Ground-state energy and stability limit of ^{3}He droplets

E. Sola, J. Casulleras, and J. Boronat

Departamento de Física i Enginyeria Nuclear, Campus Nord B4-B5, Universitat Politècnica de Catalunya, E-08034 Barcelona, Spain

Small and stable drops of ^{3}He atoms can only exist above a minimum number of particles, due to the combination of the ^{3}He atom Fermi statistics and its light mass. An accurate estimation of this minimum number using microscopic theory has been difficult due to the inhomogeneous and fermionic nature of these systems. We present a diffusion Monte Carlo calculation of ^{3}He drops with sizes near the minimum in order to determine the stability threshold. The results show that the minimum self-bound drop is formed by $N=30$ atoms with preferred orbitals for open shells corresponding to maximum value of the spin.

Liquid ^{3}He drops offer a unique combination in nature of Fermi statistics, neutral charge and self-bound character.[1,2] The interest in their physical knowledge explains the continued effort from both the experimental[3] and theoretical sides[4–8] towards a better description of their particular properties. Experimentally, ^{3}He drops are currently generated in the laboratory by means of a free jet expansion from a stagnation source chamber through a thin walled nozzle.[1] The estimated temperature of the Fermi drops is 0.15 K and therefore they are in their normal state. The nonsuperfluid character of ^{3}He drops has been detected in a series of experiments[9] where single molecules were embedded in the drops. If the molecule is surrounded by ^{4}He, the rotational spectrum presents a sharp structure which has been attributed to the superfluid nature of ^{4}He, whereas for a ^{3}He drop a broad peak is observed.

The smaller mass of the ^{3}He atom, and more fundamentally its Fermi statistics, introduces sizeable differences in the stability of the drop with respect to ^{4}He. Two ^{4}He atoms form a bound state but a minimum number of ^{3}He atoms is necessary to form a self-bound system.[5] This difference is observed in the experimental setup since very small ^{4}He drops are detected in the jet, starting from the dimer, whereas it has been proven to be difficult to observe ^{3}He drops with less than 1000 atoms.[3] Reducing this number and generating drops with a number of atoms closer to the threshold limit is one of the most important challenges for the next future. Small ^{3}He drops are expected to present "magic numbers"[4] whose experimental determination would be a clear signature of their Fermi statistics and a new benchmark for quantum many-body theories for inhomogeneous systems.

As a result of the complexity arising from the combination of Fermi statistics and inhomogeneity, the number of microscopic works on ^{3}He drops is significantly smaller than those devoted to ^{4}He drops. The first systematic study was carried out by Pandharipande *et al.*[5] in the eighties using the variational Monte Carlo (VMC) method. This calculation used a trial wave function incorporating backflow correlations to correct the nodal surface of the noninteracting system and predicted that a drop with 40 atoms is self bound. More recently, Guardiola and Navarro[7] carried out a detailed VMC calculation of small ^{3}He drops including in the trial wave function configuration-interactionlike correlations.

These new correlations improved the energy in a significant way and the smallest bound drop was estimated to be the one formed by 35 atoms. Recently, the same authors[8] have obtained a lower value (32) for this upper-bound threshold in a diffusion Monte Carlo calculation (DMC) restricted to the atom's number range 31–34. Therefore, the improvement of both the trial wave function and the theoretical approach has progressively reduced the minimum number of atoms required for a self-bound drop. It is worth mentioning that using a nonlocal density-functional approach[6] this critical number was estimated to be slightly smaller (29) than these microscopic calculations.

In this report, we present a DMC calculation of small ^{3}He drops around the threshold limit for self-binding. In the simulation we use the fixed-node (FN) approximation,[10] which provides an upper bound to the exact eigenvalue, and the release-node (RN) method[11] to estimate the quality of the FN upper bound. The approach is the same we followed in the past in the DMC calculation of the equation of state of bulk[12] and two-dimensional (Ref. 13) ^{3}He. The results show that the minimum number for a self-bound drop is 30 and that in open-shell configurations the optimal energy corresponds to maximum spin. The latter conclusion was also pointed out in previous density functional[6] and VMC work.[7]

The sign problem in a DMC simulation is usually dealt within the FN approximation.[10] Along the calculation, the wave function $f(\mathbf{R},\tau)=\psi_T(\mathbf{R})\Psi(\mathbf{R},\tau)$ $(\mathbf{R}=\mathbf{r}_1,\ldots,\mathbf{r}_N)$ evolves according to the imaginary-time (τ) Schrödinger equation, with ψ_T acting as importance sampling function and nodal constraint. For long enough time $\tau\to\infty$, $\Psi(\mathbf{R})$ is the lowest energy state compatible with the nodal surface imposed by $\psi_T(\mathbf{R})$. The trial wave function we have used has a Jastrow-Slater form

$$\psi_T(\mathbf{R})=\prod_{i<j}^{N}f(r_{ij})\Phi_\uparrow\Phi_\downarrow, \tag{1}$$

with a two-body correlation factor

$$f(r)=\exp\left\{-\frac{1}{2}\left[\frac{(\alpha r)^\beta}{N}+\left(\frac{b}{r}\right)^\nu\right]\right\}. \tag{2}$$

