$^3$He on preplated graphite

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By using the diffusion Monte Carlo method, we obtained the full phase diagram of $^3$He on top of graphite preplated with a solid layer of $^4$He. All the $^4$He atoms of the substrate were explicitly considered and allowed to move during the simulation. We found that the ground state is a liquid of density $0.007 \pm 0.001 ~\text{Å}^{-2}$, in good agreement with available experimental data. This is significantly different from the case of $^3$He on clean graphite, in which both theory and experiment agree on the existence of a gas-liquid transition at low densities. Upon an increase in $^3$He density, we predict a first-order phase transition between a dense liquid and a registered 7/12 phase, the 4/7 phase being found metastable in our calculations. At larger second-layer densities, a final transition is produced to an incommensurate triangular phase.

I. INTRODUCTION

Recent heat-capacity studies of $^3$He on graphite\textsuperscript{1–3} both clean and preplated, have reopened the interest on the phase diagram of this quasi-two dimensional (quasi-2D) system. In essence, those results confirm a previous hint about the existence of a stable $^3$He liquid at very low densities\textsuperscript{3} and are the continuation of a wealth of experimental work on this quantum fluid\textsuperscript{2,14,15}. On clean graphite, the introduction of corrugation in the theoretical description of the surface\textsuperscript{14,15} proved to be the necessary ingredient to produce a good match between those calculations and the experimental results of Sato et al.\textsuperscript{2} that indicated the existence of a gas-liquid transition at very low densities. When flat substrates were considered, no liquid was found to be stable in most calculations\textsuperscript{16–21}, but not in all of them\textsuperscript{22}.

When $^3$He is adsorbed on $^4$He-preplated graphite, the experimental data of Ref. 2 shows a linear dependence of the heat capacity versus the second layer density in the range $\rho \sim 0.007 ~\text{Å}^{-2}$. This suggests that the ground state of that $^3$He monolayer is a homogeneous liquid with a density fixed by the upper bound of the interval. For a smaller number of atoms on the same surface, the system is fragmented in drops of 0.007 Å\(^{-2}\) with enough empty space between them to produce the average 2D helium concentration we consider. This is different from what happens in the first layer adsorbed directly on graphite\textsuperscript{2,14,15} in which there is a gas of $\sim 0.006 ~\text{Å}^{-2}$ in equilibrium with a liquid of $\sim 0.014 ~\text{Å}^{-2}$. In this second case, what we have are drops of density 0.014 Å\(^{-2}\) surrounded by a gas of 0.006 Å\(^{-2}\) in the right proportions to produce any density in that range. When $\rho$ is above 0.007 and 0.014 Å\(^{-2}\) for the second and first layers, respectively, we have homogeneous liquids covering all the surface.

In this work, we calculate the complete phase diagram of $^3$He on $^4$He preplated graphite, including the high density solid region. Given the importance of considering corrugation in the description of the bare substrate, we considered explicitly all the $^4$He atoms on the first layer, allowing them to move during the simulations. This means that there can be deviations between the actual positions of those atoms and their corresponding crystallographical sites. That produced larger error bars in the statistical sampling than in the case of a flat surface, but it was found to be necessary to reproduce the experimental results, both in the low and high density regimes.

The rest of the paper is organized as follows. In Sec. II, we discuss the quantum Monte Carlo method used for the simulations and account for the interaction models and trial wave functions used as importance sampling. The obtained results and their comparison with experimental data are reported in Sec. III. Finally, Sec. IV comprises the main conclusions of the present work.

II. METHOD

We rely on a microscopic approach to the physical problem of a second layer of $^3$He atoms adsorbed on $^4$He-preplated graphite. The Hamiltonian is written as,

$$
H = \frac{\hbar^2}{2m_3} \sum_{i=1}^{N_3} \nabla_i^2 - \frac{\hbar^2}{2m_4} \sum_{i=1}^{N_4} \nabla_i^2 + \sum_{i=1}^{N} V_{\text{ext}}(\mathbf{r}_i) + \sum_{i<j}^{N} V(\mathbf{r}_{ij}) ,
$$

where $N_3$ and $N_4$ are the number of $^3$He and $^4$He atoms ($N = N_3 + N_4$), of masses $m_3$ and $m_4$, respectively. $V_{\text{ext}}(\mathbf{r})$ is the total interaction of each helium atom at position $\mathbf{r}$ with all the carbon atoms in the graphite substrate. This term is made up of a sum of individual C-He interactions, each of them modeled by the anisotropic Carlos-Cole potential\textsuperscript{26-27}, even though no influence of the C-He anisotropy is expected in the behaviour of the second layer, as has been already proved for a second
layer of $^4$He on the same substrate. As in previous works, the substrate structure was modeled by an stack of honeycomb 2D-lattices separated by 3.35 Å in the typical A-B-A-B graphite disposition. $V(r)$ is the standard Aziz potential for the helium-helium interaction of two atoms separated a distance $r$.

