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Puddles of helium in two dimensions: A Monte Carlo study

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In this paper we report simulation studies of two-dimensional puddles of liquid ^{4}He, using variational wave functions. We extract the line tension of these puddles and the energy in the bulk. We also study the surface profile and width of the surface region of the puddles as a function of system size. The wave functions are used to investigate the binding of boson ^{3}He. We find that mass-3 bosons are very weakly self-bound in two dimensions and fermions of the same mass are very unlikely to bind. Further we believe that a monolayer film of ^{3}He on graphite will not bind into a liquid puddle; it would be a gas at low densities and a solid at very high densities. [S0163-1829(99)06113-5]

I. INTRODUCTION

The properties of liquid and solid ^{4}He in two dimensions (2D) are interesting for the study of simple adsorbed systems and because they serve as a simple example of a two-dimensional quantum system. The properties of the homogeneous phase have been studied, theoretically,[1,2] using molecular dynamics,[3] and using Monte Carlo techniques.[4–8] In this paper we investigate the simplest inhomogeneous phase in two dimensions, viz., a puddle, using shadow wave functions. This is a natural continuation of a similar study of three-dimensional helium drops.[9,10] It provides us with insight into the nature of quantum droplets and quantum interfaces. One would like to know if a finite collection of He atoms will bind in 2D, how the binding energy varies with system size, and how these results change upon substitution of ^{4}He with ^{3}He. One would also like to know the nature of the interface between the liquid and vacuum and the line tension of the interface. It is known that thermal capillary wave fluctuations grow unbounded with system size in classical drops in two and three dimensions.[11,12] Do interfacial fluctuations grow with system size in these quantum systems and are these unbounded at absolute zero? Some of these questions are answered in the present investigation, while others need more extensive study and simulations.

A related question concerns the properties of ^{3}He in 2D. Variational calculations suggest that one needs about eight ^{3}He atoms to form a bound state in 3D.[10] What about two dimensions? Theoretical studies by Miller and Nasanow[13] first indicated the absence of a self-bound liquid phase for 2D ^{3}He. They also believed that the liquid phase would not be present in a monolayer film of ^{3}He adsorbed on a substrate. However, variational Monte Carlo simulations by Brami and co-workers[14] seemed to contradict this last result. They found a bound phase for a monolayer film of ^{3}He on graphite with a binding energy of 150–200 mK (relative to the energy of a single ^{3}He atom interacting with the graphite substrate). Nuclear magnetic resonance (NMR) experiments performed on a monolayer film of ^{3}He by Godfrin and Lauter[15] lead them to believe that the liquid phase is absent in the first layer of ^{3}He on graphite and with increasing coverage the film goes from a dense gas to a solid phase. We have simulated ^{3}He bosons in 2D, both finite size puddles as well as the homogeneous phase. Using this together with the Wu-Feenberg expansion[16] we estimate the energy of fermion ^{3}He and conclude that it will not bind. We have also estimated corrections to this energy in the presence of a graphite substrate and find that the system would continue to remain unbound.

Section II describes the variational wave function used in the simulation along with a brief description of the simulation method. The results from simulations of ^{4}He are presented in Sec. III. We then proceed to investigate ^{3}He puddles, both bosonic and fermionic, in Sec. IV and discuss our results for films of ^{3}He on graphite.

II. METHOD

The Hamiltonian for a system of N helium atoms in two dimensions is given by

$$H=-\frac{\hbar^2}{2m}\sum_i^N \nabla_i^2+\sum_{i<j}^N V(r_{ij}). \tag{1}$$

We use the Aziz I potential developed by Aziz and co-workers[17] to represent the interaction between helium atoms. The variational theorem gives us an upper bound on the true ground state energy E_0 of this system of helium atoms that form a drop,

$$E_0\leq E_T=\frac{\int dR\ \ \psi_T(R)H\ \ \psi_T(R)}{\int dR\ \ \psi_T(R)\ \ \psi_T(R)}. \tag{2}$$

