

Structure and Stability of ^3He Droplets

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We have studied the structure of ^3He droplets at zero temperature using a density functional approach plus a configuration interaction calculation in an harmonic oscillator major shell. The most salient feature of open shell drops is that the valence atoms couple their spins to the maximum value compatible with Pauli's principle, building a large magnetic moment. We have determined that 29 atoms constitute the smallest self-bound droplet. [S0031-9007(97)03450-9]

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The structure and dynamical properties of liquid helium drops have been the subject of many experimental and theoretical studies during the past 15 years (for recent reviews, see [1,2]). During this time, a severe experimental limitation has been the impossibility of selecting or detecting and identifying quantitatively small van der Waals clusters [3]. The situation has recently changed, and new scattering deflection methods seem able to determine and select the size of large helium clusters [4]. A new method based on diffracting a molecular beam from a transmission grating looks very promising in doing the same for small-size van der Waals clusters [5]. This might shed light onto the ground state structure of ^3He droplets. Prompted by this experimental possibility, we present here a realistic calculation of the structure of these systems using a reliable density functional and powerful techniques borrowed from the shell model description of the atomic nucleus (see, e.g., [6]).

The first systematic calculation of the ground state properties of ^4He and ^3He drops was carried out by Pandharipande and co-workers using a variational Monte Carlo (VMC) technique [7] and by Stringari and Treiner within a local, zero range energy density functional (LDF) approximation [8]. At present, diffusion Monte Carlo calculations are also available for ^4He drops [9,10]. For ^3He , there are also two recent systematic calculations which make use of nonlocal, finite-range density functionals (NLDF) built so as to reproduce a large number of properties of the homogeneous and inhomogeneous liquid [11,12]. Within LDF, a random-phase approximation calculation of the collective spectrum of closed shell ^3He drops is also available [13].

One of the more interesting issues of these studies is the existence of a minimum number N_{\min} of atoms below which ^3He droplets are unbound. That number was estimated to be between 20 and 40 [7], since for $N = 20$ the system was unbound and weakly bound for $N = 40$. Calculations carried out employing local or non-local functionals have reproduced this microscopic feature [8,11,12]. These two numbers belong to the $(p +$

$1)(p + 2)(p + 3)/3$ sequence with $p = 0, 1, 2, \dots$ characteristic of the harmonic oscillator (HO) well, each of them defining a major shell closure.

NLDF calculations of large drops show departures from that sequence above $N = 168$ [11,12]. These functionals are fitted to reproduce many properties of the homogeneous liquid and of its free surface, as well as Landau parameters and dynamical properties of the liquid [14], thus representing a considerable improvement over LDF [8]. Yet total energies of small drops depend to a considerable extent on the parametrization one uses. That can be appreciated in Table I, where we collect the energies of closed shell drops up to $N = 168$ obtained in [7,8,11,12] completed with those calculated with the NLDF of [14], which is an improved version of that of [12] in the sense that it better reproduces the properties of the homogeneous liquid, it is Galilean invariant in the spin channel, and yields binding energies of finite drops in better agreement with those of [7], at the price of only slightly worsening the surface tension of the liquid free surface, which is of $0.115 \text{ K}\text{\AA}^{-2}$ for the functional of [12] and $0.120 \text{ K}\text{\AA}^{-2}$ for that of [14], as compared with the experimental value of $0.113 \text{ K}\text{\AA}^{-2}$. The results we shall present here have been obtained using that functional. As compared with VMC [7], all density functional calculations yield some overbinding. However, it is worthwhile noticing that the VMC approach of [7] underbinds the homogeneous liquid by 0.13 K/atom [15].

Within NLDF, an estimate of N_{\min} may be obtained using the uniform filling approximation for open shell drops. It consists in uniformly distributing the valence atoms between the magnetic substates. This yields $N_{\min} = 34$, as can be seen in Fig. 1 (dashed line). Since correlations not included in the mean field are known to play an important role in open shell drops, that estimate may be too crude. Indeed, the interaction between different configurations arising in the $p = 3$ major shell, made of $l = 1$ and $l = 3$ orbital angular momentum substates, could produce a sizeable extra binding.

