

## Variational calculations for $^3\text{He}$ impurities on $^4\text{He}$ droplets

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Variational Monte Carlo method is used to calculate ground state properties of  $^4\text{He}$  droplets, containing 70, 112, 168, 240, 330, and 728 particles. The resulting particle and kinetic energy densities are used as an input in the Feynman-Lekner theory for  $^3\text{He}$  impurities. The kinetic energy density of  $^4\text{He}$  atoms and the energy of the  $^3\text{He}$  surface states are compared with the results of previous phenomenological calculations.

### I. INTRODUCTION

Helium droplets have attracted some interest in recent years. A major motivation is the fact that they represent a prototype of finite size quantum systems, behaving as fluid of strongly interacting structureless particles. Several theoretical schemes have been developed for pure helium droplets.<sup>1-6</sup> The comparison between theory and experiments is still elusive, mainly because helium droplets are so weakly bound objects that their experimental characterization is very difficult. An interesting approach, from this point of view, is the use of atomic and molecular impurities as probes.<sup>7,8</sup> Theoretical calculations for droplets with impurities have been recently done with both Monte Carlo<sup>9</sup> and density functional methods.<sup>10,11</sup> Impurities heavier than helium atoms are expected to have small zero point motion, so that they can be treated as classical objects in a quantum fluid.<sup>11</sup> This is certainly not true for  $^3\text{He}$  and hydrogen impurities, for which a full quantum mechanical treatment is needed. The case of  $^3\text{He}$  impurities is particularly interesting from the theoretical viewpoint. Since  $^3\text{He}$  and  $^4\text{He}$  atoms interact through the same potential, the properties of their mixed systems are determined only by quantum effects, i.e., the different statistics and the different zero point motion. One  $^3\text{He}$  atom, being lighter than  $^4\text{He}$ , tends to move in regions of low  $^4\text{He}$  density. This is the origin of the so-called Andreev state of  $^3\text{He}$  on a  $^4\text{He}$  liquid-vapor interface at low temperature.<sup>12-17</sup> The same behavior is observed in  $^4\text{He}$  films on solid substrates,<sup>18</sup> where the layer structure of the  $^4\text{He}$  density produces a rich variety of  $^3\text{He}$  states.<sup>19-22</sup> Predictions on the Andreev states of  $^3\text{He}$  atoms on  $^4\text{He}$  droplets have been already given<sup>10</sup> using a phenomenological density functional, as well as the variational Feynman-Lekner theory. A key ingredient in

the latter approach is the  $^4\text{He}$  kinetic energy density, for which a simple approximate expression was proposed in Ref. 15.

In the present work we calculate ground state properties of pure  $^4\text{He}$  droplets by means of a variational Monte Carlo (VMC) method. We follow the same procedure as in Ref. 2, but with a different parametrization for the variational wave function. A detailed discussion about the results for the ground state of the droplets is given elsewhere.<sup>23</sup> Here we present the first microscopic results for the kinetic energy density, which are shown to be in good agreement with the predictions of Ref. 15. Finally, we use the ground state properties of  $^4\text{He}$  droplets as an input in the Feynman-Lekner theory for  $^3\text{He}$  impurities. We calculate the binding energy of the  $^3\text{He}$  surface states for several  $^4\text{He}$  droplets. The results are extrapolated to estimate the binding energy of  $^3\text{He}$  atoms on a planar  $^4\text{He}$  surface, in good agreement with the experimental value.<sup>14</sup>

The work is organized as follows. Section II contains the short description of the VMC method and the results for the kinetic energy density of  $^4\text{He}$  droplets. The Feynman-Lekner approach is briefly introduced in Sec. III, where the results for the Andreev states are also discussed. Section IV is devoted to conclusions.

### II. PARTICLE AND KINETIC ENERGY DENSITY OF PURE $^4\text{He}$ DROPLETS

First, we want to calculate the ground state energy, density profile, and kinetic energy density for droplets of given number of particle  $N$ . We use the VMC method. In the variational approach a suitable form for the many-body wave function  $\Psi_v(\mathbf{r}_1, \dots, \mathbf{r}_N)$  is chosen, containing

a set of parameters that are varied in order to minimize the energy of the system. The Hamiltonian has the usual form

$$H_0 = \sum_j \frac{-\hbar^2}{2m_4} \nabla_j^2 + \sum_{i<j} v(r_{ij}), \quad (1)$$

where  $m_4$  is the mass of  ${}^4\text{He}$  atoms. The Aziz HFDHE2 potential<sup>24</sup> is used as the pair interatomic potential  $v(r_{ij})$ .