We use a shadow wave function,[18] modified by a form factor, as the trial function $\psi_T(R)$, i.e.,

$$\psi_T(R)=\int \Xi(R,S)dS, \tag{3}$$

where $R\equiv(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)$, denotes the positions of the He atoms and $S\equiv(\mathbf{s}_1,\mathbf{s}_2,\ldots,\mathbf{s}_N)$ denotes N auxiliary shadow variables which are coupled to each other and to the real variables, and are integrated out in the definition of the wave function. The function Ξ is chosen to be

$$\Xi(R,S)=F_1(S)\exp\left(-\sum_{i<j}^{N} u_r(r_{ij})-C\sum_{i}^{N}(\mathbf{r}_i-\mathbf{s}_i)^2-\sum_{i<j}^{N} v_s(s_{ij})\right). \tag{4}$$

The pseudopotential for the real particles $u_r(r_{ij})$ is of the McMillan form $u_r(r_{ij})=(b/r_{ij})^5$, while the pseudopotential for the shadow variables is a scaled version of the Aziz potential,[17] $v_s(s_{ij})=\alpha V_{Aziz}(\delta s_{ij})$. Here b, α, and δ are variational parameters. Reals and shadows are coupled in Eq. (4) through the Gaussian coupling term whose strength C is a variational parameter. $F_1(S)$ is a one-body form factor which is a product of Fermi functions,

$$F_1(S)=\prod_{i}^{N}\left[\frac{1}{1+\exp[(s_i-s_0)/d_0]}\right], \tag{5}$$

where s_0 and d_0 are additional variational parameters, and the s_i are measured relative to the center of mass of the shadow particles. The wave function without the form factor has been used to simulate the homogeneous phase of ^{4}He in two dimensions and provides a good description of this system.[8] The form factor introduces inhomogeneity in the system and binds the shadows, to their center of mass, consequently binding the real particles. It is of course well known that some type of form factor is necessary to stabilize large 3D drops. They have been widely used,[10] and are usually expressed in terms of the particle coordinates. We have used the shadow variables. With this type of from factor it can be shown[9] that when a particle is removed to a large distance from the center of mass of the puddle the wave function delays exponentially. One can also show[9] that the energy obtained variationally with this form factor is very close to that obtained with a wave function in which there is no form factor but in which the shadow pseudopotential is a function of the local density. Both energies are close to the values obtained by Green's Function Monte Carlo (GFMC) simulations.[10] The trial energy for this wave function is given by

$$E_T=\int\int\int dR\, dS\, dS' p(R,S,S')\left[\frac{H\ \Xi(R,S')}{\Xi(R,S')}\right], \tag{6}$$

where the probability density

$$p(r,S,S')=\frac{\Xi(R,S)\ \Xi(R,S')}{\int\int\int dR\ dS\ dS'\ \Xi(R,S)\ \Xi(R,S')}. \tag{7}$$

The Metropolis method is used to sample this probability density and estimate the energy in square brackets in Eq. (6). The variational parameters are varied until one obtains a minimum in the energy for each system size that is simulated.

TABLE I. Variational parameters and energies as a function of number of particles, N ($\sigma=2.556$ Å).

N	$C\sigma^2$	b/σ	δ (K)	α	s_0/σ	d_0/σ	E/N (K)
2	2.8	1.12	0.09	0.92	1.65	1.3	−0.09 (1)
9	3.0	1.12	0.09	0.93	3.50	1.2	−0.26 (2)
16	3.0	1.12	0.09	0.91	4.70	1.2	−0.380(8)
36	3.1	1.12	0.09	0.93	7.10	1.3	−0.471(7)
64	3.1	1.12	0.08	0.93	9.40	1.4	−0.528(5)
121	3.1	1.12	0.09	0.93	12.70	1.5	−0.570(7)
165	3.4	1.12	0.08	0.94	14.30	1.45	−0.602(7)
512	3.1	1.13	0.09	0.93	26.00	2.0	−0.621(2)
1024	3.1	1.13	0.09	0.93	36.60	2.1	−0.640(2)

