Variational Study of ^{3}He Droplets

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We report variational calculations of energies of $^3\mathrm{He}_N$ droplets ($20 \leq N \leq 40$), using Aziz atom-atom interactions. The trial wave function has a simple structure, combining two- and three-body correlation functions coming from a translationally invariant configuration-interaction description, superimposed to a Jastrow-type correlated wave function with backflow. We find that the smallest bound drop has $N = 35$ atoms, and that for each N the minimum energy states have the highest spin values.

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The experimental research on liquid helium clusters has been limited up to recently by the difficulties in selecting their size. A new method based on the diffraction of a beam from a transmission grating [1] has proven to be successful in detecting clusters as small as ^{4}He trimer and dimer [2]. This experimental achievement opens interesting perspectives on the study of helium droplets. On the theoretical side ^{4}He droplets have been widely studied, and essentially exact ground state energies are provided by diffusion Monte Carlo (DMC) calculations [3]. In contrast, no DMC results are yet available for ^{3}He droplets. Such systems have been studied by means of either variational Monte Carlo (VMC) techniques [4] or phenomenological density functionals (DF) [5]. These studies have shown that there is a minimum number of atoms of ^{3}He, estimated to be between 20 and 40 atoms, to form a bound system. These numbers define two closed shells in the harmonic oscillator scheme. A shell-model calculation [6] in the open $1f2p$ shell has led to the prediction that 29 atoms is the minimum number required to form a bound system. It was also found that valence atoms couple their spins to the maximum value compatible with Pauli's principle. The calculations of Ref. [6] are based on a DF determined in the bulk, and thus its extrapolation to such light systems is not exempted from uncertainties. The purpose of this Letter is to analyze these predictions within a fully microscopic framework.

Recently we reported [7] accurate variational calculations for ^{4}He droplets based on trial functions which combine two- and three-body correlations, coming from a translationally invariant configuration interaction (CI) description, and Jastrow-type short-range correlations. Such wave functions represent a sizable improvement in the ground state energy, as compared with previous ones. The resulting upper bounds to the ground state energies are very close to the diffusion Monte Carlo ones for $^4\mathrm{He}_N$ droplets with $N \leq 40$. In this Letter we present the extension of this Jastrow CI (JCI) scheme to ^{3}He droplets.

The trial wave function is written as the product of four terms

$$\Psi(\mathbf{R}) = \Phi_\uparrow \Phi_\downarrow \prod_{i<j} g(r_{ij}) \times \left(1 + \sum_{i<j} f_2(r_{ij}) + \sum_{i<j<k} f_3(r_{ij}, r_{ik}, r_{jk})\right). \tag{1}$$

with $\mathbf{R}$ representing all single-particle coordinates.

The first two terms $\Phi_{\uparrow(\downarrow)}$ are Slater determinants, referred to as spin-up (-down) particles, built up from single-particle harmonic oscillator wave functions characterized by the inverse length parameter α. Their role is to incorporate the statistics and roughly confine the system. By using harmonic oscillator single-particle wave functions we secure the translational invariance of the full wave function. The Slater determinants include the Feynman-Cohen backflow [8] in the form proposed by Schmidt *et al.* [9] by replacing each coordinate $\mathbf{r}_i$ by

$$\mathbf{r}_i \to \mathbf{r}_i + \sum_{i \neq j} \eta(r_{ij}) (\mathbf{r}_i - \mathbf{r}_j). \tag{2}$$

In our calculations we have taken for the backflow function $\eta(r)$ the long-range form used by Pandharipande *et al.* [4]

$$\eta(r) = \frac{\lambda}{r^3}, \tag{3}$$

with the same value for the parameter $\lambda = 5$ Å^3.