Several forms of the variational wave function of the pure  ${}^4\text{He}$  liquid have been proposed in the literature.<sup>1,2,5,6,23</sup> They are expected to give very similar results in the context of the present work. We use the formalism of Ref. 23, where the wave function is taken of the same form as in Ref. 2,

$$\Psi_v = \prod_{i \leq N} f_1(r_i) \prod_{i < j \leq N} f_2(r_{ij}) \prod_{i < j < k \leq N} f_3(r_{ij}, r_{jk}, r_{ki}), \quad (2)$$

but with an improved version of the correlation functions  $f_1$ ,  $f_2$ , and  $f_3$ . This choice ensures a correct asymptotic behavior of the wave function far outside the droplet, which has an important role for the  ${}^3\text{He}$  and hydrogen impurity states.

Once the optimal wave function is found, it is used to calculate the one-body density and the kinetic energy density, given by

$$\rho(\mathbf{r}_1) = N \int d\mathbf{r}_2 \cdots d\mathbf{r}_N |\Psi_v|^2 \quad (3)$$

and

$$\tau(\mathbf{r}_1) = \frac{N}{\rho(\mathbf{r}_1)} \int d\mathbf{r}_2 \cdots d\mathbf{r}_N \Psi_v^* \left( \frac{-\hbar^2}{2m_4} \nabla_1^2 \right) \Psi_v, \quad (4)$$

respectively.

We consider droplets with 70, 112, 168, 240, 330, and 728 particles. The resulting density profiles and the kinetic energy density are shown in Fig. 1. It is

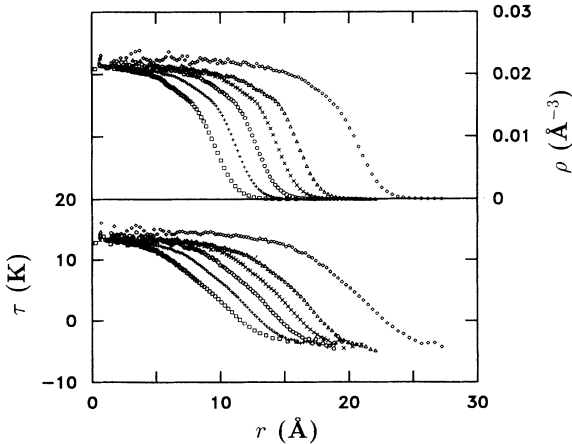


FIG. 1. Particle density and kinetic energy density for droplets of 70 (squares), 112 (plus signs), 168 (circles), 240 (crosses), 330 (triangles), and 728 (diamonds) atoms.

worth noticing that both Monte Carlo<sup>1,2,4-6,23</sup> and density functional<sup>3,11</sup> calculations predict an extrapolated free surface thickness (distance between the points where the density is 90% and 10% of the bulk value) of the order of 6–8 Å, which is consistent with our results.

The kinetic energy density is shown in the lower part of Fig. 1. It decreases smoothly from the inner value, close to the value of the kinetic energy per particle in bulk liquid, to the asymptotic limit  $\mu_4(N)$ , i.e., the chemical potential of the drop containing  $N$   ${}^4\text{He}$  atoms. This limit follows from the behavior of the one-body factor which in our parametrization<sup>23</sup> is

$$f_1(r \rightarrow \infty) \propto \sqrt{\rho(r \rightarrow \infty)} \propto \frac{1}{r} \exp \left[ - \left( \frac{2m_4 |\mu_4|}{\hbar^2} \right)^{\frac{1}{2}} r \right], \quad (5)$$

which dominates the  $r \rightarrow \infty$  limit of the wave function, yielding