III. PUDDLES OF ^{4}He

We simulated the system with the number of particles ranging from 2 to 1024. The system was put in a circular box with hard walls. At the start of each run the particles were distributed uniformly in a circle (smaller than the box), with a density $\rho_b=0.0395$ Å^{-2} which is equal to the zero-pressure value determined from simulations of the homogeneous system[8] [the homogeneous simulations use a trial wave function which is a shadow wave function $\psi_T(R)$ without the one-body form factor imposed on the shadows]. The system was allowed to equilibrate for about 100 000 passes. Each pass consisted of 5 sweeps and during each sweep an attempt was made to move every particle in the system once. After equilibration, the simulation was run for a further 100 000 passes during which the energy was computed by averaging uncorrelated estimates. The parameters were varied until a minimum in energy was obtained for each system size. Table I shows the minimum energy per particle along with the optimum variational parameters. For the case of 512 and 1024 particles, a neighbor table scheme was used to reduce the computation time, and the simulation cell was changed from a circular box to a square box with periodic boundary conditions. In these large systems, a cutoff of 2.5σ was chosen for the potential. A tail correction[19] was made to the energy. We tested this correction by running a 512-particle system with and without the cutoff and found identical results within the statistical uncertainties of the calculations. It can be seen from the table that the parameters that optimize the inner region, C, b, δ, and α, do not vary much with system size. Their values are also close to the optimum values one would expect from simulations of bulk ^{4}He at a density ρ_b.[8] The parameters s_0 and d_0 are of course sensitive to the size of the system. However, it is seen that the energy is fairly insensitive to variations in d_0. This is because the interface fluctuates considerably in these puddles, leading to fluctuations in the energy in the course of a run with a given set of parameters. Consequently changing the interfacial width by a small amount by changing d_0 does not affect the energy significantly. The energy per particle can be fit to a quadratic function with bulk, surface, and ''curvature'' terms,

$$E/N=e_b+e_s x+e_c x^2. \tag{8}$$

Here $x=N^{-1/2}$, e_b should be very close to the bulk energy per particle of the homogeneous phase in the limit of large x, and $e_s^2=4\pi\gamma/\rho_b$, where γ is the line tension of the fluid. The results from fitting the parameters in Eq. (8) are shown in Table II along with the extracted line tension, and the line

TABLE II. Parameters that fit the energy in Eq. (8) and the corresponding line tension.

Range of N	e_b (K)	e_s (K)	e_c (K)	γ (K/Å)
9–1024	-0.676 (9)	1.1 (1)	0.4 (4)	0.063 (8)
16–1024	-0.68 (1)	1.2 (2)	0.04 (75)	0.07 (1)
36–1024	-0.66 (1)	0.7 (3)	2.5 (17)	0.07 (2)

of best fit is indicated in Fig. 1. We see that the line tension has a value of about 0.07 (2) K/Å. The experimental value for the surface tension of a three-dimensional drop is 0.27 K/Å ^{2}. A GFMC simulation of the planar interface[19] yields a value of 0.265. Å $^{-2}$. This close agreement with experiment shows that the Aziz I potential is accurate enough to provide a reliable value for the 3D surface tension. It is interesting to compare the value we have obtained for the line tension with the 3D surface tension. If we define a reduced surface tension $T\rho_3^{-2/3}$ and a reduced line tension $\gamma\rho_2^{-1/2}$, with ρ_3 and ρ_2 the 3D and 2D densities, respectively, then these reduced quantities both have the dimensions of energy. We find that the reduced line tension is approximately 10 times smaller than the reduced surface tension. However, there does not seem to be a unique, natural, way to define these reduced quantites. The other, plausible, method is to use the microscopic length scale σ, which appears in the two-body Aziz potential. Using σ to define reduced surface and line tensions we again find a ratio of approximately 10. We speculate that the binding energy of puddles, on a smooth graphite substrate, will be very close to those of the strictly 2D system. Our basis for this speculation is that the binding energy of the fluid first layer on graphite is only 5% greater than that of the 2D system.[25] We expect puddles to show a very similar difference.

