PATH INTEGRAL MONTE CARLO SIMULATIONS OF
THIN 4HE FILMS ON A H2 SURFACE

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Abstract

Atomically thin 4He films of up to three monolayers on molecular hydrogen (1,1,1) surfaces are studied at T=0.5 K, using path integral Monte Carlo. We compute the binding energy of 4He to the H2 substrate as a function of 4He coverage and obtain evidence of the prewetting transition. Density profiles perpendicular to the 4He-H2 interface are obtained, as well as the zero point motion and effective mass of 4He parallel to the substrate surface. The superfluid density of 4He v.s. coverage is calculated, and the intermediate scattering function is computed, from which we estimate the speed of third sound. Finally, we calculate the vorticity-vorticity correlation function.
I. INTRODUCTION

Thin films of liquid $^4$He adsorbed on H$_2$ surfaces at low temperatures are particularly interesting physical systems because of the large quantum effects of both constituents. By adjusting the $^4$He film thickness it is possible to go from a two dimensional monolayer to three dimensional bulk $^4$He. Much of the physics at the $^4$He-H$_2$ interface is dominated by quantum many-body effects, notably the interpretation of the experimental means of investigation; third sound and heat capacity measurements.

But in spite of a long history of experimental$^{1-12}$ and theoretical$^{3-5}$ research done on $^4$He films on various substrates, even the results for the ground state energy of a single $^4$He atom bound to the H$_2$ surface vary by a factor of about 1.5$^{6,7}$. This seems disconcerting because the pair potential$^8$ between a $^4$He atom and a H$_2$ molecule is known reasonably accurately. The situation is worse yet for the strength of the potential between a $^4$He atom and the H$_2$ surface, because the controversy about its asymptotic long distance attraction ranges over a factor of four$^{9-12}$. From the theoretical side, no $ab\ initio$ calculations have dealt with this system.

We believe to be able to fill some of the gaps with path integral simulations. Nothing other than the semi-empirical pair potentials among isolated $^4$He atoms and H$_2$-molecules are input, and all zero point, thermal, and exchange effects of the $^4$He-H$_2$ system can be calculated without approximation from first principles. The Path Integral Monte Carlo (PIMC) method underlying our computations has been described in detail by Ceperley and Pollock$^{13}$. In a companion paper$^{14}$ we discussed our simulations of the bare hydrogen surface and introduced a general procedure to incorporate the long range contributions of a heterogeneous and layered substrate-adsorbate system in an “external potential”.

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In sections III and IV, respectively, we report energies and density profiles of $^4$He films on the H$_2$ surface at T=0.5K for $^4$He coverages from 0.1 to 3 monolayers. Recently, wetting and prewetting transitions of $^4$He films on weakly binding substrates are getting much attention$^{15,16}$, and Cheng et al.$^{15}$ have predicted a prewetting transition for $^4$He on H$_2$ with a density functional method. We shall discuss our binding energy as a function of $^4$He coverage in that context. Whitlock et al.$^{17}$ have computed zero temperature ground state properties of free 2D $^4$He films with Green's Function Monte Carlo (GFMC) and Variational Monte Carlo (VMC).

Even though the Kosterlitz-Thouless (KT)$^{18}$ theory treats superfluidity in two dimensions in a general way, the influence of the substrate on the onset of superfluidity and the superfluid density needs further investigation. In particular, the possible presence of non-superfluid, or inert partial layers of $^4$He at the substrate-adsorbate interface complicates the interpretation of experiment.

An important experimental probe of $^4$He films on substrates is “third sound”, which is a surface wave of the superfluid component of the $^4$He film. The speed of third sound is conventionally derived in the framework of continuum mechanics, assuming a small wave amplitude of the liquid film perpendicular to the substrate$^{19}$. With the aim to narrow the gap between continuum methods of interpretation of experiment and microscopic ab initio calculations we discuss superfluidity and third sound modes, of the $^4$He-H$_2$ system obtained with PIMC in sections VII and VIII. In the process of calculating the third sound speed we obtain excitation energies. These can be compared to the work of Epstein and Krotscheck$^{20}$, Krotscheck and Tymczak$^{21}$, and Clements et al.$^{22}$, who have carried out density functional calculations at zero temperature of the dynamic structure factor of liquid $^4$He films, the chemical potential, and third sound speed.
Recently, Chen et al.\textsuperscript{23} observed in their third sound measurements of $^4$He on H$_2$ surfaces, a new sound mode that coexists in some region of temperature and $^4$He thickness with the conventional third sound signal. This is a striking discovery, because two modes of third sound have never been observed. In spite of experimental and theoretical effort, it is at this point not known what this new phase of sound mode coexistence is. Zhang\textsuperscript{24} proposed the existence of a lattice of vortex-anti-vortex pairs to explain this new sound mode but we see no evidence in support of that idea, as we shall mention when we compute the vorticity-vorticity correlation function in section IX.

