Diffusion Monte Carlo study of two-dimensional liquid $^4$He

S. Giorgini, J. Boronat and J. Casulleras
Departament de Física i Enginyeria Nuclear, Campus Nord B4–B5,
Universitat Politècnica de Catalunya, E–08028 Barcelona, Spain

Abstract

The ground-state properties of two-dimensional liquid $^4$He at zero temperature are studied by means of a quadratic diffusion Monte Carlo method. As interatomic potential we use a revised version of the HFDHE2 Aziz potential which is expected to give a better description of the interaction between helium atoms. The equation of state is determined with great accuracy over a wide range of densities in the liquid phase from the spinodal point up to the freezing density. The spinodal decomposition density is estimated and other properties of the liquid, such as radial distribution function, static form factor, momentum distribution and density dependence of the condensate fraction are all presented.

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I. INTRODUCTION

In recent years a great deal of interest has been devoted in the literature to the study of quantum boson liquids in restricted geometries.\textsuperscript{1−15} From a theoretical point of view thin films of liquid \textsuperscript{4}He adsorbed on different solid substrates have been studied using variational techniques, based on the hypernetted-chain/Euler-Lagrange (HNC/EL) theory,\textsuperscript{1−3} Density Functional theories (DFT),\textsuperscript{4−6} and also Monte Carlo techniques.\textsuperscript{12,13} According to these investigations, liquid \textsuperscript{4}He films display different behaviours depending on the strength of the substrate potential.

In the case of weak-binding and long-range potentials, such as some alkali metal substrates, the helium film does not exhibit at all two-dimensional characteristics. By adding \textsuperscript{4}He atoms to the substrate surface, three-dimensional clusters form and helium uniformly covers the surface only for coverages many atoms thick (prewetting transition). For some substrates a stable coverage is found only for infinitely thick films (wetting transition).

More interesting, from the point of view of the role of dimensionality, is the case of substrates with deep and narrow potential wells for which the degree of freedom perpendicular to the surface is practically “frozen out” and low-coverage films are stabilized at the surface forming two-dimensional (2D) systems. To this category belong graphite, solid H\textsubscript{2} on glass and some alkali metal substrates like Li and Mg. On these substrates the growth of the first liquid layers is predicted to proceed via layering transitions\textsuperscript{1,3,4} by increasing the surface coverage single atomic monolayers develop and become stable one on the top of the other. This prediction has been confirmed experimentally for helium on graphite by heat capacity\textsuperscript{8} and third sound measurements.\textsuperscript{9}

Variational calculations of ground-state and dynamic properties of single \textsuperscript{4}He monolayers at zero temperature reveal a striking 2D behaviour over a wide range of coverages.\textsuperscript{1,2} Such calculations seem to indicate that single monolayers of liquid helium adsorbed on strong-binding substrates represent physical realizations of 2D homogeneous quantum liquids.

A condition which can be relevant for theories aiming to describe \textsuperscript{4}He films is that
they possess the correct limiting behaviour of homogeneous three-dimensional and two-
dimensional systems. Homogeneous three dimensional (3D) $^4$He is a well studied system
both experimentally and theoretically. In the case of homogeneous 2D $^4$He no experimental
results are available and “exact” results coming from \textit{ab initio} calculations can be extremely
useful.

From the point of view of Monte Carlo simulations, $^4$He in confined geometries has
already been the object of several studies.$^{10-15}$ Variational Monte Carlo techniques have
been applied to the study of both inhomogeneous films on substrates$^{12}$ and more extensively
to homogeneous 2D $^4$He, where the simulation is easier.$^{10,15}$ However, these methods rely
heavily on the choice of the variational wavefunction used in the simulation.

From the side of more exact Monte Carlo methods, thin $^4$He films on molecular hydro-
gen surfaces at low temperature have been recently studied by Wagner and Ceperley$^{13}$ using
Path Integral Monte Carlo (PIMC) techniques. Strictly two-dimensional $^4$He has also been
the object of Monte Carlo calculations.$^{11,14}$ At $T = 0$ Whitlock \textit{et al.}$^{11}$ have calculated, using
Green’s Function Monte Carlo (GFMC), the equation of state and other ground-state prop-
erties of liquid and solid $^4$He in two dimensions, giving an estimate of the freezing and melting
densities. At finite temperature, PIMC techniques have been employed by Ceperley \textit{et al.}$^{14}$ to investigate the superfluid transition, which in 2D belongs to the Kosterlitz-Thouless
universality class.