In Eq. (1), $\Phi_\uparrow$ ($\Phi_\downarrow$) is a Slater determinant for the spin-up (spin-down) particles filled up with single-particle orbitals

corresponding to the polynomial part of the harmonic oscillator basis. From a practical point of view, the use of this basis is clearly advantageous since the resulting Slater determinant is of the Vandermonde type and therefore translationally invariant. With this model, and considering the Jastrow part as a function of relative distances only, spurious energy contributions due to the movement of the center of mass are not present. The orbitals are chosen in its Cartesian coordinate representation, and for incomplete shells we have followed the prescription used by Guardiola and Navarro[7] which warrants invariance under 90 deg rotations with respect to the Cartesian axis.

The *quality* of the upper bound in the FN approach depends on the accuracy of the nodal surface defined by $\psi_T(\mathbf{R})$. The nodal surface of $\psi_T(\mathbf{R})$ (1) corresponds to the one of a noninteracting system. Therefore, this model is expected to be too simple for describing a correlated liquid as ^{3}He. Using the imaginary-time Schrödinger equation, one can prove that the first correction (corresponding to a short imaginary-time interval) to the noninteracting nodal surface corresponds to a displacement of the coordinates due to correlations with all the other particles.[15] These corrections are known as backflow correlations and are constructed by replacing the coordinates $\mathbf{r}_i$ of particles in the Slater determinants by

$$\widetilde{\mathbf{r}}_i = \mathbf{r}_i + \lambda_B \sum_{j \neq i}^{N} \eta(r_{ij}) \mathbf{r}_{ij}. \quad (3)$$

Similarly to previous studies of the homogeneous liquid,[12,13] we have used for the function $\eta(r)$ a Gaussian, $\eta(r) = \exp\{-[(r-r_B)/\omega_B]^2\}$; λ_B, r_B, and ω_B are variational parameters.

The FN method provides a rigorous upper bound on the ground-state energy but does not provide information on the quality of the upper bound, i.e., the difference between the energy obtained and the exact eigenvalue. In order to estimate the bias due to the particular model nodal surface we have used the RN technique.[11] In the RN method, the walkers are allowed to cross the nodal surface determined by $\psi_T(\mathbf{R})$ for a finite lifetime t_r and a sign + or − is assigned to each one. To this end, the importance sampling wave function is chosen positive

$$\psi(\mathbf{R}) = [\psi_T(\mathbf{R})^2 + a^2]^{1/2}, \quad (4)$$

with a as a constant, and the fermionic energy is obtained by projecting on the antisymmetric component $\psi_T(\mathbf{R})$. The method would arrive to the exact ground-state energy for $t_r \rightarrow \infty$, but this limit is not accesible in liquid ^{3}He due to the rapid emergence of bosonic noise. Nevertheless, the initial slope can be well determined and its value can be used for comparing different nodal surfaces and for an estimate of the magnitude of the bias introduced by the FN approximation.[12]

All the DMC simulations have been carried out using the HFD-B(HE) Aziz potential,[14] which has proved high accuracy in the microscopic description of the bulk phases of liquid ^{4}He and ^{3}He.[15] The parameters entering into $\psi_T(\mathbf{R})$, Eqs. (2) and (3), are adjusted variationally. The dependence on the number of atoms in the drop is only significant for the parameter α (2): for $N=30$, $\alpha=4.0\ \sigma^{-1}$ and for $N=40$, α

TABLE I. Total (E) and kinetic (K) energy per particle of small ^{3}He drops as a function of the number of atoms. S_z is the z component of the total spin of the drop. Figures in parentheses are the statistical errors.