$$\tau(r \rightarrow \infty) = \mu_4. \quad (6)$$

In Ref. 15 the approximate expression

$$\tau = \tau_0 \left( \frac{\rho}{\rho_0} \right)^n - \frac{\hbar^2}{2m_4} \frac{\nabla^2 \sqrt{\rho}}{\sqrt{\rho}}, \quad (7)$$

was proposed for  $\tau$ , where  $\tau_0$  and  $\rho_0$  are the ground state kinetic energy per particle and the particle density in bulk  ${}^4\text{He}$  at zero pressure, while  $n$  is a phenomenological parameter. Equation (7) is an interpolation between the expected behavior of  $\tau$  in the two opposite limits  $\rho \rightarrow \rho_0$  and  $\rho \rightarrow 0$ . It has never been checked so far with microscopic calculation. In Ref. 15 the values of  $\tau_0$ , and  $n$  were fixed in a phenomenological way to reproduce known properties of  ${}^3\text{He}$  impurities in bulk liquid  ${}^4\text{He}$ , while the saturation density was taken directly from

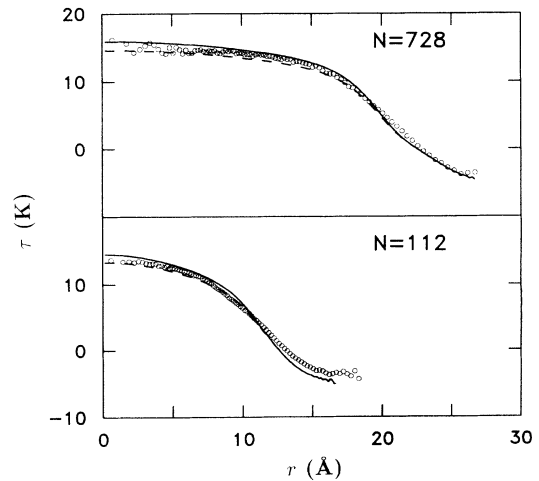


FIG. 2. Kinetic energy density for droplets of 728 and 112 atoms. Circles: VMC; solid lines: Eq. (7) with  $\rho_0 = 0.362\sigma^{-3}$ ,  $\tau_0 = 14.52$  K, and  $n = 1.77$ ; dashed lines: the same equation with  $\rho_0 = 0.365\sigma^{-3}$ ,  $\tau_0 = 13.34$  K, and  $n = 1.76$ .

experiments ( $\tau_0 = 13.34$  K,  $n = 1.76$ ,  $\rho_0 = 0.365\sigma^{-3}$ , where  $\sigma = 2.556$  Å). The same quantities can be calculated microscopically. For instance, the kinetic energy per particle in bulk liquid at several densities was calculated in Ref. 25 using the variational wave function (2). From those data one extracts  $\rho_0 = 0.362\sigma^{-3}$ ,  $\tau_0 = 14.52$  K, and  $n = 1.77$ . In Fig. 2 we compare the VMC results for  $\tau$ , for droplets with 112 and 728 atoms, with the ones of the approximated formula (7). The solid line corresponds to the variational values of  $\tau_0$ ,  $n$ , and  $\rho_0$ , while the dashed line corresponds to the phenomenological parameters of Ref. 15. In both cases we have used Eq. (7) with the VMC density of the corresponding droplets, and the high frequency statistical fluctuations have been filtered out in the calculation of the second derivative. The figure reveals that the approximate formula (7) works very well, especially for large droplets.

### III. LEKNER-FEYNMAN THEORY FOR $^3\text{He}$ IMPURITIES

Once the ground state properties of pure  $^4\text{He}$  droplets are obtained, one can calculate the energy and wave function of one  $^3\text{He}$  impurity. Consider a droplet consisting of  $(N - 1)$   $^4\text{He}$  atoms and one  $^3\text{He}$  impurity atom. Since the interatomic potential is the same for  $^3\text{He}$  and  $^4\text{He}$ , one can write the Hamiltonian in the form

$$H = H_0 + H_I, \quad (8)$$

where  $H_0$  is given in Eq. (1) and

$$H_I = -\frac{\hbar^2}{2m_4} \left( \frac{m_4}{m_3} - 1 \right) \nabla_1^2. \quad (9)$$

A realistic trial wave function would be

$$\Psi = f(\mathbf{r}_1) \prod_{i=2}^N F(\mathbf{r}_1, \mathbf{r}_i) \Psi_0, \quad (10)$$

where  $\Psi_0$  is the ground state of the Hamiltonian  $H_0$ , and  $f$  and  $F$  are variational functions to be determined. In principle, the optimal  $f$  and  $F$  can be found by minimizing the total energy of the system  $\langle \Psi | H | \Psi \rangle$ , which can be done either by solving resulting Euler equations or by direct Monte Carlo simulations.