In the present work we present a Diffusion Monte Carlo (DMC) calculation of homoge-
nous liquid $^4$He in two dimensions at zero temperature. Our main purpose is to provide an
exhaustive calculation of the equation of state and other relevant properties of the ground
state such as distribution function, structure form factor, momentum distribution and con-
densate fraction for a wide range of densities within the liquid phase. In particular, we have
performed a detailed analysis of the equation of state in the region of negative pressures
in order to give an accurate estimation of the spinodal density of the 2D $^4$He liquid. The
spinodal density is defined as the density below which the system becomes macroscopically
unstable against density fluctuations. Its determination is relevant for the physics of $^4$He
monolayers on strong-binding substrates because the spinodal density coincides with the coverage at which the uniform monolayer breaks down into 2D clusters in equilibrium with the vacuum, and it is thus the density at which the first layering transition occurs.\textsuperscript{1,3}

In our DMC simulation we have used as interatomic potential a revised version of the HFDHE2 Aziz potential\textsuperscript{16} which was proposed by Aziz \textit{et al.}\textsuperscript{17} in 1987 and is known as HFD-B(HE) potential. This renewed interatomic potential was recently used by two of us\textsuperscript{18} (J.B. and J.C.) to study 3D bulk helium at zero temperature and it has been proven to give results for the density dependence of the pressure and the system compressibility which are closer to experimental data than the older Aziz potential. We are thus inclined to think that it gives also better results for two-dimensional $^4$He.

The structure of the paper is as follows: in Sec. II we outline briefly the quadratic Diffusion Monte Carlo method used to solve the Schrödinger equation. In Sec. III we present results on both the equation of state in the liquid phase and on the distribution and structure functions. In Sec. IV are collected the results for the density dependence of the condensate fraction, which differ substantially from the previous analysis by Whitlock and coworkers,\textsuperscript{11} and of the momentum distribution for different values of the density. A brief discussion and conclusions are included in Sec. V.

II. COMPUTATIONAL METHOD

The aim of Diffusion Monte Carlo algorithms is to solve the time dependent Schrödinger equation of a system of $N$ particles in imaginary time

$$-\frac{\partial \Psi(R, t)}{\partial t} = (\hat{H} - E)\Psi(R, t), \quad (1)$$

where $R = (r_1, ..., r_N)$ is the configuration vector for the positions of the $N$ particles and $t$ is measured in units of $\hbar$. In Eq. (1) the Hamiltonian $\hat{H}$ has the usual form

$$\hat{H} = -\frac{\hbar^2}{2m} \nabla_{\mathbf{R}}^2 + V(\mathbf{R}), \quad (2)$$
and $E$ is a parameter representing an energy shift. Provided the initial wavefunction
$\Psi(R, t = 0)$ has a nonzero overlap with the ground state $\Phi_0(R)$, the solution of Eq. (1) gives exactly the ground state wavefunction in the asymptotic limit of large times:
$\Psi(R, t \to \infty) = \Phi_0(R)$.

When numerically integrating Eq. (1) importance sampling techniques are employed to guide quickly the solution towards the ground state. To this aim the Schrödinger equation (1) is rewritten for the function

$$f(R, t) = \psi_T(R)\Psi(R, t),$$

where $\psi_T(R)$ is a time independent trial function. One gets

$$-\frac{\partial f(R, t)}{\partial t} = -D \nabla^2_R f(R, t) + D \nabla_R (F(R)f(R, t)) + (E_L(R) - E) f(R, t) \equiv \hat{A} f(R, t).$$

Eq. (4) has the form of a classical diffusion equation for the distribution function $f(R, t)$. The first term describes the diffusive process with diffusion constant $D = \hbar^2/2m$. The second term contains the drift force

$$F(R) = 2\psi_T(R)^{-1}\nabla_R \psi_T(R),$$

which drives the system towards the region in configuration space where the trial function $\psi_T(R)$ is relevant. The third term in Eq. (4) represents a branching term which depends on the local energy $E_L(R) = \psi_T(R)^{-1}\hat{H}\psi_T(R)$.

By introducing the time dependent Green’s function

$$G(R', R, \Delta t) = \langle R' | e^{-\hat{A}\Delta t} | R \rangle$$

the solution of Eq. (4) can be written formally as

$$f(R', t + \Delta t) = \int dR \ G(R', R, \Delta t) f(R, t).$$

If the Green’s function $G(R', R, \Delta t)$ is known for infinitesimal time steps $\Delta t$, the asymptotic solution for large times $f(R, t \to \infty)$ can be obtained by solving iteratively Eq. (4).
In the most usual implementations of DMC algorithms to solve the many body Schrödinger equation, \( G(R', R, \Delta t) \) is approximated up to order \( \Delta t \) for small time steps.\(^{19}\) In this case, the obtained energy of the ground state depends linearly on the time step \( \Delta t \) and several calculations for different values of \( \Delta t \) are needed in order to extrapolate the correct result in the limit \( \Delta t \to 0 \).