The third term is the two-body Jastrow correlation factor, which we take to be of the McMillan type

$$g(r) = \exp\left[-\frac{1}{2}\left(\frac{b}{r}\right)^\nu\right]. \tag{4}$$

As previously shown for ^{4}He drops, this suffices to conveniently screen the strong short-range atom-atom repulsion. This form, with $\nu = 5$, reflects the cusp condition for a 12-6 Lennard-Jones potential. For the Aziz-type potentials HFDHE-2 [10] and HFD-B(HE) [11] considered in this Letter, we have used the values $\nu = 5.2$ Å and $b = 2.85$ Å. The parameter b has been modified with respect to the value 2.95 Å used in our previous calculations on ^{4}He systems, in order to properly account for the different masses of ^{3}He and ^{4}He atoms.

Finally the fourth term in Eq. (1) corresponds to a special version of the CI expansion describing two- and three-particle excitations, with the role of incorporating fine details to the wave function at medium and long ranges. This form is inspired by a linearized version of the coupled cluster method (CCM) [12] at the SUB(3) truncation, i.e., containing translationally invariant excitations up to three particles, three holes. The linearized version of the CCM is actually a special CI scheme [13], much more efficient than the usual CI methods when dealing with realistic interactions. It is worthwhile noticing that the present mixed JCI scheme is inspired in the correlated basis functions (CBF) method as developed by Feenberg, Clark, and Krotscheck [14] for extended systems, in which Jastrow correlations are combined with nonorthogonal perturbation theory. Our approach combines additive CI and multiplicative Jastrow correlations.

The CI correlation functions f_2 and f_3 could be determined by minimizing the ground state energy, giving rise to a system of coupled integro-differential equations for f_2 and f_3. A practical alternative is to expand these functions in a set of Gaussians:

$$1 + \sum_{i<j} f_2(r_{ij}) + \sum_{i<j<k} f_3(r_{ij}, r_{ik}, r_{jk}) = \sum_{p\le q\le r=1}^{N_\beta} C_{pqr} G_{pqr} \,, \tag{5}$$

with

$$G_{pqr} = S\left\{ \sum_{i<j<k} e^{-\beta_p r_{ij}^2} e^{-\beta_q r_{ik}^2} e^{-\beta_r r_{jk}^2} \right\}, \tag{6}$$

where S indicates symmetrization with respect to the particle labels. It is convenient to fix one of the parameters $\{\beta_p\}$, say β_1, to zero. In this way we cover the three possibilities contained in the ansatz wave function: by restricting the three labels p, q, r to 1, no CI correlations are considered; by keeping two labels equal to 1, amounts to include the linear two-body correlations; finally the unrestricted choice corresponds to the complete ansatz, with two- and three-body CI correlations included in the trial wave function. These three choices will be referred to later on as J, JCI2, and JCI3 results, respectively. Our previous experience with nuclear systems as well as ^{4}He drops indicates that the selection of the set of parameters $\{\beta_p\}$ is not very critical, as far as a sufficiently large interval of length ranges $(1/\sqrt{\beta_p})$ is included. In the calculations presented below we have used the set $\{\beta_p/\alpha^2\} = \{0, 0.5, 1, 2, 4\}$. Note that the special form of G_{pqr} does not spoil the antisymmetry nor the quantum numbers of the determinantal wave function.

To get the bound state energies one has to minimize the Hamiltonian expectation value $\langle H\rangle$ with respect to the free parameters α and $\{C_\mu\}$, where μ stands for the three labels $\{p, q, r\}$. The wave function being linear in the amplitudes $\{C_\mu\}$, their determination requires the solution of a generalized eigenvalue problem with Hamiltonian and norm matrices given by

$$\mathcal{H}_{\mu\nu} = \left\langle \Phi_\uparrow\Phi_\downarrow \prod_{i<j} g(r_{ij}) G_\mu |H| G_\nu \prod_{i<j} g(r_{ij}) \Phi_\uparrow\Phi_\downarrow \right\rangle,$$
$$\mathcal{N}_{\mu\nu} = \left\langle \Phi_\uparrow\Phi_\downarrow \prod_{i<j} g(r_{ij}) G_\mu \,|\, G_\nu \prod_{i<j} g(r_{ij}) \Phi_\uparrow\Phi_\downarrow \right\rangle. \tag{7}$$

These matrices have been computed by means of the Monte Carlo method, by carrying out a random walk guided by the probability function $|\prod_{i<j} g(r_{ij}) \Phi_\uparrow\Phi_\downarrow|^2$.