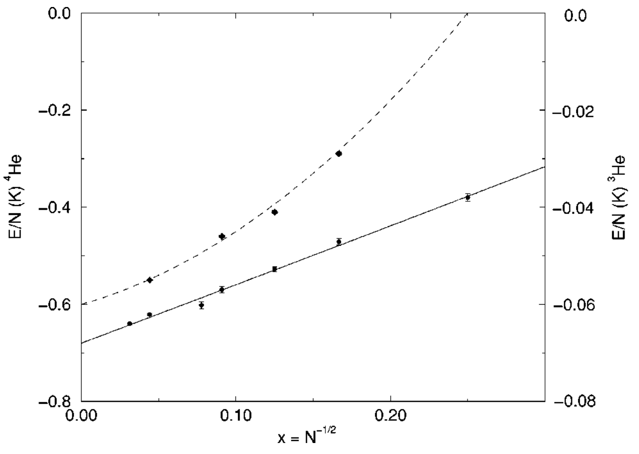

FIG. 1. The energy per particle fit to a quadratic function defined in Eq. (8). The data points for ^{4}He are fit by the solid line, the scale for which is indicated on the left, while the data points for ^{3}He are fit by the dashed line, the scale for which is indicated on the right.

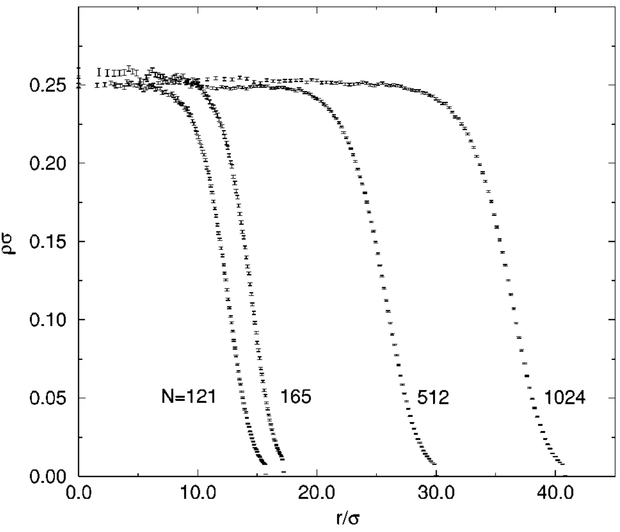

FIG. 2. The density profile in systems with $N = 121, 165, 512, 1024$ particles.

The density of the puddles as a function of radial distance is shown for the larger puddle sizes in Fig. 2. The density of the auxiliary ‘‘shadow’’ variables (which is not shown here) follows the density of the real particles very closely; the two are almost identical. We also observe that the density in the interior of the droplet is about 0.038 Å $^{-2}$, which is close to the value $\rho_b = 0.04$ Å $^{-2}$ in bulk ^{4}He. We see that the puddles have an interface whose width increases with increasing size. One measure of the width is the value of d_0 in Table I. d_0 varies from 3.07 to 5.37 Å as N varies from 16 to 1024. This is to be compared with a variation in d_0 of 3.83 to 6.65 Å as N varies from 20 to 700 particles in 3D drops.[10] A more direct measure of the width of the interface is the distance over which the density falls from 90% of its bulk value to 10%. This is tabulated for various drops sizes in Table III. A plot of this width vs $N^{1/4}$ is shown in Fig. 3. Although one cannot fit a perfect straight line through the points in the plot, it seems plausible that $W \sim N^{1/4}$, i.e., $W \sim \sqrt{L}$. This result is expected for a classical interface at finite temperature.[11,12] The reason we see this in our system is because any wave function whose form is given by Eq. (3) leads to a probability density that can be mapped to a classical system. With pseudopotentials of the kind we have used the equivalent potential functions of the classical system are short ranged. We would thus expect, in the absence of gravity, that the interfacial width in 2D will grow proportional to the square root of the length of the interface. This classical result arises from the excitation of long-wavelength surface waves on a 2D line. This argument essentially explains why we find the classical result for our wave function. In 3D the mean square width increases logarithmically with the linear dimension of the surface. In most circumstances in 3D, gravity acts per-

TABLE III. Width W of the interface as a function of N. Here W is the distance over which the density falls from 90% of its bulk value to 10%.