A significant simplification is obtained by taking  $F(\mathbf{r}_1, \mathbf{r}_i) \equiv 1$ .<sup>26</sup> This corresponds to the assumption that the correlations between  $^3\text{He}$  impurity and  $^4\text{He}$  atoms are the same as those among  $^4\text{He}$  atoms. The Euler equation for  $f$  then becomes

$$-\frac{\hbar^2}{2m_3} \nabla_1 \cdot [\rho(\mathbf{r}_1) \nabla_1 f(\mathbf{r}_1)] + \left( \frac{m_4}{m_3} - 1 \right) f(\mathbf{r}_1) \tau(\mathbf{r}_1) \rho(\mathbf{r}_1) - \lambda f(\mathbf{r}_1) \rho(\mathbf{r}_1) = 0, \quad (11)$$

where  $\rho$  and  $\tau$  are the particle and kinetic energy densities of the pure  $^4\text{He}$  droplet [see Eqs. (3) and (4)]. The same equation can be rewritten in the form of a Schrödinger

equation

$$-\frac{\hbar^2}{2m_3} \nabla^2 \chi(\mathbf{r}) + V_3(\mathbf{r}) \chi(\mathbf{r}) = \epsilon \chi(\mathbf{r}), \quad (12)$$

where

$$\chi(\mathbf{r}) = f(\mathbf{r}) \sqrt{\rho(\mathbf{r})} \quad (13)$$

is the impurity wave function and

$$V_3(\mathbf{r}) = \left( \frac{m_4}{m_3} - 1 \right) \tau(\mathbf{r}) + \frac{\hbar^2}{2m_3} \frac{\nabla^2 \sqrt{\rho(\mathbf{r})}}{\sqrt{\rho(\mathbf{r})}} + \mu_4 \quad (14)$$

is an effective potential seen by the  $^3\text{He}$  atom. The chemical potential  $\mu_4$  of the droplet of the considered size is added for convenience, so that the eigenvalue  $\epsilon$  in Eq. (12) is referred to the state in vacuum. In principle, an additional term should appear in the effective potential  $V_3(\mathbf{r})$  because the exact ground state of pure  $^4\text{He}$  droplet  $\Psi_0$  is approximated by the variational wave function  $\Psi_v$ . This term is expected to be small due to the closeness of these two wave functions, and is neglected in the present treatment.

The assumption  $F(\mathbf{r}_1, \mathbf{r}_i) \equiv 1$  in Eq. (10) corresponds to the Feynman-Lekner theory,<sup>26</sup> already used in the past to predict the properties of the Andreev state on the  $^4\text{He}$  free surface,<sup>13,15</sup> on films,<sup>22</sup> as well as on droplets.<sup>10</sup> Unlike in the previous calculations, where phenomenological  $\rho$  and  $\tau$  were used, here we take them from *ab initio* calculations.

In order to find the energy and the wave function of the impurity on a droplet Eq. (12) is rewritten in spherical coordinates:

$$-\frac{\hbar^2}{2m_3} \frac{d^2}{dr^2} \chi_{nl} + \left( V_3 + \frac{\hbar^2}{2m_3} \frac{l(l+1)}{r^2} \right) \chi_{nl} = \epsilon_{nl} \chi_{nl}. \quad (15)$$