The other adjustable parameter, α, has been minimized directly. Its value is sensibly constant, around 0.24–0.25 Å^{-1} for N between 30 and 40.

A challenging question is the determination of the Slater determinants for a number of atoms between 20 and 40. These two extreme cases are trivial, the first case corresponding to the complete filling of $1s$, $1p$, $1d$, and $2s$ shells, and the second case to the completion of the next $1f2p$ major shell. Obtaining wave functions with good L and S quantum numbers for partial occupation of the last major shell is very cumbersome. Even the mere question of finding the Russell-Saunders terms is a quite complicated task because of the very large number of them (see, e.g., [15]). Certainly, some extreme cases (few particles or holes) are still manageable. In general, when the calculation is restricted to *two-body* operators, it may be solved by using standard shell-model techniques, as was done in Ref. [6], but the extension of this procedure to a fully correlated wave function would require a tremendous effort.

By limiting our scope to the determination of upper bounds one may relax the requirement of having good angular momentum quantum numbers. Keeping in mind this idea we have chosen to work in the *Cartesian coordinate* representation of the single-particle orbitals, and select the occupied levels in such a form that there is still invariance under 90° rotations with respect to the X, Y, and/or Z axes. These invariant many-particle wave functions are obtained by constructing third order monomials in coordinates x, y, z. Table I displays the orbitals and the occupation numbers for the symmetric configurations corresponding to particles with the same spin projection σ_z, for the number of particles n in the open shell. Notice that not all values of n give rise to wave functions with the above mentioned Cartesian symmetry, and those do not appear in this table.

We have performed calculations using two Aziz He-He interactions, and the results are presented in Table II. The optimal value of α is 0.25 Å^{-1}, for N greater than 33, and 0.24 Å^{-1} for the lighter systems. The oldest Aziz potential HFDHE-2 [10] has been used in order to compare

TABLE I. Occupation numbers for configurations in the $1f2p$ active shell in Cartesian coordinates giving rise to wave functions invariant under 90° rotations, as a function of the number of atoms of a given σ_z.

Orbitals	$n = 10$	$n = 9$	$n = 7$	$n = 6$	$n = 4$	$n = 3$	$n = 1$
x^3	1	1	0	0	1	1	0
y^3	1	1	0	0	1	1	0
z^3	1	1	0	0	1	1	0
x^2y	1	1	1	1	0	0	0
x^2z	1	1	1	1	0	0	0
y^2x	1	1	1	1	0	0	0
y^2z	1	1	1	1	0	0	0
z^2x	1	1	1	1	0	0	0
z^2y	1	1	1	1	0	0	0
xyz	1	0	1	0	1	0	1

our approach with the work of Pandharipande *et al.* [4], which is the only previous microscopic determination of ^{3}He drops. This work considered only closed-shell cases, obtaining the energy value 4.12 ± 0.14 K (compared with ours, 3.44 ± 0.05 K) for the system with $N = 20$ atoms, and -1.44 ± 0.08 K (ours is -2.55 ± 0.07 K) for $N = 40$ atoms. Note that this is the *total* energy of the system and not the energy per particle; in fact, the binding energies are very small compared with the experimental value of the uniform ^{3}He liquid, $E/N = -2.473$ K [16], showing that we are very close to the stability threshold. It should be mentioned that the calculations of Ref. [4] include a three-body Jastrow correlation in addition to the two-body Jastrow term and the backflow effects. The gain in energy of our approach indicates that the CI way of introducing the three-body correlations is more effective than the usual way of introducing them through a Jastrow factor. The situation is similar to what we obtained for ^{4}He drops [7], where our JCI3 energies are better than Jastrow VMC calculations with triplet correlations, being comparable to DMC results.

TABLE II. Binding energy (in K) determined at the JCI3 approximation for several $^3\mathrm{He}_N$ drops as a function of the number of spin-up ($N_\uparrow$) and spin-down ($N_\downarrow$) atoms. Results are given for the two Aziz potentials HFDHE-2 [10] and HFD-B(HE) [11].