TABLE I. Total energies (K) for several closed shell ${}^3\text{He}_N$ drops. PPW results are from [7], ST from [8], WR from [11], BJHNS from [12], and BHN from [14]. The ∞ values correspond to the energy per particle in the homogeneous liquid.

N	PPW [7]	ST [8]	WR [11]	BJHNS [12]	BHN [14]
20	4.12	1.6	0.97	2.05	2.70
40	-1.44	-7.6	-9.80	-7.49	-4.28
70	-19.25	-35.0	-37.3	-33.7	-27.1
112	-51.52	-84.0	-86.5	-81.3	-69.9
168	-103.8	-159.6	-162.4	-155.0	-139.7
∞	-2.36 [15]	-2.49	-2.49	-2.49	-2.49

To make quantitative that statement, we have resorted to a configuration interaction calculation in the $p = 3$ major shell, whose basic ingredients are the two-body matrix elements of the residual atom-atom interaction, which we have taken to be the effective ${}^3\text{He}$ - ${}^3\text{He}$ interaction deduced from the NLDF of [14]. For each specific drop, the NLDF in the uniform filling approximation provides both the single particle wave functions, to evaluate the two-body matrix elements, and the single particle energies. The dimension of the m -scheme variational space (number of different Slater determinants in the space) reaches $D = \binom{20}{10} = 184\,756$ for $N = 30$. We have solved the secular problem using the code ANTOINE [16], a fast implementation of the Lanczos method mostly oriented to nuclear structure problems. This procedure takes care of relevant correlations that are absent in the uniform filling HF approximation and can accommodate possible spherical symmetry breaking. The method has been successfully applied to the description of deformed nuclei in the laboratory frame [17] using a spherical shell model basis, which plays, in that case, the same role as the uniform filling HF approximation does here.

Table II displays the most important matrix elements obtained for $N = 30$ and $N = 40$. Although in a different representation, they bear some of the characteristics of the triplet pairing matrix elements of liquid ${}^3\text{He}$ [18]: Notice that the matrix elements in a single l orbit are very re-

pulsive for $L = 0$, essentially zero for even L values, and attractive for odd L values, which, in addition, are very similar. The diagonal matrix elements involving two l orbits are much more attractive in the $S = 1$ channel than in the $S = 0$. Besides the $S = 1$ matrix elements are nearly L independent. We shall see that these features of the matrix elements completely determine the structure of open shell droplets. They are also responsible for the quantitative and qualitative differences between the present results and those of [12], where the use, for the sake of simplicity, of a zero-range ${}^3\text{He}$ - ${}^3\text{He}$ residual interaction to compute the pairing matrix elements within one active l orbit led to the incorrect prediction that small ${}^3\text{He}$ drops could display a pairing of BCS type.

The calculated correlation energies E_C are plotted in Fig. 1 (dash-dotted line). They smoothly depend on the number of particles in the shell having a pronounced minimum at midshell. Moreover, they roughly have the same value for particles and for holes. Adding the correlation to the NLDF ground state energy, we obtain the total energy, which is also shown in Fig. 1 (solid line). It is seen that correlations move the limit of stability

TABLE II. Matrix elements $\langle l_1, l_2(L) | V | l_3, l_4(L) \rangle$ (K) for $N = 30$ and 40 drops.

l_1, l_2, l_3, l_4	LS	$N = 30$	$N = 40$
3, 3, 3, 3	00	0.724	0.896
	11	-0.247	-0.228
	20	-0.019	-0.010
	31	-0.210	-0.235
	40	0.015	0.024
	51	-0.203	-0.239
1, 1, 1, 1	60	0.010	-0.003
	00	0.307	0.506
	11	-0.137	-0.155
3, 1, 3, 1	20	-0.018	0.081
	20	-0.024	-0.012
	21	-0.237	-0.264
	30	-0.197	-0.245
	31	-0.188	-0.264
	40	-0.005	-0.053
	41	-0.235	-0.287