II. METHODS

The simulations described in this paper use semi-empirical pair potentials\textsuperscript{25,8,26} for the interactions between the atoms and molecules $^4$He-$^4$He, $^4$He-H$_2$, and H$_2$-H$_2$, and are shown in Fig. (1). The interaction between $^4$He atoms is represented by the semiempirical HFDHE2 potential of Aziz et al., considered to have an uncertainty of 0.15K per atom. The ‘Aziz’ potential is the de facto standard for the $^4$He-$^4$He interaction in many-body computations, and has been used, for example, in Green’s Function Monte Carlo (GFMC) and Variational Monte Carlo (VMC) studies by Whitlock et al.\textsuperscript{17}, and in density functional calculations by Epstein and Krotscheck\textsuperscript{20}, Krotscheck and Tymczak\textsuperscript{21}, and Clements et al.\textsuperscript{22}. In PIMC, the HFDHE2 potential was successfully employed in various calculations of $^4$He properties at finite temperatures both above and below the lambda transition in three dimensions, such as bulk superfluid density\textsuperscript{27}, energies, heat capacities, and condensate fraction\textsuperscript{28}, as well as energies and the superfluid density in two dimensions\textsuperscript{29}.

We choose the semiempirical potential from Silvera and Goldmann\textsuperscript{25} to represent the
interaction between two H\textsubscript{2} molecules because it seems to be the most reliable pair potential in the regime of low pressure and temperature, and has already been used successfully in PIMC simulations of H\textsubscript{2} clusters by Sindzingre et al.\textsuperscript{30} and, more recently, by Scharf et al.\textsuperscript{31}. More details are in the companion paper\textsuperscript{14}.

We describe \(^4\text{He}-\text{H}_2\) interaction by the \textit{ab initio} pair potential from Meyer et al.\textsuperscript{8}, as discussed by Pierre et al.\textsuperscript{7}. Its uncertainty in the well region amounts to about 0.3K out of 13.9K. According to Meyer et al.\textsuperscript{8}, the next most reliable pair potential would be that obtained from a fit to experimental data carried out by Shafer and Gordon\textsuperscript{32}, which has already a well depth of 15.4K. Given that the best available \(^4\text{He}-\text{H}_2\) pair potentials differ by some 10\% in the well region, it should be kept in mind that these uncertainties will carry over into our own calculations as systematic errors. The errors we state later are the statistical errors of the Monte Carlo simulation only.

Not in our calculations, but only here, for the sake of comparison of the above pair potentials with those used in other investigations, we cast the substrate potential felt by a single adatom in the form adopted by Cheng et al.\textsuperscript{15}:

\[
V(z) = \frac{4 C_3^3}{27 D^2} z^9 - \frac{C_3}{z^3},
\]

where the substrate has been assumed to homogeneously occupy the semi-infinite space \(z < 0\). We note that the well depth between the substrate surface and the adsorbed particle may change when the surface corrugations parallel to the interface are averaged over. A substrate-adsorbate potential generated by integrating out the surface potential corrugations will underestimate the binding energy of \(^4\text{He}\) atoms to the substrate for submonolayer coverages, because the less favorable “on-top” surface sites would be weighted the same as the deeper binding “in-between” sites, centered above the middle of a triangle of H\textsubscript{2} molecules,
and there are twice as many “in-between” than there are “on-top” sites. This will turn out to
be relevant at least for the first adsorbed $^4$He layer, where too weak binding energies of $^4$He
to the H$_2$ substrate would be obtained. However, the outward relaxation of the H$_2$ surface,
as discussed in section IV, leads to a lower H$_2$ density near substrate-adsorbate interface,
which will partly compensate this effect for low $^4$He coverages. The numerical values for the
three possible combinations of particles and adsorbates are given in Table 1. The explanation
for the discrepancy by a factor of about 4.5 in the experimental values of the asymptotic
attraction of a $^4$He atom to a H$_2$ surface is so far open, and hard to understand theoretically.

Our PIMC simulation is carried out in a box with periodic boundary conditions in the
$x$-$y$ plane, which is parallel to the $^4$He-H$_2$ interface. Both $^4$He atoms and H$_2$ molecules are
treated as spherical particles and moved by the PIMC program. The interaction between all
particles is given by the pair potentials described above. For details of the PIMC formalism,
see Ref. [13].