N	$N_\uparrow$	$N_\downarrow$	S_z	HFDHE-2	HFD-B(HE)
40	20	20	0	-2.55 ± 0.07	-3.90 ± 0.07
39	20	19	1/2	-1.87 ± 0.09	-3.17 ± 0.10
38	19	19	0	-1.05 ± 0.11	-2.29 ± 0.11
37	20	17	3/2	-0.42 ± 0.08	-1.62 ± 0.09
36	20	16	2	0.06 ± 0.09	-1.09 ± 0.09
36	19	17	1	0.30 ± 0.10	-0.86 ± 0.10
35	19	16	3/2	0.76 ± 0.08	-0.33 ± 0.09
34	20	14	3	1.13 ± 0.06	0.09 ± 0.06
34	17	17	0	1.71 ± 0.06	0.67 ± 0.06
33	20	13	7/2	1.49 ± 0.09	0.56 ± 0.09
33	19	14	5/2	1.58 ± 0.08	0.66 ± 0.09
33	17	16	1/2	2.07 ± 0.09	1.15 ± 0.10
32	19	13	3	1.92 ± 0.09	1.04 ± 0.09
32	16	16	0	2.68 ± 0.07	1.81 ± 0.08
31	20	11	9/2	2.24 ± 0.07	1.42 ± 0.07
31	17	14	5/2	2.46 ± 0.09	1.62 ± 0.09
30	20	10	5	2.15 ± 0.09	1.35 ± 0.09
30	19	11	4	2.53 ± 0.07	1.73 ± 0.07
30	17	13	2	2.82 ± 0.06	2.02 ± 0.06
30	16	14	1	2.89 ± 0.06	2.09 ± 0.07
20	10	10	0	3.44 ± 0.05	3.01 ± 0.05

In Table II we present also the results obtained with the modern Aziz potential HFD-B(HE) [11]. DMC calculations using this force reproduce the experimental measurements for both ^{4}He [17] and ^{3}He liquids [18], so we believe our calculations to be real predictions for droplets. The number of particles of the smallest bound system turns out to be less than or equal to 35, a number higher than the value of 29 found in a DF plus shell-model calculation [6]. Here it should be mentioned that we are obtaining upper bounds and that we use wave functions without good angular momentum quantum numbers. There are some exceptions to the last comment in the calculations presented: the closed-shell systems ($N_\uparrow = 20, N_\downarrow = 20$) and $(10, 10)$ have obviously $L = 0$ and $S = 0$. Also our trial function for $(20, 19)$ has $L = 3$ and $S = 1/2$ (a f-shell hole state), and the $(20, 10)$ is a pure $L = 0$ and $S = 5$ system. The other cases have a spin equal to or larger than the value quoted in the table for S_z and a mixture of orbital angular momentum states. In conclusion, almost all quoted values could be improved by substituting the simple product of spin-up and spin-down Slater determinants by adequate linear combination of open-shell Slater determinants.

The last result which emerges from our calculations is the preference of the system for a wave function with the maximum value of the spin S. There are some systems in which our way of constructing the trial functions gives several configurations with different values of S_z. It can be seen in Table II that for a given number of atoms N, the lowest energy is reached for the configurations having the maximum value of S_z, even considering statistical errors. This result gives support to the predictions of Ref. [6]. The preference for high values of the spin bears some similarity with the familiar Hund's rule for atomic systems.

To conclude, we have presented a powerful method based in an efficient way of building variational self-adjustable trial functions, very much analogous to an Euler-Lagrange procedure to determine the linear two- and three-body correlations. Our calculations significantly improve previous microscopic determinations of these fermionic droplets. We find an upper bound $N = 35$ to the minimal number of constituents able to form a bound state. By improving the description of the Slater-determinantal part and/or by using DMC-like algorithms this number could be still slightly lowered. We hope our present results will stimulate further theoretical and experimental work on these peculiar finite fermionic systems.

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