Recently a quadratic DMC algorithm\(^{20}\) has been proven to work efficiently in the description of \(^4\)He droplets\(^{21}\) and bulk 3D liquid \(^4\)He.\(^{18}\) This method relies on an expansion of the Green’s function \( G(R', R, \Delta t) \) up to order \( (\Delta t)^2 \), which generates a quadratic dependence of the energy eigenvalue on \( \Delta t \), permitting thus to avoid the extrapolation to the limit \( \Delta t \to 0 \) by working with a single time step. More details concerning this procedure can be found in Ref. 18.

In the present work we have used in all the calculations \( \Delta t = 2 \times 10^{-3} \tau \), where \( \tau \) is the appropriate time unit: \( \tau = \frac{m \sigma^2}{2 \hbar^2} \) (\( \sigma = 2.556 \) Å). Time step analysis have been performed at the density \( \rho = 0.275 \sigma^{-2} \), close to the liquid equilibrium density, and at \( \rho = 0.400 \sigma^{-2} \), near the freezing density, and no changes have been observed in the results for the energy eigenvalue by reducing the time step \( \Delta t \).

The mixed estimator \( \langle \psi_T | \hat{O} | \Phi_0 \rangle \) of a generic operator \( \hat{O} \), is the direct output of DMC algorithms. If the operator \( \hat{O} \) commutes with the Hamiltonian of the system, the mixed estimator coincides with the pure expectation value on the ground state \( \langle \Phi_0 | \hat{O} | \Phi_0 \rangle \). If, on the contrary, the operator \( \hat{O} \) does not correspond to a conserved quantity, its expectation value on the ground state can be obtained by means of a linear extrapolation\(^{22}\)

\[
\langle \Phi_0 | \hat{O} | \Phi_0 \rangle = 2 \langle \psi_T | \hat{O} | \Phi_0 \rangle - \langle \psi_T | \hat{O} | \psi_T \rangle .
\]  

(8)

The above method to get pure estimators is the most widely used in DMC simulations which must thus be supplemented by a variational Monte Carlo (VMC) calculation to determine the variational expectation value \( \langle \psi_T | \hat{O} | \psi_T \rangle \). The linear extrapolation (8), which is obtained by writing the ground state wavefunction \( \Phi_0 \) as \( \Phi_0(R) = \psi_T(R) + \delta \psi(R) \), is correct to linear order in the functional variation \( \delta \psi \).
To go beyond this approximation, removing the dependence on the trial wavefunction, several algorithms have been proposed to calculate pure estimators.\textsuperscript{23,24} On this line an algorithm based on “forward walking" has been recently presented by two of us\textsuperscript{25} (J. C. and J. B.) which is easy to incorporate in the original Monte Carlo algorithm and allows the calculation of pure expectation values of coordinate operators $\hat{O}(\mathbf{R})$ with satisfactory stability and reliability. The method applied to bulk 3D liquid helium\textsuperscript{25} gives values for the particle-particle distribution function and static structure factor in very good agreement with experimental results and without any significant dependence on the function used as importance sampling. The results given in the present work for the potential energy, distribution and structure functions are calculated by using this method.

Another important parameter in the calculation is the population of walkers $n_w$, which represents the number of points $\mathbf{R}_i$ in configuration space at which the distribution function $f(\mathbf{R}, t)$ is sampled. In our calculations we have used a mean walker population of $n_w = 450$. At the densities $\rho = 0.275\sigma^{-2}$ and $\rho = 0.400\sigma^{-2}$ the walker population was increased to $n_w = 900$ and no appreciable change in the results was observed.

Our simulation box contains 64 particles. At the 2D saturation density $\rho_0 \simeq 0.284\sigma^{-2}$ this corresponds to a simulation box length of $\sim 38$ Å, roughly 14 times larger than the mean interparticle distance at this density. Such box size is large enough to neglect finite volume effects. In fact, in 3D bulk liquid $^4$He finite size effects have been proven to be negligible already for a box length of 8 times larger than the mean interparticle distance.\textsuperscript{18}

Finally, as importance sampling we have used a simple McMillan\textsuperscript{26} two-body trial function

\begin{equation}
\psi_T(\mathbf{R}) = \prod_{i<j} \exp \left( -\frac{1}{2} \left( \frac{b}{r_{ij}} \right)^5 \right) .
\end{equation}

We have taken for all the densities $b = 1.205\sigma$. This value, which minimizes the energy at the equilibrium density, coincides with the one found by Whitlock \textit{et al.}\textsuperscript{11} in their VMC calculation. For the highest density calculated, $\rho = 0.420\sigma^{-2}$, we have also used as importance sampling the two-body function proposed by Reatto\textsuperscript{27}.
\[ \psi_T(R) = \prod_{i<j} \exp \left( -\frac{1}{2} \left( \frac{b}{r_{ij}} \right)^5 - \frac{L}{2} \exp \left( -\left( \frac{r_{ij} - \lambda}{\Lambda} \right)^2 \right) \right), \quad (10) \]

with \( L = 0.2, \lambda = 2.0\sigma, \Lambda = 0.6\sigma \) and \( b = 1.225\sigma \). Although this trial function gives at the density \( \rho = 0.420\sigma^{-2} \) a VMC energy lower than the McMillan function (9), no appreciable change in the results of the DMC simulation is found.

