - \frac{2}{N} \left\langle \sum_{i<j} e^{i\vec{q} \cdot \vec{r}_{ij}} \right\rangle \right\}, \quad (23)$$

$$M_0(q) = N+2 \left\langle \sum_{i<j} e^{i\vec{q} \cdot \vec{r}_{ij}} \right\rangle - \left| \left\langle \sum_i e^{i\vec{q} \cdot (\vec{r}_i - \vec{R})} \right\rangle \right|^2, \quad (24)$$

where the expectation values refer to the ground-state wave function.

From these sum rules we define the dispersion relation

$$\hbar\omega = \frac{M_1(q)}{M_0(q)} \quad (25)$$

and the static structure factor

$$S(q) = M_0(q)/N. \quad (26)$$

Figure 1 displays our calculated dispersion relation for the drops with  $N=8, 12, 20, 30,$  and  $40$  atoms. The presence of the maxon in the dispersion relation is related to superfluidity, and in this respect it has been shown<sup>11</sup> that the onset of superfluidity in finite droplets appears for a number of atoms greater than  $\approx 60$ . Our results show that for  $N=40$  atoms a shoulder is insinuating in the dispersion relation at a momentum transfer  $q \approx 1 \text{ \AA}^{-1}$ .

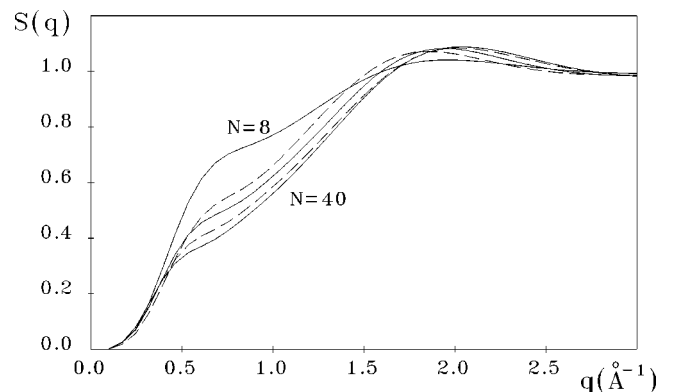


FIG. 2. Structure factor  $S(q)$  for drops with  $N=8, 12, 20, 30,$  and  $40,$  respectively, from bottom to top.

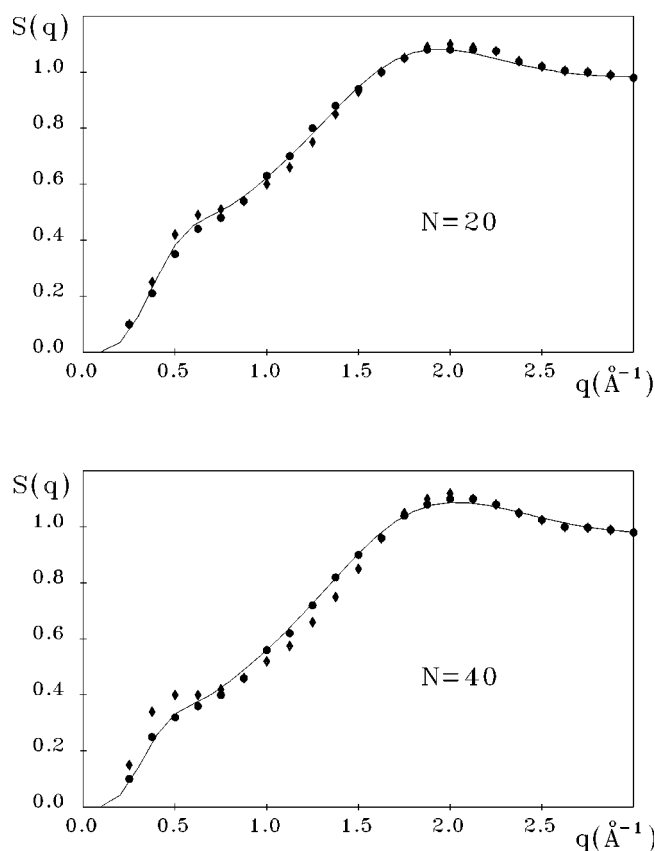


FIG. 3. Structure factor  $S(q)$  for drops with  $N=20$  and  $40$ . Dots and diamonds correspond respectively to the VMC and DMC results of Ref. 7.

Figures 2 and 3 refer to the static structure factors, the former displaying our calculated  $S(q)$  for the same number of atoms as in Fig. 1. The systems with 20 and 40 atoms are compared in Fig. 3 with the results of Ref. 7, which refer to VMC and DMC calculations. It appears that for 20 atoms, J-CI3 results (solid line) are in between VMC (dots) and DMC (diamonds) results, whereas for 40 atoms, both J-CI3 and VMC are practically coincident. This may be an indication of the limits of the present J-CI3 calculations, as compared to DMC results, already mentioned in the ground-state energy results.

## V. CONCLUSIONS

The richness and adaptability of the J-CI3 ansatz is the clue of our efficient method of describing lowest-energy states of  ${}^4\text{He}$  drops with good angular momentum quantum numbers ( $L=0, 2$ , and  $4$  have been studied here). Even if the method of obtaining the two- and three-body configuration interaction parts of the wave function by means of a nonorthogonal Gaussian basis is not strictly equivalent to an exact Euler-Lagrange method, it maintains a close analogy with it and is almost equivalent.

A minimum number of constituents is required to have  $L \neq 0$  bound states. Actually,  $N \geq 10$  are needed to have a  $L=2$  bound state, and the threshold for  $L=4$  bound excitations is close to  $N=30$ . Below these values of  $N$  the variational method provides excitation energies located in the continuum. These states could be interpreted as *virtual* states, in the language of potential scattering theories or even resonances. However, this interpretation is not rigorous. Within the statistical fluctuations, the excitation energies are roughly constant as a function of  $N$ , with a value of around  $1.5$  K for states with  $L=2$  and  $3.1$  K for  $L=4$ , thus indicating approximate vibrational spectra.

Our scheme, however, is not flexible enough to describe monopolar excitations. In that case one must ensure strict orthogonality with respect to the presumed exact ground-state wave function, as was done in Ref. 5. This condition is automatically provided in our method through the solution of the generalized eigenvalue problem. However, the obtained energies are quite unstable with respect to small changes in the Gaussian basis. We interpret this disappointing property as a signal of the lack of flexibility of our trial function family to accommodate simultaneously both the ground state and the  $L=0$  excitations.

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