Stability chart of small mixed ^{4}He-^{3}He clusters

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A stability chart of mixed ^{4}He and ^{3}He clusters has been obtained by means of the diffusion Monte Carlo method, using both the Aziz HFD-B and the Tang-Toennies-Yiu atom-atom interaction. The investigated clusters contain up to eight ^{4}He atoms and up to 20 ^{3}He atoms. One single ^{4}He binds 20 ^{3}He atoms, and two ^{4}He bind 1, 2, 8, and more than 14 ^{3}He atoms. All clusters with three or more ^{4}He atoms are bound, although the combinations $^4\mathrm{He}_3{}^3\mathrm{He}_{9,10,11}$ and $^4\mathrm{He}_4{}^3\mathrm{He}_9$ are metastable. Clusters with 2, 8, and 20 ^{3}He atoms are particularly stable and define magic ^{3}He numbers.

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Helium clusters are weakly bound quantum systems, as a consequence of both the small atomic mass and the weak van der Waals interaction between helium atoms. A detailed analysis of their structure and dynamics has been done employing microscopic many-body techniques [1–9], as well as density-functional methods [10–14]. Systems formed by a mixture of both helium isotopes are particularly interesting, since they are made of bosons and fermions with different mass interacting through the same potential. Clusters with a single fermion, $^4\mathrm{He}_N{}^3\mathrm{He}$, have been studied using a density-functional approach [15] and microscopic methods [16–18]. Such clusters form bound states for $N \geqslant 2$. The excess in kinetic energy pushes the ^{3}He atom to the surface, where its wave function becomes quasi-two-dimensional, similar to the Andreev state describing one ^{3}He impurity in ^{4}He bulk. Compounds with a large number of both ^{3}He and ^{4}He atoms have been studied employing a nonlocal finite range density functional [19,20].

The study of the lightest clusters has a great theoretical interest, as they are a challenge for theoretical methods due to their very small binding energy. Moreover, they are particularly interesting from the experimental point of view because one may expect in the near future the improvement of the detection systems [21], so as to resolve clearly the clusters of mass up to 50 amu, approximately [22]. The main objective of this paper is to present a precise determination of the stability chart of small mixed clusters with $N_B \leqslant 8$ bosons and $N_F \leqslant 20$ fermions. We have used the diffusion Monte Carlo (DMC) method, with the Aziz HFD-B [29] as well as the Tang-Toennies-Yiu (TTY) [30] He-He interactions.

In previous works [23,24] we have investigated the stability of small mixed systems by means of a variational Monte Carlo (VMC) calculation, based on a trial wave function which combines short-range correlations, described by a Jastrow factor, and medium- and long-range correlations, described by a configuration-interaction term up to pair level. We concluded that a single ^{4}He atom cannot bind a system with $N_F \leqslant 20$ fermions, and nor can two ^{4}He atoms bind a system with $3 < N_F < 17$. We also found metastability islands for some combinations with three and four bosons. However, only upper bounds to the binding energies are obtained from a variational calculation, so that the limits of the instability regions may be only approximate. Moreover, the choice of the atom-atom interaction could play a relevant role in establishing the stability boundaries.

Recently, Bressanini and collaborators [25,26] have considered systems with $N_B = 2-17$ and $N_F = 1-3$, by means of the DMC method. Apart from the fact that DMC calculations provide exact ground-state energies (or improved upper bounds, if fermions are present), whereas VMC generates only upper bounds, there are two main differences between their approach and our previous work. The first one is related to the He-He interaction: the TTY [30] interaction used in Ref. [26] gives slightly less binding energies than the HFD-B [29] interaction used in Ref. [23]. The second difference is related to the importance sampling wave function for the systems with three fermions. Bressanini and Morosi assumed an $L = 0$ configuration and concluded that the minimum number of bosons required to bind three fermions is nine. In contrast, an $L = 1$ state was considered in Ref. [23] and we concluded that all systems with $N_B \geqslant 4$ and $N_F = 3$ are bound. Obviously, as far as the instability regions are obtained from differences of ground-state energies, they may change when improving the VMC calculations with the DMC method. However, the main question addressed by this comparison refers to the quantum numbers of the ground state.