N	W/σ
36	4.3 (1)
64	4.7 (1)
121	4.98 (5)
165	5.1 (5)
512	6.8 (1)
1024	7.0 (2)

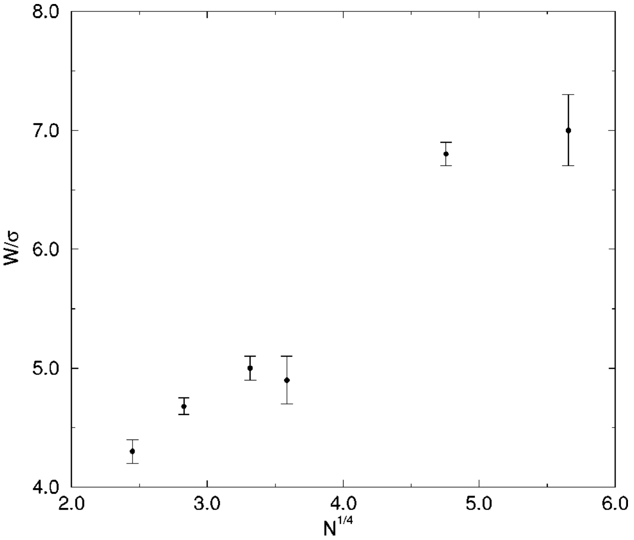

FIG. 3. The width W of the interface as a function of system size. W is the distance over which the density falls from 90% of its bulk value to 10%.

pendicular to the interface and suppresses interfacial fluctuations leading to a finite interfacial width in 3D.[11] However, physical realizations of a 2D system are often in the form of a film on a substrate which is perpendicular to the gravitational field. Consequently stabilization of the interfacial fluctuations is not possible in such systems. We believe, however, that if our wave function is modified to include the zero-point fluctuations of these surface waves, long-range correlations will be present in the wave function and these will be sufficient to invalidate the classical analogy.

A closer examination of the interfacial region of the puddles reveals a fundamental asymmetry in the interface. The one-body form factor has the property that it is symmetric about the point at which the density has dropped to one-half of the value in the interior of the drop. The form factor has a reflection symmetry about this point. Figure 4 is a plot of the density profile in a puddle with 512 particles. Also shown on the same graph is a 728-particle 3D drop simulated by Pieper and co-workers.[21] The x and y axes have been rescaled on this graph. The y axis has been rescaled so that the bulk density in the interior of the puddle asymptotes to a value of 1, and the x axis is rescaled so that the second derivative of the profile in the radial direction goes to zero at $x=1$. This is done so that we can compare 2D and 3D drops of different sizes on the same scale. The solid line is the Fermi function in the one-body form factor $F_1(S)$ used for the 512-particle simulation and is rescaled in the same fashion. It can be seen that the density profile does not follow the one-body form factor accurately in the interfacial region. No amount of adjusting of parameters (keeping the energy low) yields a symmetric profile. This is a feature observed in all the puddles that we have simulated. This is markedly different from classical simulations of three-dimensional drops[22] where the density profile is very well described by a hyperbolic tangent (a shifted and rescaled version of the Fermi function) and is symmetric. We do not know if this feature is fundamentally characteristic of helium. The density profiles in 3D ^{4}He drops, studied variationally by Pieper and co-workers,[21] also possess this asymmetry.

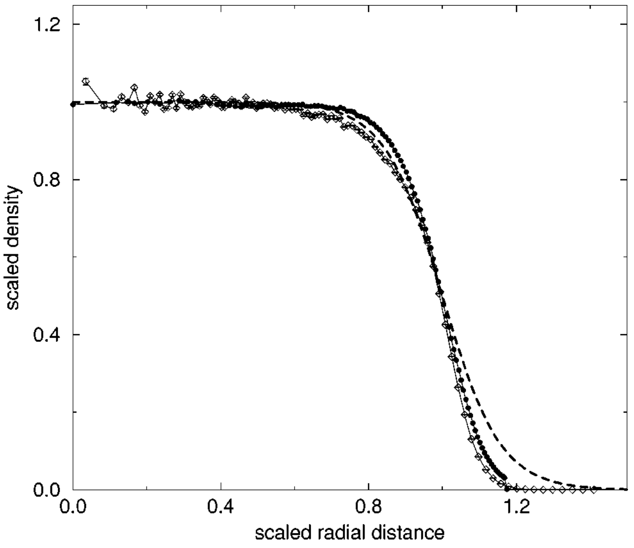

FIG. 4. The scaled density profile and the corresponding Fermi function (dashed line). The circles are the scaled density profile for a 512-particle 2D puddle while the diamonds represent a scaled 728-particle 3D drop.