The crucial point is that the effective potential  $V_3$  has a well on the surface of the droplet, so that the lowest eigenstates of the Schrödinger equation are localized on the surface. The potential well originates from a balance between the excess kinetic energy of an  $^3\text{He}$  atom in the bulk with respect to the one of  $^4\text{He}$  atoms, which tends to push the  $^3\text{He}$  atom out, and the He-He interaction, which binds the  $^3\text{He}$  atom to the liquid. A typical situation is shown in Fig. 3 for a droplet of 112 atoms. Results for the energy of the lowest ( $n = 0$ ) impurity states on six droplets are given in Fig. 4 as a function of  $N^{-1/3}$ . We note that a suitable smoothing procedure has been applied to the VMC data in order to avoid spurious effects of statistical fluctuations of  $\rho$  in the calculation of the second derivative. This affects mainly the external tail of  $V_3$  where the density is very small. The error bars in Fig. 4 correspond to the consequent inaccuracy in the results, estimated by choosing different smoothing methods. The extrapolation to the case of the planar surface ( $N \rightarrow \infty$ ) can be done by a linear fit, even if the accuracy of the fit is relatively poor. We obtain  $\epsilon(\infty) \simeq -4.9$  K, rather close to the experimental estimate  $\epsilon(\infty) = (-5.02 \pm 0.03)$  K quoted in Ref. 14.

In Fig. 4 the results of the VMC calculation are com-

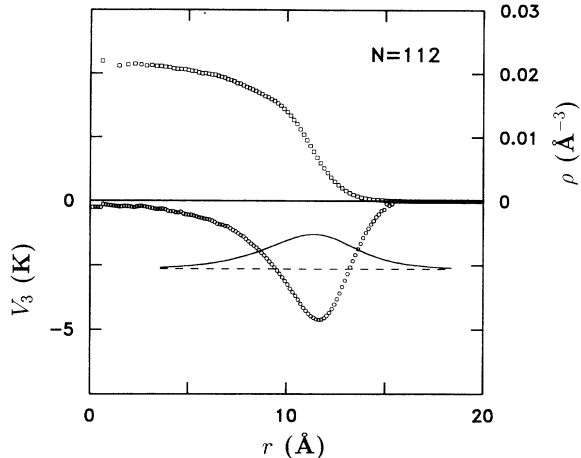


FIG. 3. Lowest eigenstate of one  $^3\text{He}$  impurity on a droplet of 112 atoms. Solid line:  $^3\text{He}$  wave function; squares:  $^4\text{He}$  density; circles: effective potential  $V_3$ .

pared with predictions of Ref. 10 (solid line) obtained using the Feynman-Lekner theory with approximation (7) for  $\tau$  and density profiles of Ref. 3. The significant difference between the two predictions is mainly due to the different density profiles. The curvature of the surface profiles in the outer region is underestimated in the density functional calculations of Ref. 3 with respect to the VMC results (see Fig. 5). This makes the potential well for the impurity wider, and the binding energy lower. The density profiles calculated with a more recent density functional<sup>27,11</sup> are sharper than the ones of Ref. 3 and the corresponding predictions for the  $^3\text{He}$  binding energy are closer to the VMC results (dashed lines in Figs. 4 and 5). Part of the remaining discrepancy is due to the use of the approximated expression (7) for  $\tau$ ; we have

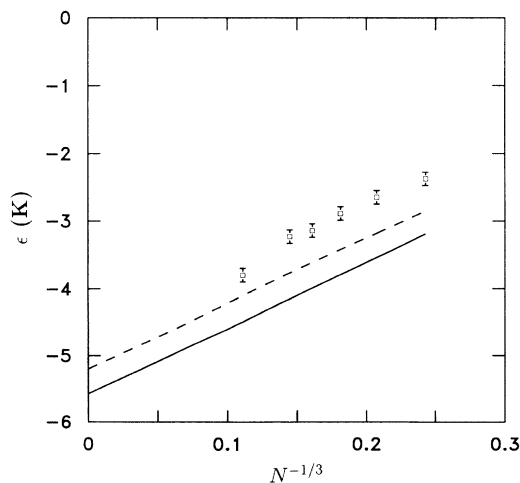


FIG. 4. Binding energy of the lowest impurity state as a function of  $N^{-1/3}$ . Points with error bars: Feynman-Lekner theory with  $\tau$  and  $\rho$  from VMC calculations; solid line: Feynman-Lekner results of Ref. 10; dashed line: same theory but with more recent density functional calculations for the density profiles, as in Ref. 11.