### III. RESULTS

In this section, we present our numerical results for the energy and structure properties of the ground state. First we analyze the equation of state in the liquid phase, comparing our results with the ones presented in Ref. 11. Ground-state properties such as the radial distribution function and the static structure factor are also discussed in the first subsection. The second subsection contains our results concerning the density dependence of the condensate fraction and the momentum distribution.

#### A. Equation of state and structure properties

In Ref. 11, the equation of state for both the liquid and the solid phase of 2D homogeneous \(^4\)He has been investigated by means of VMC and GFMC techniques. The interatomic potential employed in this study is the two-body HFDHE2 Aziz potential.\(^{16}\) The estimated value for the liquid freezing density from the GFMC calculation is \( \rho_l = 0.443\sigma^{-2} \). For the sake of consistency we have calculated the energy per particle using the Aziz potential at two densities in the liquid phase: at \( \rho = 0.275\sigma^{-2} \), close to the equilibrium density, and at \( \rho = 0.400\sigma^{-2} \), near freezing. The values for the energy obtained from our DMC calculation are in good agreement with the GFMC results reported in Ref. 11 for the same two densities.

Once the equivalence between our DMC algorithm and the GFMC algorithm employed in Ref. 11 has been tested by using the same interatomic potential, we have proceeded to the study of the equation of state in the liquid phase with the HFD-B(HE) potential proposed by Aziz et al.\(^{17}\) (henceforth referred to as Aziz II). In Table I we present the results of our
DMC calculations for the total, potential and kinetic energy per particle for some of the densities calculated. As discussed in Sec. II the values of the potential energy per particle have been obtained by employing the algorithm for pure estimators.\textsuperscript{25}

The Aziz II potential is slightly more attractive than the Aziz potential and the binding energies at the different densities are therefore somewhat larger with respect to the ones obtained in Ref. 11. For example, at $\rho = 0.275\sigma^{-2}$ near to equilibrium the energies per particle are $E/N = -0.8519 \pm 0.0044$ K and $E/N = -0.8950 \pm 0.0019$ K for the Aziz and Aziz II potentials respectively. In Fig. 1 we show our DMC results for the equation of state in the liquid phase together with the GFMC results of Ref. 11.

The equation of state of 2D liquid $^4$He is usually fitted by using a polynomial cubic function of the form

$$e = e_0 + B \left( \frac{\rho - \rho_0}{\rho_0} \right)^2 + C \left( \frac{\rho - \rho_0}{\rho_0} \right)^3,$$  \hspace{1cm} (11)

where $e = E/N$ and $\rho_0$ is the equilibrium density. In Table II we report the values of the parameters which best fit our results and we compare them with the values reported in Ref. 11. The values for the equilibrium density are very close, whereas appreciably different are the values for the $B$ and $C$ parameters. These differences affect the predictions for the density dependence of the surface pressure and compressibility, as well as the estimation of the spinodal density. The two fits are shown in Fig. 1 together with the Monte Carlo data. The cubic polynomial fit (11) fits our data rather well and no significant improvement in the $\chi^2$ quality of the fit is found by increasing the order of the polynomial function used.

Once the equation of state function $e(\rho)$ is known, one can calculate straightforwardly the surface pressure, defined as $P(\rho) = \rho^2(\partial e/\partial \rho)$, and the isothermal compressibility

$$\kappa(\rho) = \frac{1}{\rho} \left( \frac{\partial \rho}{\partial P} \right).$$  \hspace{1cm} (12)

From the inverse compressibility $1/\kappa$ one obtains the velocity of sound

$$c(\rho) = \left( \frac{1}{m\rho\kappa} \right)^{1/2}. \hspace{1cm} (13)$$
In Fig. 2 we show the comparison between the surface pressure obtained from our equation of state and the result of Ref. 11. An appreciable difference is found in the regimes of low and high densities. A larger difference is found by comparing the predictions of the two fits for the velocity of sound, shown in Fig. 3. The velocity of sound at the 2D equilibrium density is \( c(\rho_0) = (92.8 \pm 0.6) \text{ m/sec} \), which is nearly 3 times smaller than the velocity of sound at the saturation density of 3D bulk liquid \(^4\text{He} \), \( c^{3D}(\rho_0) = 238.3 \text{ m/sec} \).28