There is a special feature of the interfaces of three- and two-dimensional boson drops which might explain the asymmetry. In drops of ^{4}He in 3D it has been shown that the condensed Bose state has a high amplitude in the interfacial region.[23,24] In the interior of the droplet the amplitude of the condensed state is a constant, with the value expected from bulk simulations. However, it peaks in the interfacial region. In principle we could attempt to calculate the condensate fraction for our 2D system and see if it also peaked in the interfacial region. Unfortunately our shadow wave function poses serious technical difficulties if one wishes to compute the condensed state function. These have been overcome for the homogeneous fluid,[18] but we have, as yet, no reliable method for inhomogeneous systems.

We should mention that the profiles of fermionic 3D drops are markedly different from their bosonic counterparts. Simulations reveal an interfacial region[10] that is much larger than a ^{4}He drop with the same number of particles. Furthermore, the density oscillates significantly in the bulk region of the drop due to filling of momentum shells, a feature absent in the bosonic simulations.

The two-dimensional puddles are low-density systems and we know from studying the homogeneous system that they are very weakly correlated. For this reason we believe that a good variation calculation will provide a reasonably accurate description of these puddles. We know that for the homogeneous system the variation energy, at low densities, is within 10% of the GFMC value.[7,8] Since the interfacial region is even less dense, we expect that our variation calculations will be at least as accurate for the properties of the interface. This claim is supported by the fact that the difference in the surface tension found in variational and GFMC calculations is only 4%.[19]

IV. PUDDLES OF ^{3}He

We now turn our attention to the existence of puddles of ^{3}He. We start by dealing with the strictly two-dimensional

TABLE IV. Variational parameters and energies for puddles of boson ^{3}He as a function of N ($\sigma = 2.556$ Å).

N	$C\sigma^2$	b/σ	δ (K)	α	s_0/σ	d_0/σ	E/N (K)
2	1.5	1.12	0.09	0.93	3.1	1.2	-0.001 (10)
3	1.4	1.12	0.09	0.93	4.0	1.2	0.010 (20)
9	1.4	1.12	0.09	0.93	6.8	1.2	-0.009 (10)
10	1.4	1.12	0.09	0.93	7.0	1.2	-0.003 (6)
11	1.4	1.12	0.09	0.93	7.6	1.2	-0.001 (10)
12	1.4	1.12	0.09	0.93	7.8	1.2	-0.010 (5)
13	1.4	1.12	0.09	0.93	8.6	1.2	-0.013 (5)
16	1.4	1.12	0.09	0.93	8.9	1.2	-0.016 (6)
36	1.3	1.12	0.09	0.93	13.4	1.3	-0.029 (3)
64	1.4	1.12	0.09	0.92	17.5	1.5	-0.041 (3)
121	1.4	1.12	0.09	0.93	22.5	1.6	-0.046 (2)
512	1.4	1.12	0.09	0.93	45.0	2.0	-0.055 (1)

system. It has been suspected for some time that 2D ^{3}He will not bind.[13] If this is true, then puddles of ^{3}He will not form. Before making any predictions for 2D ^{3}He we first studied the mass-3 boson system in 2D. Once again we simulated the homogeneous liquid phase of this system using a trial shadow wave function $\psi_B(R)$ which is identical to $\psi_T(R)$ in Eq. (3) except that the one-body form factor for the shadows is not included. We determined the equilibrium density (at zero pressure) to be $\rho_e^{(3)} = 0.0121$ Å^{-2} and the energy at this density to be $e_b^{(3)} = -0.063$ (2) K per particle, respectively. The corresponding density and energy for mass-4 bosons in 2D are $\rho_e^{(4)} = 0.0395$ Å^{-2} and $e_b^{(4)} = -0.687$ (2) K, respectively.[7] We see that bosons of mass 3 are very weakly bound in 2D. Further the equilibrium density is also extremely low. Next, we proceeded to determine the minimum number of mass-3 bosons that would bind in 2D. We simulated puddles of bosonic ^{3}He with the same trial shadow wave function $\psi_T(R)$ described in Sec. II. The results of these simulations are shown in Table IV. The 512-particle run used a neighbor table scheme with a cutoff of 5.0σ since the density in these systems is very low. We see that the minimum number of mass-3 bosons that bind in 2D is greater than or equal to 12. For puddles with fewer particles the fluctuations in the energy are large enough to destroy binding. We also fit the energy per particle to a quadratic in $x = N^{-1/2}$ as we did in Sec. III. The results of the fitting are shown in Table V and the best fitting curve in Fig. 1. We see that e_b in Table V is close to the bulk value $e_b^{(3)} = -0.063$ K. The line tension for mass-3 bosons is an order of magnitude lower than the line tension for the mass-4 case, and cannot be accurately determined from these simulations.