N	S_z	E/N (K)	K/N (K)
29	9/2	0.0194(10)	3.395(65)
30	5	−0.0006(11)	3.630(13)
	4	0.0067(12)	3.630(15)
	3	0.0184(12)	3.595(13)
31	9/2	−0.0078(12)	3.682(18)
	3/2	0.0056(12)	3.666(13)
32	4	−0.0258(18)	3.808(15)
	3	−0.0180(12)	3.729(11)
	0	0.0003(10)	3.751(12)
33	7/2	−0.0377(13)	3.822(13)
	5/2	−0.0340(11)	3.836(14)
	1/2	−0.0190(13)	3.857(15)
34	3	−0.0535(15)	3.935(16)
	0	−0.0330(12)	3.942(16)
35	5/2	−0.0649(13)	4.02(2)
	3/2	−0.0639(16)	3.999(19)
36	2	−0.0839(15)	4.122(13)
	1	−0.0792(17)	4.124(18)
37	3/2	−0.1016(15)	4.228(17)
38	1	−0.1211(19)	4.26(3)
	0	−0.118(2)	4.241(18)
39	1/2	−0.1372(16)	4.349(16)
40	0	−0.1564(17)	4.442(17)
43	3/2	−0.1702(15)	4.583(16)
55	15/2	−0.2848(18)	5.463(16)
70	0	−0.412(2)	5.72(2)

$=4.4\ \sigma^{-1}$, increasing linearly with N ($\sigma=2.556$ Å). The optimal values of the rest of parameters are: $\beta=1$, $\nu=5$, $b=1.14\ \sigma$, $\lambda_B=0.34$, $r_B=0.75\ \sigma$, and $\omega_B=0.54\ \sigma$.

Table I contains results for the total (E/N) and kinetic energy (K/N) per particle as a function of the number of atoms N in the drop. With the exception of the first row for $N=32$, 35, and 38 the calculations have been made using orbitals with invariance under 90 deg rotations with respect to the coordinate axis. According to our results, the threshold limit for a self-bound drop is $N=30$ and all cases studied with equal N but different spin S_z show a preferred state corresponding to the maximum value of the spin. The latter result can be also interpreted taking into account the magic numbers which close a shell (of the spin-up or spin-down atoms), which in the range studied correspond to values 10, 20, and 35. The results contained in Table I show that the optimal energies follow the rule of having at least one closed shell, with preference for the smallest one: 10 for $N=29$ and 20 for $N=31$–39.

The lowest total energies of the ^{3}He drops are shown in Fig. 1 as a function of their number of atoms N. The line on top of the DMC data corresponds to polynomial fits and are
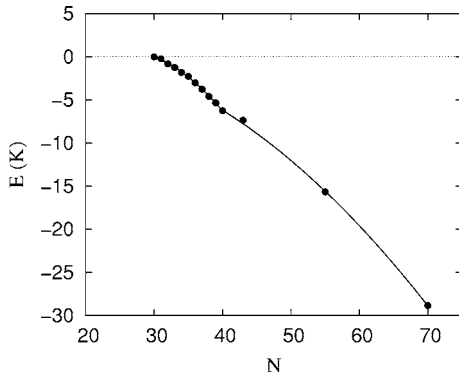
FIG. 1. Total energy of ^{3}He drops as a function of the number of atoms N. The error bars are smaller than the symbol size. The line on top of the DMC data is a guide to the eye.

only intended as to guide the eye. As one can see in the figure, the behavior of the energy with N is not monotonous in the regime studied showing a kink for $N=40$, a doubly magic number $N_\uparrow = N_\downarrow = 20$. This kink is a remnant of the shell model chosen to describe the antisymmetry of the system in absence of correlations. Dynamical correlations induced by the interatomic potential smooth significantly this effect but, for these small drops, it is still clearly observable. On the other hand, Fig. 1 shows that in the regime $N=30$–40 the DMC data display a regular behavior, which is well reproduced by a second-degree polynomial. This feature yields us confidence on the calculation itself and on the method followed for the selection of orbitals in the Slater determinants.

Although the search for an exact and stable quantum Monte Carlo algorithm for solving the N-fermion Schrödinger equation continues, the intrinsic difficulty of the problem raises the question about the maximum information one can obtain at present from the available Monte Carlo methods. The only really stable method, which can manage a significant number of fermions, is FN. With FN one is able to compute rigorous upper bounds to the ground-state energy, with the only constraint of the model nodal surface contained in ψ_T. The introduction of backflow correlations in the model has proven to be of crucial importance in order to decrease the bias introduced by ψ_T. Also for ^{3}He drops, this introduction allows for a much better description: for $N=40$, backflow correlations make the energy per particle to decrease 0.03 K, roughly a relative improvement of 25%.