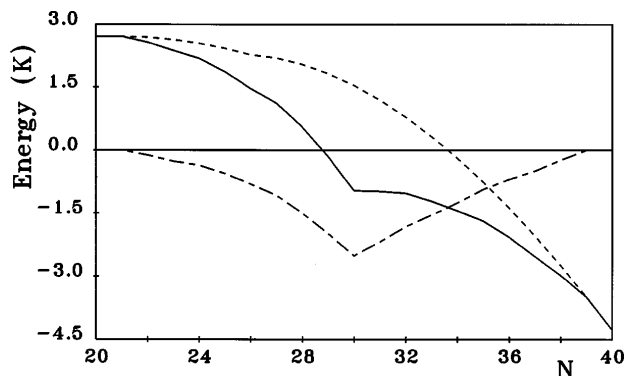


FIG. 1. NLDF energies (short-dashed line), correlation energies E_C (short-long-dashed line), and total energies (solid line) as a function of the number N of atoms in the droplet.

down by about five atomic units. Consequently, we predict $N_{\min} = 29$. The very pronounced E_C minimum at midshell allows one to state that the droplet with 30 atoms is bound, even taking into consideration the uncertainties of the NLDF mean field results, as shown in Table I. That size is in the reach of new detection techniques [5].

The structure of small droplets is also a major issue. A common feature of droplets with $N = 20$ to 40 is that their ground states have the maximum spin value allowed by Pauli's principle, i.e., $S_{\max} = \tilde{n}/2$, where \tilde{n} is the number of particles (holes) in the valence space below (above) midshell. This is due to the properties of the residual interaction discussed above: The energy minimum within one l orbit is obtained for configurations having the maximum antisymmetry in orbital space and, consequently, the maximum symmetry in spin space. In addition, due to the dominance of the spin triplet channel in the interaction between atoms in different orbits, the aligned states of each orbit couple their spins to the maximum allowed value.

Another relevant quantity is the spin gap Δ_S , defined as the energy difference between the ground state and the lowest excited state with spin $S = S_{\max} - 1$. The gap is plotted in Fig. 2. It is seen that Δ_S is roughly proportional to \tilde{n} , or equivalently to the ground state spin. Embedded in the spin gap they may lay several states with different L values. Only at midshell \pm one atom, is the spin gap empty. An explanation of this behavior is given below. Exploratory calculations carried out in the $p = 4$ and $p = 5$ major shells indicate that these features, i.e., maximum spin alignment and large E_C and Δ_S at midshell, still persist.

We can go one step further taking advantage of the specificities of the residual interaction. Looking at the two-body matrix elements in Table II, one realizes that the diagonal matrix elements $\langle l^2(L)|V|l^2(L)\rangle = V^L$ split into three quasidegenerate blocks according to their L value: $L = 0$, even $L \neq 0$, and odd L , in increasing order of attraction. Let us consider a single l orbit and assume that exact degeneracy holds within each

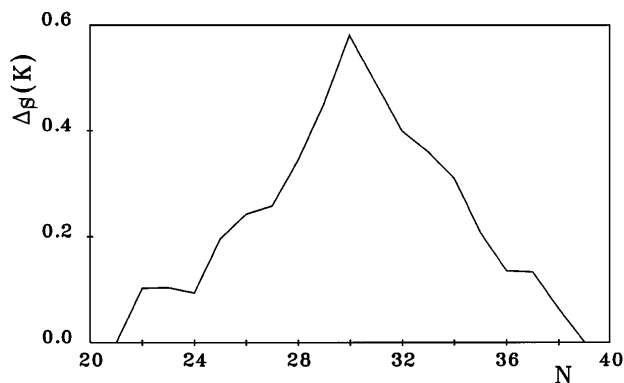


FIG. 2. The spin gap Δ_S as a function of the number N of atoms in the droplet.