To estimate the binding energy for ^{3}He fermions we used the Wu-Feenberg expansion.[16] This expansion appears to be very rapidly convergent at these very low densities. We assume that the shadow wave function $\psi_B(R)$ used to simulate the homogeneous phase of mass-3 bosons is an exact ground state. For the fermion system we choose a wave function $\psi_F(R)$, which is a product of $\psi_B(R)$, and a Slater determinant of plane waves with wave vectors $\mathbf{k}_i$ and spin functions χ_i filling a Fermi disk of radius $k_F = (2\pi\rho)^{1/2}$, where ρ is the density, i.e.,

$$\psi_F(R) = \psi_B(R)\,\mathrm{Det}[\exp(i\mathbf{k}_i\cdot\mathbf{r}_j)\ \ \chi_i(\sigma_j)]. \tag{9}$$

A few definitions are in order. Let $g_B(r)$ be the pair correlation function for the boson system described by $\psi_B(R)$, and $u(k)$ be defined by

$$u(k) \equiv S_B(k) - 1 = \rho \int \exp(i\mathbf{k}\cdot\mathbf{r})[g_B(r) - 1], \tag{10}$$

where $S_B(k)$ is the structure factor.

The energy per particle for the 2D fermion system can be expanded in a permutation expansion,[16]

$$e_F = e_B + e_{WF}^0 + e_{WF}^1, \tag{11}$$

where $e_B = e_b^{(3)} = -0.063$ K is the energy for the mass-3 boson system. The zeroth- and first-order corrections (in permutations) are given by $e_{WF}^0 = \epsilon_F/2$, and $e_{WF}^1 = \epsilon_F\rho\Delta$, where ϵ_F is the Fermi energy and

$$\Delta = \int_0^1 16x^3 u(2k_F x)\left[\pi/2 - x(1-x^2)^{1/2} + \arctan\left(\frac{-x}{(1-x^2)^{1/2}}\right)\right]dx. \tag{12}$$

At the equilibrium density $\rho_e^{(3)}$, the Fermi energy $\epsilon_F = 0.6114$ K per particle, and $\Delta = -0.305\,821$. Here Δ was determined by simulating the system at a density $\rho_e^{(3)}$, to find $g_B(r)$, and then numerically integrating Eq. (12). This yields corrections $e_{WF}^0 = 0.3057$ K, and $e_{WF}^1 = -0.0027$ K. We see that the first-order correction is two orders of magnitude smaller than the zeroth order. Higher-order corrections to the energy involve higher powers of the density, and we suspect that they contribute an extremely small amount to the energy of the fermion system. The bulk of the contribution to the energy of the fermion system in 2D is from the energy of the free Fermi gas. Because the mass-3 boson system condenses at such a very low density, we believe that our variational estimate of the energy will be very close to the exact value. For this reason we can be confident in asserting that ^{3}He will not bind in 2D.[20]

Can this lack of binding be observed in some experimental realization of the 2D system? The closest one could come to a real 2D system is a monolayer of ^{3}He on graphite. One could ask if this thin film would puddle and form a region of dense liquid. We have already estimated how much the binding energy of a thin film of ^{4}He on graphite would be lowered relative to its 2D value due to opening out of the wave function in the z direction.[25] We expect the opening of the wave function to lower the energy because while the greater volume available to the particles will decrease the magnitude of the potential energy it will at the same time decrease the kinetic energy to a larger extent. We follow the same proce-

TABLE V. Parameters that fit the energy per particle in Eq. (8) and the corresponding line tension for boson ^{3}He.