We considered unpolarized $^3$He, i.e., $N_\uparrow = N_\downarrow = N_3/2$. To solve the $N$-body Schrödinger equation corresponding to the Hamiltonian of Eq. 1, we used the fixed-node Diffusion Monte Carlo (FN-DMC) method. This stochastic technique solves the Schrödinger equation in imaginary time, providing the exact energy for the ground state of a Bose system and an upper bound for a Fermi one. Following Ref. 15, the one given in Ref. 11 (the experimental limit given in Ref. 2, a limit slightly larger than the one given in Ref. 11 ($\rho \sim 0.116$ Å$^{-2}$). The parameter $b_3$ in Eq. 3 is variationally optimized; its value is set to $b_3 = 2.96$ Å, as in Refs. 13 and 20. The coordinates in the Slater determinants were corrected by backflow terms in the standard way,

$$\tilde{x}_i = x_i + \lambda \sum_{j \neq i} \exp[-(r_{ij} - r_b)^2/\omega^2](x_i - x_j) \quad (4)$$

$$\tilde{y}_i = y_i + \lambda \sum_{j \neq i} \exp[-(r_{ij} - r_b)^2/\omega^2](y_i - y_j). \quad (5)$$

The optimal values for the parameters in the backflow term were those of the bulk three-dimensional system, i.e., $\lambda = 0.35$; $\omega = 1.38$ Å, and $r_b = 1.89$ Å, since they were proved to give lower energies, that those corresponding to a pure 2D system.

The part of the trial wave function corresponding to the first $^4$He layer is taken as

$$\Psi_4(\mathbf{r}_{N_3+1}, \mathbf{r}_{N_3+2}, \ldots, \mathbf{r}_N) = \prod_{i} u_4(\mathbf{r}_i) \times \prod_{i<j} \exp \left[-\frac{1}{2} \left(\frac{b_4}{r_{ij}}\right)^2\right] \times \prod_{i} \exp \left\{-a_4[(x_i - x_{\text{site}})^2 + (y_i - y_{\text{site}})^2]\right\} \quad (6)$$

The last (Nosanov) term in the equation pins the atoms around their crystallographic positions. The free parameters in $\Psi_4$: $b_4 = 3.07$ Å (as in Refs. 22 and 30), and $a_4$ has the same value that in a previous study of the second layer of $^4$He on $^4$He ($0.55$ Å$^{-2}$). As before, $u_4(\mathbf{r}_i)$ is the numerical solution of the one-body Schrödinger equation for one $^4$He atom on top of graphite.

Eq. 2 defines adequately the $^3$He layer when it is a liquid or a gas. If this is not so, we have to introduce a Nosanov term to confine the $^3$He atoms,

$$\prod_{i} \exp \left\{-a_3[(x_i - x_{\text{site}})^2 + (y_i - y_{\text{site}})^2]\right\} \quad (7)$$

in which we used the same $a_3$ parameter for all the solid phases and densities ($a_3 = 0.24$ Å$^{-2}$). We checked that neither an increase or a decrease of up to 50% in $a_3$ varies the stability ranges of the solids described. The trial wave function (2) is factorized in a term depending only on the $^3$He fermions. The one-body function $u_3(\mathbf{r})$ is the numerical solution of the Schrödinger equation that describes a single $^3$He atom on top of a triangular lattice formed by $^4$He atoms located in the crystallographic positions ($x_{\text{site}}, y_{\text{site}}$) corresponding to an incommensurate triangular phase. To solve this equation, we used the same technique as in Ref. 13 and neglected the influence of the underlying graphite structure. In the present work, we used two triangular $^4$He lattices at different densities: 0.112 Å$^{-2}$ (corresponding to a separation of 3.2 Å), and 0.12 Å$^{-2}$ (with a He-He lattice constant of 3.1 Å). The first density corresponds to the $^4$He promotion to the second layer, while the latter is the upper experimental limit given in Ref. 2, a limit slightly larger than the one given in Ref. 11 ($\rho \sim 0.116$ Å$^{-2}$). The parameter $b_3$ in Eq. 3 is variationally optimized; its value is set to $b_3 = 2.96$ Å, as in Refs. 13 and 20. The coordinates in the Slater determinants were corrected by backflow terms in the standard way,

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Eq. 2 defines adequately the $^3$He layer when it is a liquid or a gas. If this is not so, we have to introduce a Nosanov term to confine the $^3$He atoms,

$$\prod_{i} \exp \left\{-a_3[(x_i - x_{\text{site}})^2 + (y_i - y_{\text{site}})^2]\right\} \quad (7)$$

in which we used the same $a_3$ parameter for all the solid phases and densities ($a_3 = 0.24$ Å$^{-2}$). We checked that neither an increase or a decrease of up to 50% in $a_3$ varies the stability ranges of the solids described. The trial wave function (2) is factorized in a term depending only on the $^3$He coordinates and other only on the $^4$He ones. Simulations including a Jastrow term relating atoms in both layers produced no significant change in the obtained results.