The DMC description is based in a variational or importance sampling wave function. Such a wave function is required to control the variance of the ground-state energy. Moreover, when dealing with fermions, it allows us to build up a positive-definite quantity which may be thought of as a probability distribution function. We have used a rather simple form which contains the basic required properties. It is written as a product of five factors:

$$\Psi(\mathbf{R}) = \Psi_{BB}\Psi_{FF}\Psi_{BF}D_{\uparrow}D_{\downarrow}, \tag{1}$$

containing a Jastrow form for the boson-boson (BB), fermion-fermion (FF), and boson-fermion (BF) parts, and the spin-up and -down Slater determinants required by the Pauli

exclusion principle. As compared with the trial wave function used in Ref. [23] it is simpler because it does not include the self-adjustable configuration-interaction terms. Each Jastrow part has the generic form

$$\Psi_{MN}=\prod_{i,j}\exp\left(-\frac{1}{2}\left[\frac{b_{MN}}{r_{ij}}\right]^{\nu}-\alpha_{MN}r_{ij}\right), \qquad (2)$$

where indices M,N represent bosons (B) or fermions (F), and indices i,j run over the corresponding atoms. This Jastrow part includes the short-range repulsion, related to parameters b_{MN}, and the long-range confining part, related to α_{MN}. Functions Ψ_{MN} are explicitly symmetric under the exchange of particles.

The antisymmetry required for fermions is described in the Slater determinants $D_{\uparrow}$ and $D_{\downarrow}$, related to the spin-up and -down fermions. These Slater determinants are of primary relevance, because they define the sets of spin-up and -down nodal surfaces which strongly constrain the DMC algorithm. As in our previous work [23] we have assumed a harmonic-oscillator (HO) ordering of shells, with major shells $1s$ (up to 2 fermions), $1p$ (up to 8 fermions), and $2s1d$ (up to 20 fermions). Moreover, based on our findings on pure ^{3}He clusters [7] we have assumed in all cases a filling scheme with maximum total spin, i.e., the atomic physics Hund's rule. In conclusion, the single-particle orbitals entering the Slater determinants have been taken as harmonic polynomials in terms of the Cartesian coordinates of the fermions, i.e., 1, x, y, z, x^2, y^2, z^2, xy, xz, and yz, in that order. The major shells are therefore characterized by the power 0, 1, and 2 of the harmonic polynomials. Note that the confining part of the single-particle orbitals is already included in the Jastrow part of the variational wave function.

There are some properties related to this simple choice which are worth commenting on. First, the Slater determinants are translationally invariant, having a Vandermonde-like structure. Second, up to $N_F=8$ the resulting wave functions have a well-defined orbital angular-momentum (L) value; it is a trivial consequence of the fact that configurations $1s^n$ and $1p^n$ have a unique term with maximum spin. This is not the case when filling the $2s1d$ shell ($8<N_F\leqslant 20$), and deliberately we have used single-particle wave functions which are a mixture of $2s$ and $1d$ orbitals, with the hope that the DMC procedure will select the more adequate ones.

In all cases, the short-range parameters have been fixed to the values $\nu=5.2$, $b_{BB}=2.95$ Å, $b_{FF}=2.85$ Å, and $b_{BF}=2.90$ Å, as in our previous studies. We have also included the so-called Feynman-Cohen backflow [27,28] in both the fermionic exponential and the Slater determinants, according to the form used by Pandharipande *et al.* [2]. The trial or importance sampling wave function contains thus only three free parameters α_{BB}, α_{BF}, and α_{FF} which have been determined by minimizing the ground-state expectation value of the Hamiltonian.