At \( T = 0 \), and in the limit of a 2D film, the third sound velocity \( c_3 \) coincides with the velocity of sound \( c^{2D} \) of the purely 2D liquid.4 Then, for low coverages \( c^{2D} \) must reproduce quite accurately the \( c_3 \) in monolayers adsorbed on strongly binding substrates. A direct comparison between \( c^{2D} \) and experimental \( c_3 \) for low-coverage films is however difficult by the present uncertainties in the determination of the liquid coverages.29

The density dependence of the velocity of sound is linear over a wide range of densities. Only when approaching the spinodal density, where the system becomes unstable against density fluctuations, the velocity of sound drops suddenly to zero and the compressibility diverges. The estimation of the spinodal density from our fit gives: \( \rho_{sp} = (0.228 \pm 0.002)\sigma^{-2} \), which is significantly smaller than the value \( \rho_{sp} = 0.247\sigma^{-2} \) obtained from the data of Ref. 11. The spinodal density of 2D homogeneous liquid \(^4\text{He} \) has been estimated also by Clements et al.1 from the fit of their HNC/EL variational results for the equation of state: they find \( \rho_{sp} = 0.202\sigma^{-2} \).

At the spinodal density \( \rho_{sp} = 0.228\sigma^{-2} \) the mean interparticle distance is \( r_{sp} = (1/\pi\rho_{sp})^{1/2} = 3.02\text{Å} \). For 3D bulk liquid \(^4\text{He} \) an accurate estimation of the spinodal density has been given in Ref. 30 with the result \( \rho_{sp}^{3D} = (0.264 \pm 0.002)\sigma^{-3} \), corresponding to a mean interparticle distance of \( r_{sp}^{3D} = (3/4\pi\rho_{sp}^{3D})^{1/3} = 2.47\text{Å} \). It is interesting to notice that at the 2D freezing density \( \rho_l = 0.443\sigma^{-2} \), estimated in Ref. 11, the mean interparticle distance is only \( r_l = 2.17\text{Å} \), whereas at the 2D equilibrium density the interparticle spacing is \( r_{eq} = 2.71\text{Å} \). The corresponding values of the average interatomic distance in the 3D case are respectively: \( r_{l}^{3D} = 2.10\text{Å} \) and \( r_{eq}^{3D} = 2.22\text{Å} \). The mean interparticle distances of the 2D and 3D liquid \(^4\text{He} \) barely overlap; at freezing the particles are only 3% further apart in 2D than
in 3D, at the equilibrium density the difference increases to 22% and the same difference persists down to the spinodal density.

Important information on the structure of the ground state is obtained from the two-body radial distribution function

\[ g(r_{12}) = \frac{N(N-1)}{\rho^2} \frac{\int \Phi_0(r_1, \ldots, r_N)|^2 dr_3 \ldots dr_N}{\int \Phi_0(r_1, \ldots, r_N)|^2 dr_1 \ldots dr_N} \]  \hspace{1cm} (14)

and from its Fourier transform, the static structure factor

\[ S(k) = 1 + \rho \int dr e^{i k \cdot r} (g(r) - 1) = \frac{1}{N} \frac{\langle \Phi_0 | \rho_q \rho_{-q} | \Phi_0 \rangle}{\langle \Phi_0 | \Phi_0 \rangle}, \]  \hspace{1cm} (15)

with

\[ \rho_q = \sum_{i=1}^{N} e^{i q \cdot r_i}. \]  \hspace{1cm} (16)

Both these quantities can be calculated using the method for pure estimators described in Ref. 25. In Fig. 4 we show the results obtained for the radial distribution function at three different densities. As the density increases more peaks at large interparticle distances appear. At the highest density shown \( \rho = 0.420 \sigma^{-2} \), which is just before freezing, four peaks are clearly visible in the distribution function: a clear indication that the system is close to solidification. In Table III, we report the position \( r_m \) and the height \( g(r_m) \) of the first peak in the radial distribution function for some of the densities calculated. The height of the first peak in \( g(r) \) increases with the density and it shifts towards smaller interparticle distances. It is interesting to compare the height of the first peak in the radial distribution function for the 2D and 3D systems: in 2D at the equilibrium density \( g(r_m) \approx 1.25 \) whereas the corresponding value in 3D is 1.38. This is a clear indication that the 2D system is more dilute and possesses less correlations at equilibrium than its 3D counterpart. Close to the freezing density the heights of the first peak in \( g(r) \) of the 2D and 3D systems become comparable. In fact, as previously discussed, the mean interparticle distance of the two systems become similar when freezing is approached.