As in a previous work on bulk liquid ^{3}He, we have used the RN technique to get some insight on the quality of the upper bound provided by the FN method. Although the RN method is unstable in the sense that the subjacent bosonic component asymptotically overwhelms the Fermi signal, the initial slope of the energy with the released time t_r can be determined with enough precision to make a comparison between different model nodal surfaces possible.[12] This kind of analysis is shown in Fig. 2, where the RN energies calculated with a ψ_T containing or not backflow correlations are plotted as a function of t_r. Figure 2 shows a reduction of the slope by a factor of 2 when backflow correlations are present in ψ_T pointing to a significant improvement of the upper bound (an exact wave function would show zero slope). However, this
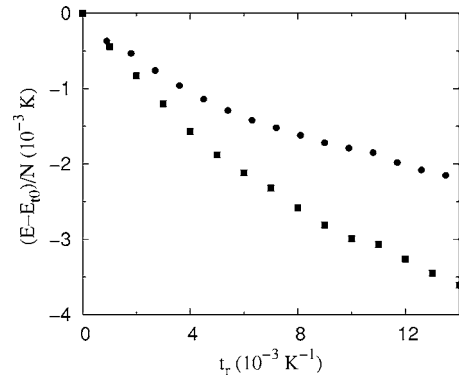

FIG. 2. Evolution of the energy with released time t_r for a $N=40$ ^{3}He drop. Circles and squares correspond to Jastrow-Slater wave functions with and without backflow correlations, respectively. In both cases, we subtract to the RN energies the energy at initial time $E_{t0} = E(t_r=0)$ for an easier comparison. The statistical error bars are essentially constant in this t_r range; they are not shown for major clarity of the released signal.

improvement is worse than the one we observed in the past in a similar calculation of bulk ^{3}He.[12] In fact, also the variance observed in the present simulation of the drops is larger than the one estimated in bulk. Both comparisons point to a somehow incomplete treatment of the inhomogeneity inherent to drops. Possible improvements on this line could be obtained by changing the functional dependence of correlations, from $(|\mathbf{r_i}-\mathbf{r_j}|)$ to $(\mathbf{r_i},\mathbf{r_j})$.[16] Notwithstanding, this modification would introduce additional complexity in the calculation and require from new correlation functions which at present are not very well known.

The present DMC results for ^{3}He drops, near the threshold limit for self-binding, are compared with recent MC data in Fig. 3. In Fig. 3, the VMC energies correspond to the best variational calculation up to date.[7] The high quality of the upper bounds there achieved comes from the introduction of configuration-interaction correlations in the Jastrow part. The resulting limit for self-binding was $N=35$, quite close to our result ($N=30$), and the difference with respect to the present results increases slightly with N. The orbitals we have used,
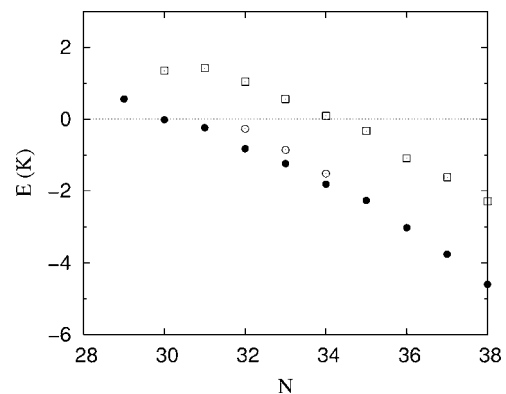

FIG. 3. Threshold limit for self-binding in ^{3}He drops. Solid circles correspond to the present results. Open circles stand for the FN-DMC calculation from Ref. 8, and squares for the VMC results from Ref. 7.

which are essentially the same used in this VMC estimation, do not have in general good angular momentum quantum numbers. In principle, that can be considered a defficiency of the model and the results obtained could be worse than the ones obtained with better wave functions. However, Guardiola[17] proved using VMC that the energies of ^{3}He drops are independent of the orbital angular moment and that they depend only on the spin.

Recently, Guardiola and Navarro[8] have reported FN-DMC results for ^{3}He drops in the range $N=31-34$ (also shown in Fig. 3). They use the same interatomic potential and the same Jastrow factor but a different shell structure in the Slater determinant, and also a different form for the backflow function. Their results show a minimal number $N=32$ for a bound drop and energies that are above our results in the number range considered. We have verified that a significant part of the difference between that calculation and the present one lies in the different functional form used for backflow correlations. In Ref. 8, the form $\eta(r)=\lambda/r^3$ instead of a Gaussian is used; for a $N=34$ drop it supposes an energy loss of ~ 0.26 K.

In conclusion, we have carried out an accurate calculation of small ^{3}He drops using FN-DMC and RN-DMC that has allowed for a reduction in the threshold limit for self-binding; our results show that this minimum number is $N=30$. Similarly to previous density-functional[6] and microscopic calculations,[7] our results confirm that the ground-state energy is achieved for maximum spin or, in other terms, for shell configurations where the number of atoms of one of the two species (up or down) closes a shell. In the N range studied, the energy shows kinks for $N=30$ and $N=40$ which correspond to magic numbers of the underlying shell model; its signal is however quite depressed by the relevance of ^{3}He-^{3}He dynamical correlations.

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