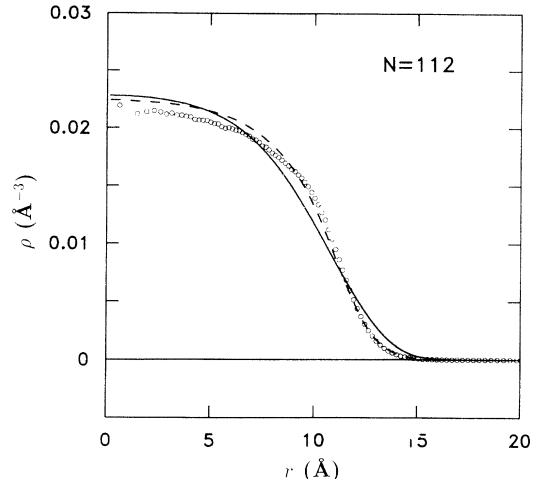


FIG. 5. Surface profile for a droplet of 112 atoms. Points: VMC results; solid line: density functional results of Ref. 3; dashed line: density functional results of Ref. 11.

checked that the corresponding effect on the  $^3\text{He}$  binding energy is small (about 0.1 K).

By solving the Schrödinger equation (12) one finds, above the lowest eigenstate, a spectrum of states with different principal quantum number  $n$  and angular momentum  $l$ . The general features of the spectrum are the same as in Ref. 10. In the limit of an infinite droplet the states with different  $l$  coincide with those of a two-dimensional Fermi gas.

To conclude, we stress again the idea of the Feynman-Lekner approach and its limits. The idea is that the two-body correlations between the  $^3\text{He}$  impurity and  $^4\text{He}$  atoms are taken to be the same as the ones between the  $^4\text{He}$  atoms. The  $^3\text{He}$  wave function is then expressed by means of the factor  $f(\mathbf{r})$  in the many-body wave function (10). The form of  $f$  is derived variationally, by solving a Schrödinger-like equation. The lowest eigenvalues are localized on the surface of the droplets, as an effect of the different mass of  $^3\text{He}$  and  $^4\text{He}$ . To improve the Feynman-Lekner theory one should account for the fact that  $^3\text{He}$ - $^4\text{He}$  correlations differ from the  $^4\text{He}$ - $^4\text{He}$  ones; i.e., one should take  $F(\mathbf{r}_1, \mathbf{r}_i) \neq 1$ . One possibility is to choose  $F = (1 + \phi)$ , where  $\phi$  is small. Keeping the terms in the expansion of the total energy up to the second order in  $\phi$  is equivalent to taking into account the energy of an elastic deformation of the residual droplet due to the presence of the impurity. Such corrections have already been studied in Refs. 28 and 20 in the case of liquid  $^4\text{He}$  films. The problem of  $^3\text{He}$ - $^4\text{He}$  correlations, in the context of variational calculations, has been also discussed in Refs. 29 and 30 for bulk  $^3\text{He}$ - $^4\text{He}$  mixtures. Work in this direction is in progress.

#### IV. CONCLUSIONS

We present the variational Monte Carlo (VMC) calculation for  $^4\text{He}$  droplets with and without  $^3\text{He}$  impurities.

We have used the variational wave function of Ref. 2 modified as in Ref. 23. The results for the density and energy of six droplets, from  $N = 70$  to  $N = 728$ , are close to the predictions of previous calculations. We have calculated the kinetic energy density, for which only approximate estimates have been given so far. Our results show that the analytic formula given in Ref. 15 for the kinetic energy density [Eq. (7)] works very well, and can be safely used in calculations involving the  $^4\text{He}$  surface.

In the second part of the work we applied the Feynman-Lekner approach to study  $^3\text{He}$  impurity states on  $^4\text{He}$

droplets. The impurity wave function turns out to be localized on the surface of the droplets, as expected. The lowest value of the binding energy is almost linearly dependent on  $N^{-1/3}$  and the value extrapolated to  $N \rightarrow \infty$  is close to the experimental binding energy of the Andreev state on a planar surface. We have also compared our results with the predictions for the  $^3\text{He}$  binding energies obtained using phenomenological calculations of the density profiles and the kinetic energy density. The general trend of these earlier results is similar to the one of VMC calculations.

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