The method we used to calculate the long range potential contributions in an “extern-
al potential” from both, the adsorbate and bulk substrate was described in a companion paper$^{14}$. The general idea is to use Gaussian density profiles to represent the layered
substrate-adsorbate system’s density outside the simulation box. These density profiles can
alternatively be taken from experiment, or computed self consistently with PIMC. Fig. (2)
shows the profiles and their Gaussian approximations for three different $^4$He films at T=0.5K
adsorbed on a H$_2$ surface, obtained from the PIMC simulation.

Let us introduce some terminology, which we will use throughout the remainder of the
paper. By an explicit layer, we mean one where the particles’ coordinates are explicitly
represented. An explicit layer can either be dynamic, in which case the particles’ paths are
dynamically sampled or frozen, in which case the coordinates of the path are unchanged
from the beginning of the calculation to the end. In contrast to explicit, in a model layer, only the mean density of its atoms interacts with the explicit particles. We use the model layers to represent all of the particles deep inside the bulk that we do not wish to explicitly carry along in the simulation. They simply contribute to an external potential which the explicit layers see. Further details are given in the companion paper\textsuperscript{14}.

Since the H\textsubscript{2} molecules get moved by the PIMC, we have calculated the surface relaxation and the effect of the \(^4\text{He}\) adsorbate on the zero point motion of the H\textsubscript{2}. However, since the total fluctuation of any observable is proportional to the square root of the total number of particles, in the regime of low \(^4\text{He}\) coverages, the error of the energy will be dominated by the energy fluctuations of the substrate. Hence, we choose three different setups to obtain a more precise value.

\textit{Substrate I} consists of four explicit dynamic layers of 16 H\textsubscript{2} molecules. The H\textsubscript{2}-\(^4\text{He}\) interface has twice this simulation cell’s surface area of 199 \(\text{Å}^2\), because we adsorb \(^4\text{He}\) on both sides of the hydrogen slab. The external potential is constructed such that the adlayer and underlying surface layer see “bulk substrate” in the form of model layers underneath. The external potential has a mirror symmetry w.r.t. the middle of the four H\textsubscript{2} layer planes.

\textit{Substrate II} consists of two frozen layers of H\textsubscript{2} plus underlying model layers. We can reduce the substrate induced total energy fluctuations by \textit{freezing} the substrate. We take an equilibrated H\textsubscript{2} configuration of \textit{substrate I} from a simulation with \(^4\text{He}\) coverage of 0.060\(\text{Å}^{-2}\), or 3/4 monolayers. This finite \(^4\text{He}\) coverage was chosen for freezing to incorporate some of the substrate compression from the adsorption. The approximation hereby lies in neglecting the change of the substrate response to different \(^4\text{He}\) coverages.

\textit{Substrate III} consists of one frozen and one dynamic layer of 30 H\textsubscript{2} molecules each. We used this to simulate the submonolayer \(^4\text{He}\) coverage region more accurately and in
finer coverage increments. Thus, we have to simulate only a relatively small number of H$_2$ molecules for the dynamic substrate, but incorporate correlations between $^4$He atoms and H$_2$ molecules. *Substrate III* is the same H$_2$ substrate used for studies of partial H$_2$ layer adsorption in Ref. [14].

III. BINDING ENERGY

Now, we can commence to investigate the adsorption of thin $^4$He films on a H$_2$ surface. In this section, we focus on the binding energy $e_{\text{bind}}$ per $^4$He atom to the H$_2$ substrate depending on the $^4$He coverage $\sigma$, i.e. the number of $^4$He atoms per area. This is a surprisingly hard problem because both $^4$He and H$_2$ are significantly delocalized due to their small mass and weak interaction, and quantum exchanges are important even for H$_2$. One can not think of $^4$He atoms as localized over a specific H$_2$ surface site, because the binding energy of a $^4$He atom to a H$_2$ molecule hardly exceeds that between two $^4$He atoms, see Fig. (1). Instead, the $^4$He atoms will move over the surface, preferentially avoiding the weaker binding sites directly on top of H$_2$ molecules, as we shall see below.