block. In this case, the residual interaction has the symmetry of the group $O(2l + 1)$, a subgroup of the complete $SU(2l + 1)$. Owing to that, the states of the l^n configuration can be classified according to the representations of the group chain $SU(2l + 1) \supset O(2l + 1) \supset O(3)$ (see Ref. [19]). The group representations are such that at midshell and midshell \pm one atom the state of maximum spin contains a single L value. For the other cases, several L values are degenerate. This explains the results of the exact calculations discussed before. Furthermore, for a given spin, a formula which only involves the simplest Casimir operators of $SU(2l + 1)$ holds for the lowest energy state of each spin:

$$E(\tilde{n}, S) = \binom{\tilde{n}}{2} [\delta V - \bar{V}] + \binom{\tilde{n}}{2} \bar{V} + \frac{1}{2} \delta V \left[S(S + 1) - \frac{1}{4} \tilde{n}(\tilde{n} + 2) \right], \quad (1)$$

where

$$\bar{V} = \frac{\sum (2L + 1)(2S + 1)V^L}{\sum (2L + 1)(2S + 1)}, \quad (2)$$

$$\delta V = V^{\text{odd}} - V^{\text{even}}. \quad (3)$$

The quantities earlier defined as correlation energy E_C and spin gap Δ_S now read

$$E_C = \binom{\tilde{n}}{2} [\delta V - \bar{V}] \equiv \binom{\tilde{n}}{2} V^C, \quad (4)$$

$$\Delta_S = \frac{1}{2} \delta V \left[S(S + 1) - \frac{1}{4} \tilde{n}(\tilde{n} + 2) \right] = \frac{1}{2} \tilde{n} \delta V, \quad (5)$$

so

$$E(\tilde{n}, S) = \binom{\tilde{n}}{2} \bar{V} + E_C + \Delta_S. \quad (6)$$

For a given valence p shell of degeneracy $2\Omega_p = (p + 1)(p + 2)$, the maximum spin is predicted to be $S = (p + 1)(p + 2)/2$. In order to build up this spin, all the l orbits have to be half filled, i.e., the Ω_p valence particles are distributed as $[n_1, n_2, \dots, n_i, \dots]$ with $n_i = 2l_i + 1$. This state is, therefore, unique. In the $p = 3$ case the maximum alignment configuration is

$$(f^7[S = 7/2] \otimes p^3[S = 3/2])^{[S=5]}. \quad (7)$$

The correlation energy of the aligned configuration with n_1 particles in the orbit l_1 and n_2 particles in the orbit l_2 is now written as

$$E_C[n_1, n_2] = E_C[n_1] + E_C[n_2] + \frac{1}{4} b_{12} \tilde{n}_1 \cdot \tilde{n}_2, \quad (8)$$

where $b_{12} = [\bar{V}_{12}^{S=1} - \bar{V}_{12}^{S=0}]$. For the spin gap we have now

$$\Delta_S = \frac{1}{2} b_{12}(\tilde{n}_1 + \tilde{n}_2). \quad (9)$$

It is clear that for a given number of valence particles, the closest to half filling the occupations the largest the spin, the correlation energy, and the gap. In many cases, the ground state partition is solely dictated by the exigence of producing maximum spin.

We have applied these analytical expressions to $N = 30$ and have obtained a correlation energy of -2.66 K to be compared with the exact value -2.51 K and a gap of 0.65 K compared to the exact value 0.58 K. The agreement is striking and justifies the use of expressions (8) and (9) in the description of larger ${}^3\text{He}$ drops as far as the matrix elements of the effective interaction keep the features already discussed. Notice that neither in the single l case nor in the case of several l 's the correlation energy depends on the number of pairs. Besides, no odd-even effects are present in any of the calculated observables.

To conclude, we have shown that the inclusion of configuration interactions in the description of ${}^3\text{He}$ droplets leads to predict rather unambiguously that ${}^3\text{He}_{30}$ is a self-bound system. Near midshell, the correlation energy is substantial, and in all cases the ground state of open shell droplets corresponds to a configuration with maximum spin alignment. The existence in these droplets of a large spin will originate a sizeable magnetic moment $\mu_{\text{drop}} = gS\mu_3$, with the ${}^3\text{He}$ Bohr magneton defined as $\mu_3 = \hbar e/2m_3c$ and the gyromagnetic factor deduced from the experimental magnetic moment of a ${}^3\text{He}$ atom [20] being $g = 12.8$. This fact might be used in beam deflecting experiments in inhomogeneous magnetic fields as a complementary detecting technique.

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