In Fig. 5 we show the static structure factor for three values of the density. As the density increases the peak in \( S(k) \) increases and the values at the lowest momenta accessible
in our calculation increase. Due to phonon excitations the form factor $S(k)$ is expected to go to zero in the long wavelength limit as $S(k) \sim k/2mc$. As the density decreases and the spinodal density is approached the velocity of sound $c$ drops to zero and consequently the slope in $S(k)$ diverges. This behaviour, which has been observed in the variational calculations of Ref. 1, agrees qualitatively with our ab initio calculations.

B. Condensate fraction and momentum distribution

Another quantity of great interest is the one-body density matrix $\rho(r', r)$, which is related to the change in the ground-state wavefunction when a particle is removed from position $r$ and replaced at position $r'$. For a homogeneous system $\rho(r)$ is defined as

$$\rho(r) = \langle \Phi_0 | \hat{\psi}^\dagger(r) \hat{\psi}(0) | \Phi_0 \rangle = N \frac{\int \Phi_0(r_1 + r, ..., r_N) \Phi_0(r_1, ..., r_N) dr_2 ... dr_N}{\int |\Phi_0(r_1, ..., r_N)|^2 dr_1 ... dr_N}, \quad (17)$$

where $\hat{\psi}(0)$ and $\hat{\psi}^\dagger(r)$ are, respectively, the field operators which destroy a particle from position $r = 0$ and create one at position $r$.

In the DMC algorithm the mixed matrix element $\langle \psi_T | \hat{\psi}^\dagger(r) \hat{\psi}(0) | \Phi_0 \rangle$, involving the trial wavefunction $\psi_T$, can be calculated by averaging over the asymptotic distribution function $f(R, t \to \infty)$ the relative change in the trial wavefunction when a particle is displaced from position $r_i$ to $r_i + r$

$$\langle \psi_T | \hat{\psi}^\dagger(r) \hat{\psi}(0) | \Phi_0 \rangle = \frac{\int f(R, t \to \infty) (\psi_T(r_1, ..., r_i + r, ..., r_N)/\psi_T(r_1, ..., r_N)) dr_1 ... dr_N}{\int f(R, t \to \infty) dr_1 ... dr_N}. \quad (18)$$

The calculation of the ground-state expectation value $\langle \Phi_0 | \hat{\psi}^\dagger(r) \hat{\psi}(0) | \Phi_0 \rangle$ can not be obtained straightforwardly from our method for pure estimators because it involves the knowledge of $\Phi_0(R')\Phi_0(R)$, where $R'$ is the configuration vector with one particle displaced by $r$, instead of $\Phi_0^2(R)$. To calculate the one-body density matrix $\rho(r)$ we employ thus the extrapolation technique described in Sec. II. For the highest density calculated, $\rho = 0.420\sigma^{-2}$, we have used as importance sampling both the McMillan wavefunction (9) and the one proposed by Reatto (10). The extrapolation technique gives, within statistical errors, the same result for $\rho(r)$ in the two cases.
The asymptotic limit of $\rho(r)$ gives the fraction of particles $n_0$ condensed into the zero-momentum state

$$n_0 = \lim_{r \to \infty} \rho(r).$$

(19)

In Fig. 6 we show the results for the condensate fraction obtained at different densities ranging from the spinodal point up to the freezing density. We have fitted our data with the quadratic polynomial

$$n_0(\rho) = n_0(\rho_0) + a \left( \frac{\rho - \rho_0}{\rho_0} \right) + b \left( \frac{\rho - \rho_0}{\rho_0} \right)^2,$$

(20)

where $\rho_0$ is the equilibrium density $\rho_0 = 0.284\sigma^{-2}$. The values of the parameters giving the best fit are the following

$$n_0(\rho_0) = 0.233 \pm 0.001$$

$$a = -0.583 \pm 0.006$$

(21)

$$b = 0.44 \pm 0.02.$$

The value for the condensate fraction at equilibrium density $n_0(\rho_0)$ is consistent with the estimation reported in Ref. 14 $n_0(\rho_0) \simeq 0.22$, obtained by extrapolating to zero temperature the PIMC results for the algebraic decay of the one-body density matrix. Our results for the condensate fraction are somewhat smaller than the ones reported in Ref. 11; for example at low density, $\rho = 0.275\sigma^{-2}$, we find $n_0 = 0.251 \pm 0.005$ whereas the GFMC calculation of Ref. 11 gives $n_0^{GFMC} = 0.36 \pm 0.05$. This discrepancy between DMC and GFMC results persists over the whole density range and only at very high density, $\rho = 0.400\sigma^{-2}$, $n_0^{DMC}$ and $n_0^{GFMC}$ become consistent. The reason for this is unclear, certainly it is not due to the revised version of the Aziz potential used in our DMC simulation. We have repeated our DMC calculation of the condensate fraction using the Aziz potential for the two densities $\rho = 0.275\sigma^{-2}$ and $\rho = 0.400\sigma^{-2}$ and no difference was found with the results obtained with Aziz II. It is interesting to notice that near the freezing density, where the mean interparticle distance in the 2D and 3D systems are comparable, also the condensate fraction is nearly the same ($n_0 \simeq 4\%$).
By Fourier transforming the one-body density matrix $\rho(r)$, one gets the momentum distribution of the system

$$n(k) = (2\pi)^2 \rho n_0 \delta(k) + \rho \int d\mathbf{r} e^{i\mathbf{k} \cdot \mathbf{r}} (\rho(r) - \rho(\infty)).$$