The binding energies of $^4$He to the H$_2$ surface are of particular interest for several reasons. According to Cheng *et al.*, the substrate potential parameters for well depth and long range attraction stated in the previous section place the "$^4$He on H$_2$ system" on the curve separating the substrates where at least the first layer of adsorbed $^4$He atoms crystallizes from those where the first layer stays liquid. Since we also compute below whether the $^4$He remains fluid, the location of that curve is thus tested. Another motivation of computing $e_{\text{bind}}$ is that the "$^4$He-H$_2$ system" has been predicted by Cheng *et al.* to undergo a prewetting transition. The signature of this would be a minimum in the $e_{\text{bind}}$ for a finite $^4$He coverage.
Fig. (3) shows the binding energy from substrates II and III. The results were consistently above those calculated using substrate I (not shown) which was expected because allowing the paths of H$_2$ to equilibrate will lower the energy. However, the binding energies obtained with substrate II come out too low (overbinding) for $^4$He coverages below about $\sigma = 0.040\,\text{Å}^{-2}$ (1/2 monolayers), because the substrate compression due to the adsorbate under which it was frozen is larger than compression the smaller $^4$He coverage would cause. Consequently, the smaller $^4$He coverage sees too high a H$_2$ density near the $^4$He-H$_2$ interface, making the effective substrate-adsorbate potential too strong. Therefore, we use the frozen substrate II for computation of binding energies for $^4$He coverages exceeding a monolayer only, where the exact response of the H$_2$ molecules at the $^4$He-H$_2$ is less relevant.

Plotted in Fig. (5) are the density profiles of two frozen H$_2$ layers of substrate II overlayed with the profiles of adsorbed $^4$He layers from simulations with $^4$He coverages from 0.020\,Å$^{-2}$ to 0.221\,Å$^{-2}$ in intervals of 0.020\,Å$^{-2}$ as well as coverages of 0.01\,Å$^{-2}$ and 0.03\,Å$^{-2}$. The continuing compression of a $^4$He layer after having reached its saturation density, even after the formation of a new layer, is clearly visible.

The line in Fig. (3) is a fit to the asymptotic behavior of the binding energy from substrate II for $\sigma \geq 0.100\,\text{Å}^{-2}$:

$$e_{\text{bind}}(\sigma) = -7 + \frac{\gamma}{\sigma} + \frac{c}{\sigma^3},$$

(2)

where $-7K$ is the bulk chemical potential, and $\gamma = -0.774K\,\text{Å}^{-2}$ is the surface energy of the $^4$He-vacuum interface and the $^4$He-H$_2$ interface, with the dynamic relaxation of H$_2$ not accounted for. $\gamma$ is negative, because the H$_2$ substrate is more attractive to an adsorbed $^4$He atom than bulk $^4$He. The long range van der Waals interaction between $^4$He and H$_2$ is given by:
\[ c = \frac{1}{2} \rho_{He}^3 \left[ C_3^{H_2-He} - C_3^{He-He} \right] = 1.365 \times 10^{-3} K \AA^{-6} . \] (3)

The fit according to Eq. (2) is shown as the continuous line in Fig. (3).

Simulations with substrate III shown in Fig. (3) indicate that for coverages less than \( \sigma_{min} = 0.046 \AA^{-2} \) the binding energy becomes more positive again, but the single particle limit can not readily be determined this way, due to the statistical error, which is dominated by the substrate for low \(^4\)He coverages, as mentioned above. Using a capillary-rise technique Paine and Seider\(^3\) measured a binding energy of \(^4\)He atoms to liquid molecular hydrogen of \(16 \pm 2\)K. Pierre et al.\(^7\) calculated a binding energy of 15.5K for a single adatom at T=0K. They accounted for the surface corrugation and zero point motion taking both as identical to the bulk values, but not for any surface relaxation or effects of the adsorbate atom on the surface. Since Pierre et al.\(^7\) used the same \(^4\)He-H\(_2\) pair potential we did, one might attribute the binding energy difference to surface relaxation perpendicular to the interface and to zero point motion of the surface.

In (1,1,1) direction an fcc crystal has A-B-C layer stacking, shown in Fig. (6). In the notation of Pierre et al.\(^7\) the adsorption sites “on top” of a H\(_2\) molecule are called A-sites, the sites above the center of a triangle spanned by three nearest neighbor H\(_2\) molecules in the top H\(_2\) layer with a H\(_2\) molecule in the second layer underneath it are named B-sites, and the sites above a H\(_2\) molecule in the third layer are C-sites.

We agree with Pierre et al.\(^7\) in that the helium adsorbs preferentially on B-sites and C-sites on the H\(_2\) surface. Fig. (7) shows the average \(^4\)He density on substrate III for an average \(^4\)He coverage of 0.0804 \AA\(^{-2}\) which coincides with the 2D H\(_2\) density per layer in this direction. Higher darkness corresponds to higher \(^4\)He density, so that the H\(_2\) molecules defining the A-sites are easily recognized as the white areas. Two primitive surface cells are displayed
to facilitate recognition of the periodicity. The grey shading, however, overemphasizes the
$^4$He density variation. The B-sites are preferred for adsorption by a factor of 1.7 over the
A-sites, while the B-sites have a $^4$He coverage exceeding the C-sites only by about 5%. This
is in nice agreement with Pierre et al. who obtain a surface potential well depth of about
23K for an A site, but more than 36K for B and C sites.