For the DMC algorithm we have used the short-time Green-function approximation [31,32] with an $O(\tau^3)$ form [33]. It is worth mentioning that this approximate Green function satisfies the microreversibility (detailed balance) condition. Moreover, the random process has been constrained so as not to traverse the nodal surfaces, using the *fixed-node approximation*. The simulations have been performed from an initial set of 1000 walkers, a time step $\tau=0.0005$ K^{-1}, 2000 time steps of stabilization, and 20 000 time steps for computing expectation values. In some special cases, particularly for systems with a very small binding energy, it was necessary to decrease the time step and to increase the number of steps. To evaluate the statistical error, and due to the unavoidable correlations of walkers related to the smallness of the time step, we have used a block average, grouping sets of successive steps in blocks. The size of the blocks was increased until the energy variance remained roughly constant, and in general this happened for groupings of 200 steps. Afterwards the resulting variance was tested by carrying out a statistical analysis of a set of independent DMC calculations for the $N_B=8, N_F=8$ cluster.

TABLE I. Ground-state binding energies (in K) of mixed clusters for several combinations of N_B and N_F, obtained in a diffusion Monte Carlo calculation using the Aziz HFD-B (HE) interaction. Figures in italic characters correspond to metastable states.

N_F	$N_B=1$	2	3	4	8
0		0.00162	0.127(3)	0.577(6)	5.125(15)
1		0.0154(16)	0.303(4)	0.931(8)	6.07(2)
2		0.110(5)	0.590(6)	1.404(8)	7.05(2)
3		*0.011(3)*	*0.578(8)*	1.508(10)	7.69(2)
4			*0.605(8)*	1.719(10)	8.42(2)
5			0.757(12)	2.011(12)	9.23(2)
6			0.923(12)	2.357(14)	10.03(3)
7			1.236(14)	2.790(16)	11.03(3)
8		0.225(14)	1.67(3)	3.36(2)	12.03(3)
9		*0.053(10)*	*1.581(16)*	*3.40(2)*	12.33(3)
10			*1.65(2)*	3.55(2)	12.74(3)
11			*1.70(2)*	3.69(2)	13.20(4)
12			1.84(2)	3.87(3)	13.71(5)
13			2.00(2)	4.15(3)	14.20(4)
14		*0.14(3)*	2.17(3)	4.45(3)	14.88(4)
15		0.46(5)	2.60(3)	4.88(3)	15.73(5)
16		0.63(3)	3.05(3)	5.43(3)	16.55(4)
17		1.19(3)	3.62(3)	6.18(4)	17.44(13)
18		1.53(4)	4.34(4)	6.81(4)	18.49(5)
19		2.15(3)	4.96(6)	7.64(4)	19.57(6)
20	0.348(14)	3.26(6)	5.76(5)	8.84(5)	20.75(5)

Table I displays the full set of calculations using the Aziz HFD-B interaction. Empty entries correspond to cases in which the DMC energy jumped to a positive value, representing an unbound system. Entries in italic represent *metastable states* in which the DMC algorithm converged to a negative energy value, but with energy higher than or equal to (within statistical errors) that of the cluster with the same number of bosons and one fermion less. The remaining entries are properly bound and stable systems. There is an obvious remark regarding the metastable states. An exact and unconstrained DMC calculation should converge to a bound

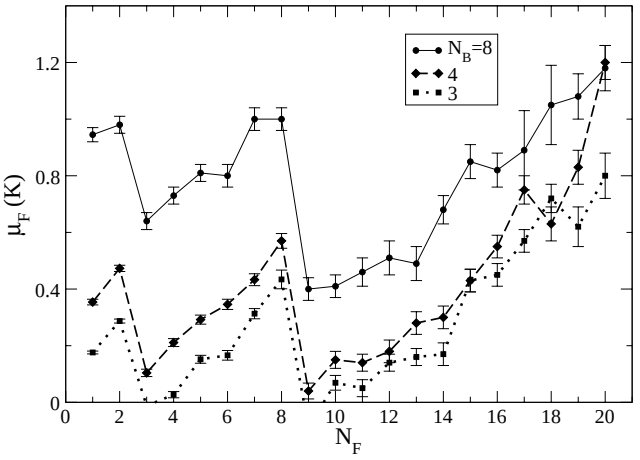

FIG. 1. Fermion separation energies (in K) as a function of N_F for $N_B=4$ and $N_B=8$.

subsystem with one fermion less plus a very far away unbound fermion. Perhaps this also happens in simulations based in an approximate Green function, if the total elapsed time is enormously long. However, the required projection on the importance sampling wave function, which is space confined, may prevent the time evolution to end up at the unbound system.