Range of N	e_b (K)	e_s (K)	e_c (K)	γ(K/Å)
36–512	-0.060 (5)	0.09 (9)	0.6 (5)	0.003 (3)

dure as in the reference mentioned above.[25] The Hamiltonian for a film of N helium atoms in the presence of graphite is

$$H^{\text{film}}=T_x+T_y+T_z+V_{\text{He-C}}+V^3_{\text{He-He}}, \tag{13}$$

where the T's represent the kinetic energy operators and the V's represents the potential energy. For simplicity we assume that the carbon can be modeled as a smooth substrate with a potential that varies only in the z direction. The above equation can be rewritten as

$$H^{\text{film}}-H^{\text{C}}=H^{\text{2D}}+V^3_{\text{He-He}}-V^2_{\text{He-He}}, \tag{14}$$

where $H^{\text{C}}=T_z+V_{\text{He-C}}$ is the part of the Hamiltonian that represents the interaction with the carbon, and H^{2D} is the Hamiltonian for 2D helium. Let $\phi(z)$ be the ground state wave function of one helium atom in the presence of the carbon substrate. Then a plausible trial wave function for the film is $\Psi_{\text{film}}(R)=\psi_B(R)\Pi_i^N\phi(z_i)$ and using this in Eq. (14) yields an upper bound for the energy (per particle) relative to the carbon binding energy:

$$e^{\text{film}}-e^{C}\leqslant e_B+\Delta e; \tag{15}$$

i.e., the binding energy changes by $\Delta e=\langle V^3_{\text{He-He}}-V^2_{\text{He-He}}\rangle$. Monte Carlo simulations yield a value $\Delta e=-0.0195\ (5)$ K per particle. We see that this correction to the binding energy e_B is still insufficient to overcome the free Fermi gas energy term from the zeroth-order Wu-Feenberg correction. This leads us to conclude that a monolayer film of ^{3}He will not bind and puddle on a graphite substrate. Increasing the density of the film would make it go from a gas phase to a high-density gas phase to a solid without an intervening liquid phase.

The work of Brami and co-workers[14] directly contradicts our results on the binding of monolayer ^{3}He films. In the same paper the authors quote results for the binding energy of a monolayer film of ^{4}He. They find a large increase in the binding energy when compared with 2D helium. This result also contradicts other work carried out by our group[25] and we have been unable to reproduce the results of Brami and co-workers.

V. CONCLUSIONS AND FUTURE WORK

We have simulated puddles of bosonic ^{4}He and ^{3}He in 2D. We see that the energy per particle in these puddles decreases with system size and approaches the value expected for bulk homogeneous 2D He. The width of the interface region increases with system size. However, to obtain more accurate information on the size dependence of the width one needs to simulate much larger systems using a geometry that consists of a strip of helium at the center of a rectangular cell with periodic boundary conditions. Furthermore, the wave function we have used in our simulation does not contain in it quantum zero-point capillary wave fluctuations. These may bind the fluctuations of the interface and prevent them from increasing indefinitely. A possible way to include this in the wave function is to use the dispersion relation for $\omega^2=k^3$ for capillary waves, assume a harmonic Hamiltonian for capillary waves, and modify the wave function in a manner similar to modifications for the bulk done by Reatto and Chester.[26] This modified wave function could be simulated, with the strip geometry described above, and one could check to see if the addition of zero-point capillary waves results in bounded interfacial fluctuations. Computation of the condensate fraction in 2D systems, particularly inhomogeneous ones, is another important direction for future work. This would enable us to check if the presence of the condensate modifies the interfacial profile for a quantum system and makes it asymmetric in contrast with the symmetric one seen in classical systems.

From our simulations for the mass-3 boson system we believe that the mass-3 fermion system will not bind, either as a pure 2D system or in the form of a monolayer of ^{3}He on graphite. Experiments seem to corroborate this hypothesis although some simulations seem to suggest otherwise. A clear-cut answer will need more careful and detailed experiments and/or simulations. Experiments also suggest that the third layer has a bound state consisting of a puddle of liquid.[15] Future work would also involve investigating in detail the properties of the second- and third-layer films of ^{3}He on graphite to check if these condense to form a self-bound liquid.

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