In Fig. 7 we show the momentum distribution plotted as $kn(k)$ for three different densities. As the density increases, more and more particles leave the condensate state and the maximum in $kn(k)$ shifts towards higher momenta. The shoulder present at high momenta is interpreted in 3D bulk $^4$He as coming from the zero-point motion of the rotons. A similar interpretation could be valid also in the 2D case, in this case though the shoulder appears more pronounced than in 3D, particularly at low densities, and shifted towards smaller $k$’s.

**IV. CONCLUSIONS**

The properties of homogeneous 2D liquid $^4$He at zero temperature have been investigated by means of a quadratic diffusion Monte Carlo method. As interatomic potential we have used a revised version of the Aziz potential which is expected to be more accurate in describing the interaction between helium atoms. The energy per particle has been calculated for a wide range of densities from the spinodal point up to the freezing density, providing an accurate determination of the equation of state function. The spinodal density of the 2D system is estimated. The radial distribution function and the static structure factor are calculated for various densities employing a recently devised method for estimating pure expectation values. The fraction of particles in the condensate state has been calculated for various densities and the density dependence of the condensate fraction has been also estimated.

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REFERENCES

1 B. E. Clements, J. L. Epstein, E. Krotscheck and M. Saarela, Phys. Rev B 48, 7450 (1993).

2 B. E. Clements, H. Forbert, E. Krotscheck, H. J. Lauter, M. Saarela and C. J. Tymczak, Phys. Rev. B 50, 6958 (1994).

3 B. E. Clements, H. Forbert, E. Krotscheck and M. Saarela, J. Low Temp. Phys. 95, 849 (1994).

4 E. Cheng, M. W. Cole, W. F. Saam and J. Treiner, Phys. Rev. B 46, 13967 (1992).

5 L. Pricaupenko and J. Treiner, J. Low Temp. Phys. 96, 19 (1994).

6 F. Dalfovo, A. Lastri, L. Pricaupenko, S. Stringari and J. Treiner, Phys. Rev. B 52, 1193 (1995).

7 E. Cheng, M. W. Cole and P. B. Shaw, J. Low Temp. Phys. 82, 49 (1991).

8 D. S. Greywall and P. A. Bush, Phys. Rev. Lett. 67, 3535 (1991).

9 G. Zimmerli, G. Mistura and M. H. W. Chan, Phys. Rev. Lett. 68, 60 (1992).

10 K. S. Liu, M. H. Kalos and G. V. Chester, Phys. Rev. B 13, 1971 (1976).

11 P. A. Whitlock, G. V. Chester and M. H. Kalos, Phys. Rev. B 38, 2418 (1988).

12 C. Carraro and M. W. Cole, Phys. Rev. B 46, 10947 (1992).

13 M. Wagner and D. Ceperley, J. Low Temp. Phys. 94, 185 (1994).

14 D. M. Ceperley and E. L. Pollock, Phys. Rev. B 39, 2084 (1989).

15 A. Belic and S. Fantoni, Physica B 194-196, 517 (1994).

16 R. A. Aziz, V. P. S. Nain, J. S. Carley, W. L. Taylor and G. T. McConville, J. Chem. Phys. 70, 4330 (1979).

17 R. A. Aziz, F. R. W. McCourt and C. C. K. Wong, Mol. Phys. 61, 1487 (1987).
18 J. Boronat and J. Casulleras, Phys. Rev. B 49, 8920 (1994).