III. RESULTS

In Fig. 1 we show the FN-DMC results for the energy per $^3$He atom as a function of the second-layer density. The upper (lower) curve corresponds to a fixed first-layer density of 0.112 (0.12) Å$^{-2}$. We chose those densities as
lower and upper limits to the magnitude at hand, and used these two sets of simulations to test the effect of a possible compression of the $^4\text{He}$ solid structure upon an increase of the number of $^3\text{He}$ atoms on top of it. The curves displayed are third-order degree polynomials obtained by least-squares fitting to the shown data.

From those fits, we can say state that the energy per $^3\text{He}$ atom on $^4\text{He}$-preplated graphite in the infinite dilution limit, $E_0$, is $-24.45 \pm 0.04 \text{ K}$ in the first case, and $-24.74 \pm 0.07 \text{ K}$ for the denser preplating, values much smaller in absolute value than the same magnitude for $^3\text{He}$ on bare graphite, $-135.77 \pm 0.001 \text{ K}$. In both cases, we used a simulation cell that comprised $14 \times 8$ incommensurate triangular lattice cells. Those rectangular unit cells contain two $^4\text{He}$ atoms, and have a surface $d_{\text{He}} \times \sqrt{3}d_{\text{He}}$, $d_{\text{He}}$ being the distance between two atoms in the first layer. This means that for the upper curve the simulation cell was $44.8 \times 44.34 \text{ Å}^2$, and for the lower one, $43.4 \times 42.95 \text{ Å}^2$. In both cases, that means $224$ $^4\text{He}$ atoms on the lower layer and enough $^3\text{He}$ atoms to produce the densities shown. This means up to $102$ $^4\text{He}$ atoms in the case of the second-layer liquids discussed below. In all the simulations, standard finite-size corrections to the fermionic wavefunction were applied. As indicated in the preceding Section, all the $^3\text{He}$ atoms on the first layer were allowed to move during the simulations, i.e., we solve the full Hamiltonian (1).

Our results, reported in Fig. 1 show second layer is a liquid, since in both cases we have self-bound structures of densities $0.007 \pm 0.001 \text{ Å}^{-2}$ (upper curve) and $0.017 \pm 0.001 \text{ Å}^{-2}$ (lower one). Those values were obtained from the fits displayed. At difference with the first layer on bare graphite structure, no gas-liquid transition was found. To our knowledge, the only experimental set of available data to compare our results to are those of Ref. 2. As indicated above, at very low densities, the experimental heat capacity depends linearly on the second layer density in the range $0-0.007 \text{ Å}^{-2}$. This would indicate a phase separation between a clean surface and an homogeneous liquid of $0.007 \text{ Å}^{-2}$. This is exactly what can be deduced from the upper curve of Fig. 1. From that, we can conclude that a model with a pre-plated density of $0.112 \text{ Å}^{-2}$ is a good description of the experimental setup.

Our results are different from the results of a previous theoretical calculation on this same system. In that work, the authors found a gas-liquid transition of the same type that the one for the first layer on bare graphite. To explain that discrepancy, we use the results displayed in Fig. 2. There, the lower set of data is the one in the previous figure after having subtracted the energy in the infinity dilution limit, $E_0$. On the other hand, the open circles correspond to a calculation in which the effect of the first $^4\text{He}$ layer and the graphite surface below it have been described by a laterally z-averaged potential, something similar to the description made in Ref. [14]. In this flat case, $E_0 = \pm 0.015 \text{ K}$. Even tough the results in Ref. [14] are not exactly the same as the ones in Fig. 2, since the helium-helium potentials are slightly different, both sets of data look pretty similar to each other. At the same time, both of them are noticeably different from the simulations made with the full Hamiltonian (1). The error bars of those results are of the size of the symbols, and not displayed for simplicity.

The missing ingredient is the corrugation of the $^4\text{He}$ substrate, as in the bare graphite case. To check that, we repeated our calculations but without allowing the movement of the $^4\text{He}$ atoms in the first layer. What we see is that those results are within the (rather large) error bars of those corresponding to the full $^4\text{He}$-moving calculation, and present a minimum at approximately the same density. In our opinion, this indicates that the introduction of a $^3\text{He}$ effective mass, used in the Ref. [14] calculations, is not enough to compensate for the use of a z-averaged potential. The error bars of these last results are comparable, though somewhat smaller than the ones for the full calculation, and again were not displayed for simplicity.