Clusters $^4He^3He_{20}$ and $^4He_2{}^3He_8$ deserve specific comments. On one hand, given that pure fermion systems with around 30 atoms are known to be bound [7], one expects that the addition of a single boson will lower the required number of fermions to produce a bound state. We have found indeed that $N_F=20$ is the minimum number of fermions bound to a single boson. On the other hand, $^4He_2{}^3He_8$ is an isolated bound cluster. Presumably it will be difficult to produce if the creation of clusters follows the expected mechanism of growing up from the previous bound system by capturing a new fermion. These two clusters, and in general clusters with 2, 8, and 20 fermions, are related to the special stability associated with the closure of $1s$, $1p$, and $2s1d$ shells, respectively, and may be properly considered as *magic* clusters.

Shell-closure effects are magnified in Fig. 1, in which the fermion chemical potentials or separation energies

$$\mu_F = E(N_B, N_F - 1) - E(N_B, N_F) \tag{3}$$

are represented as a function of the number of fermions for the clusters with 3, 4, and 8 bosons. The pattern recalls the analogous representation for the atomic ionization potentials, with a sudden dropping of the chemical potential after $N_F=2$ and 8, and a steady growing within the active shells. When there are very few bosons, this falling-off results in unstable regions. Note that this could also happen after the filling of the $2s1d$ shell, namely, for $N_F=21$, specially if the number of bosons is very small. Thus, $^4He^3He_{20}$ could also be an isolated cluster, such as $^4He_2{}^3He_8$. The fluctuations of the chemical potential within a shell are presumably of statistical nature, because after computing the energy differences the relative errors become quite large.

TABLE II. Same as Table I for the TTY interaction.

N_F	$N_B=2$	3	4	8
0	0.0013	0.1218(14)	0.558(2)	5.037(16)
1	0.0113(4)	0.297(2)	0.911(4)	5.965(8)
2	0.0983(16)	0.574(2)	1.360(4)	6.945(10)
3	*0.033(2)*	*0.553(4)*	1.460(4)	7.523(10)

The present results for the binding energies improve our previous VMC calculations with the self-adjustable configuration-interaction pair correlations [23,24], the changes being important for medium- and large-sized clusters, but not so important for clusters with less than six to eight atoms. Obviously, the relative improvement of the DMC algorithm with respect to the VMC calculations is more important near the stability edges, and this is reflected in some changes of the upper limits of the stability regions.

To ascertain the effect of the atom-atom interaction, we have used the TTY interaction to analyze the interesting $N_F=3$ region. Our DMC results are given in Table II. Even if the obtained binding energies are smaller than those computed with the HFD-B potential, the differences are not large enough to change the stability limits. These results are in very good agreement with previous calculations of Bressanini and co-workers [25,26] for $N_F=2$, which is a further test of our calculations. Our results indicate that $N_B \geqslant 4$ ^{4}He atoms can bind three ^{3}He, whereas in Ref. [26] a minimum number $N_B=9$ was required. As we have mentioned above, the reason of this apparent discrepancy lies in the angular-momentum coupling of the importance sampling wave function. We have considered the HO $1s^2 1p^1$ level ordering, which gives $L=1$ angular momentum and $S=1/2$ spin. Bressanini and Morosi considered instead the state $L=0$, $S=1/2$, as would correspond to the $1s^2 2s^1$ ordering. Our conclusion is that the $L=0$ state of the cluster $^4He_9{}^3He_3$ should be an excited state, whereas its ground state has $L=1$. One may guess that the excitation spectrum of mixed drops can be very rich.

We conclude this paper with a remark pointing towards the hopefully forthcoming experiments. Our results indicate that clusters $^4He_{2,3}{}^3He_3$ are metastable and should not be observed, whereas clusters $^4He_4{}^3He_3$ and $^4He_2{}^3He_8$ are bound, the latter being isolated. The absence or presence of peaks in the mass spectra related to these systems will ascertain the validity of the present analysis and determine the bounds of the $1p$-shell instability region.

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