19 P. J. Reynolds, D. M. Ceperley, B. J. Alder and W. A. Lester, J. Chem. Phys. 77, 5593 (1982).

20 S. A. Chin, Phys. Rev. B 42, 6991 (1990).

21 S. A. Chin and E. Krotscheck, Phys. Rev B 45, 852 (1992).

22 D. M. Ceperley and M. H. Kalos, in Monte Carlo Methods in Statistical Physics, edited by K. Binder (Springer, Berlin, 1979)

23 S. Zang and M. H. Kalos, J. Stat. Phys. 70, 515 (1993).

24 R. N. Barnett, P. J. Reynolds and W. A. Lester Jr., J. Comput. Phys. 96, 258 (1991).

25 J. Casulleras and J. Boronat, Phys. Rev. B 52, 3654 (1995).

26 W. L. McMillan, Phys. Rev. 138, 442 (1965).

27 L. Reatto, Nucl. Phys. A 328, 253 (1979).

28 B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir and P. R. Roach, Phys. Rev. A 1, 250 (1970).

29 E. Cheng, W. F. Saam, M. W. Cole, and J. Treiner, J. Low Temp. Phys. 92, 11 (1993).

30 J. Boronat and J. Casulleras, Phys. Rev. B 50, 3427 (1994).

31 H. A. Mook, Phys. Rev. Lett. 51, 1454 (1983).
FIGURES

FIG. 1. Equation of state for 2D liquid $^4$He. The solid circles correspond to the DMC energies obtained with the Aziz II potential (the statistical error bars are smaller than the size of the symbols); the solid line is the polynomial fit (11) to the calculated energies. The open diamonds are the GFMC results of Ref. 11 with the Aziz potential and the dashed line is the reported fit to these values.

FIG. 2. Density dependence of the surface pressure as obtained from our fit to the equation of state function (solid line), and from the fit of Ref. 11 (dashed line).

FIG. 3. Density dependence of the velocity of sound as obtained from our fit to the equation of state function (solid line), and from the fit of Ref. 11 (dashed line).

FIG. 4. Radial distribution function for three densities: $\rho = 0.275 \sigma^{-2}$ (solid line), $\rho = 0.320 \sigma^{-2}$ (short dashed line), $\rho = 0.420 \sigma^{-2}$ (long dashed line).

FIG. 5. Static structure function for three densities: $\rho = 0.275 \sigma^{-2}$ (solid line), $\rho = 0.320 \sigma^{-2}$ (short dashed line), $\rho = 0.420 \sigma^{-2}$ (long dashed line).

FIG. 6. Density dependence of the condensate fraction. Solid circles with error bars: results of DMC calculations; solid line: fit from eq. (20).

FIG. 7. Momentum distribution for three densities: $\rho = 0.275 \sigma^{-2}$ (solid line), $\rho = 0.320 \sigma^{-2}$ (short dashed line), $\rho = 0.420 \sigma^{-2}$ (long dashed line).
### TABLES

**TABLE I.** Results for the total and partial energies from DMC calculations. The potential energies per particle have been obtained from the calculation of pure expectation values.

| $\rho(\sigma^{-2})$ | $E/N(K)$ | $V/N(K)$ | $T/N(K)$ |
|---------------------|----------|----------|----------|
| 0.235               | -0.8480 ± 0.0016 | -3.850 ± 0.014 | 3.002 ± 0.014 |
| 0.255               | -0.8799 ± 0.0020 | -4.234 ± 0.022 | 3.354 ± 0.022 |
| 0.275               | -0.8950 ± 0.0019 | -4.680 ± 0.015 | 3.785 ± 0.016 |
| 0.320               | -0.8599 ± 0.0017 | -5.722 ± 0.013 | 4.862 ± 0.013 |
| 0.340               | -0.7932 ± 0.0031 | -6.213 ± 0.026 | 5.419 ± 0.026 |
| 0.380               | -0.5697 ± 0.0047 | -7.390 ± 0.026 | 6.820 ± 0.026 |
| 0.420               | -0.1524 ± 0.0047 | -8.532 ± 0.024 | 8.379 ± 0.025 |

**TABLE II.** Parameters of the equation of state (11) for our DMC results and the GFMC results of Ref. 11.

| Parameter | DMC          | GFMC         |
|-----------|--------------|--------------|
| $\rho_0(\sigma^{-2})$ | 0.28380 ± 0.00015 | 0.28458      |
| $e_0(K)$  | -0.89706 ± 0.00061 | -0.8357      |
| $B(K)$    | 2.065 ± 0.014 | 1.659        |
| $C(K)$    | 2.430 ± 0.035 | 3.493        |
| $\chi^2/\nu$ | 0.99        | 1.45         |
TABLE III. Position $r_m$ and height $g(r_m)$ of the first peak in the radial distribution function

| $\rho(\sigma^{-2})$ | $r_m$ (Å) | $g(r_m)$      |
|----------------------|-----------|---------------|
| 0.235                | 4.09      | 1.209 ± 0.005 |
| 0.255                | 4.03      | 1.233 ± 0.005 |
| 0.275                | 3.98      | 1.253 ± 0.002 |
| 0.320                | 3.87      | 1.323 ± 0.003 |
| 0.340                | 3.84      | 1.363 ± 0.009 |
| 0.380                | 3.63      | 1.426 ± 0.013 |
| 0.420                | 3.61      | 1.522 ± 0.006 |