Fig. 3 shows the high density end of the $T = 0$ isotherm for a $0.112 \text{ Å}^{-2}$ underlying density. There, we have displayed the energies per $^3\text{He}$ atom corresponding to a liquid phase (full squares), incommensurate triangular solid (open squares), and two standard commensurate structures on second-layer-helium structures: $4/7$ (open circle), and $7/12$ (full circle). By a simple inspection, we can see that there is practically no difference between the energies of the $4/7$ lattice, of density $0.064 \text{ Å}^{-2}$, and that of an incommensurate triangular structure of the same density. In addition, the energy per particle of a $7/12$ structure is lower than both the one cor-

\begin{figure}
\centering
\includegraphics[width=0.8\textwidth]{fig1}
\caption{(Color online) Energy per $^3\text{He}$ atom ($E/N$) on the second layer of $^3\text{He}$ on graphite as a function of the second-layer density. Upper curve, underlying $^4\text{He}$ density $0.112 \text{ Å}^{-2}$; lower curve, first layer density $0.12 \text{ Å}^{-2}$. The curves are third-order polynomial fits to the data displayed.}
\end{figure}
If, as suggested in Ref. 11, the density of the first solid of 0.070 Å\(^2\) results.

one would expect density limits for the stable phases in Fig. 4, where the symbols and lines have the same meaning as in the previous figure, and the upper one, to the pure 2D result of Ref. 24.

responding to the 4/7 commensurate solid and that of the incommensurate arrangement of equal density (0.066 Å\(^2\)). This means that we can draw a double-tangent Maxwell construction line that starts in a liquid of density 0.047±0.002 Å\(^2\) and ends in the energy corresponding to that 7/12 structure, i.e., those phases are in equilibrium with each other. When the density increases further, another Maxwell construction shows that the 7/12 solid is in equilibrium with an incommensurate triangular one of density 0.072±0.004 Å\(^2\), that is probably stable up to the third layer promotion.

In the same way as before, to check for the influence of the first layer density on the phase diagram of the second layer, we performed another series of calculations for a \(^4\)He density of 0.12 Å\(^2\). The results are displayed in Fig. 4 where the symbols and lines have the same meaning as in the previous figure. The main differences are the densities at which the different transitions are produced: the liquid is now stable up to 0.054±0.002 Å\(^2\), and it is in equilibrium with a 7/12 commensurate solid of 0.070 Å\(^2\), that undergoes a first-order phase transition to a triangular solid of \(\rho = 0.074±0.002\) Å\(^2\). If, as suggested in Ref. 11, the density of the first \(^4\)He layer in a real setup is between those considered here, one would expect density limits for the stable phases in the range 0.179 and 0.185 Å\(^2\). This is comparable to our results in both series of simulation data. In addition, magnetization and heat-capacity measurements on a double \(^3\)He layer, give a solid-solid transition region similar to the one that we see from a commensurate structure to the lower density for which the incommensurate phase is stable (total density in the range 0.179 and 0.185 Å\(^2\)). However, even though those data are close to our results, they are not directly comparable to them, since it is well known that the density of the first layer of the system \(^3\)He/\(^3\)He/graphite is ~ 5% smaller than the \(^3\)He/\(^4\)He/graphite one. Finally, the experimental upper second layer density for a liquid in equilibrium with the commensurate phase (7/12 in our case, but 4/7 in virtually all the previous literature) is 0.058±0.005 Å\(^2\). This is comparable to our 0.12 Å\(^2\) result, what would support a compression of the first layer upon population of the first one.

IV. DISCUSSION

With that in mind, we can say that our results compare favorably with the available experimental data. In particular, a solid is found for second layer densities larger than 0.072 Å\(^2\), a density which is well within the error bars of our results in both series of simulation data. This would correspond to the lower stability limit of the triangular phase. In addition, magnetization and heat-capacity measurements on a double \(^3\)He layer, give a solid-solid transition region similar to the one that we see from a commensurate structure to the lower density for which the incommensurate phase is stable (total density in the range 0.179 and 0.185 Å\(^2\))). However, even though those data are close to our results, they are not directly comparable to them, since it is well known that the density of the first layer of the system \(^3\)He/\(^3\)He/graphite is ~ 5% smaller than the \(^3\)He/\(^4\)He/graphite one. Finally, the experimental upper second layer density for a liquid in equilibrium with the commensurate phase (7/12 in our case, but 4/7 in virtually all the previous literature) is 0.058±0.005 Å\(^2\). This is comparable to our 0.12 Å\(^2\) result, what would support a compression of the first layer upon population of the first one.
The Hamiltonian does nor include any ferromagnetic or antiferromagnetic spin-spin interaction, something that could be important and eventually change the relative stability of the 4/7 and 7/12 phases.

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