Fig. (8) shows the $^4$He coverages on A-, B-, and C-sites depending on the overall $^4$He
coverage. This proves, that at least the top layer of the substrate gives rise to noticable
adsorption potential corrugation. In substrate III the second H$_2$ layer is frozen, and lies
on top of model layers, so that the ratio of B-sites to C-sites coverage is probably not
quantitatively reliable. The linear behavior indicates that the occupation of A-sites is a
single particle property – not caused by $^4$He-$^4$He interactions. A realistic model of the first
monolayer of $^4$He on H$_2$ must include the honeycomb lattice of surface states.

To emphasize the significance of the layer structure and the substrate corrugation in the
$x$-$y$-direction for the binding energy of $^4$He to the H$_2$ substrate, we solved the one-dimensional
Schroedinger equation numerically for a single $^4$He atom adsorbed on a homogeneous, non-
responding, and semi-infinite H$_2$ substrate at bulk density$^{35}$ of 0.026 Å$^{-3}$, obtaining two
bound states of the $^4$He atom, the ground state having a binding energy of $-6.7$K which
is too weak by a factor of 2.5, and one at $-4$K. This result emphasizes that the binding
energy for surface adsorption in the monolayer regime, i.e., in the immediate proximity of
the surface, is dominated by regular crystal surface corrugation.

Prewetting is a phase transition from a 2D gas to a 2D liquid. For small $\sigma$, the $^4$He atoms
would form droplets rather than a homogeneous film on the substrate. This may occur when
the substrate-$^4$He potential is weaker then the $^4$He-$^4$He potential and can thus not spread
the droplets on the surface. At a large enough coverage, the prewetting transition takes
place, joining the droplets into a homogeneous $^4$He film. In our simulation, prewetting must manifest itself in a minimum of $e_{\text{bind}}$ at some non-zero $\sigma_{\text{min}}$. In the case of a pure 2D $^4$He film at T=0K, the existence of such a minimum in the total energy has been demonstrated by Whitlock et al.$^{17}$ with Green’s Function Monte Carlo and Variational Monte Carlo.

With substrate III, we got for a submonolayer $^4$He film on a H$_2$ substrate at T=0.5K a minimum in the binding energy of $e_{\text{bind}} = 16.6 \pm 0.4$K at the coverage 0.046 ± 0.001 $\text{Å}^{-2}$. Whitlock et al.$^{17}$ obtained with GFMC for a free 2D $^4$He film at T=0K a minimum in the binding energy at 0.04356 $\text{Å}^{-2}$. Obviously, their energy minimum is not due to a substrate response. Concurrently, our result shows, that the minimum in the binding energy of the $^4$He atoms is mostly due to the $^4$He-$^4$He interaction, because the presence of the substrate and its honeycomb structure hardly affected the coverage at which it occurs. Therefore, our results confirm the existence of a prewetting jump of a submonolayer of $^4$He. This prewetting transition was predicted by Cheng et al.$^{15}$, who obtained with density functional theory a prewetting jump of 0.033 $\text{Å}^{-2}$ $^4$He, but are limited to zero temperature, and used for simplicity a non-responding homogeneous H$_2$ substrate. However, the agreement is reasonable, given the different temperatures and substrate models.

The chemical potential is given in terms of the free energy by:

$$\mu(\sigma) = f(\sigma) + \sigma \frac{\partial f(\sigma)}{\partial \sigma}.$$  \hspace{1cm} (4)

At low enough temperatures $f = e$ (the energy/particle). Fig. (4) shows the chemical potential obtained for the frozen substrate II. The minimum of $\mu$ at $\sigma = 0.125 \text{Å}^{-2}$ coincides approximately with half filling of the second layer. This agrees the prediction of “growth instabilities” of liquid $^4$He films at zero temperature on weakly attractive model substrates by Clements et al.$^{22}$, because from density plots in the $x$-$y$ plane we see droplet formation
for coverages up to about half filling of the second $^4$He layer. However, this effect is less pronounced than in the first layer. In their density functional theory calculations a growth instability is a region where the chemical potential becomes more negative with increasing $^4$He coverage. They found one such growth instability on a model substrate centered at $0.035\text{Å}^{-2}$, and another extending from $0.13\text{Å}^{-2}$ to $0.165\text{Å}^{-2}$. These growth instabilities indicate phase separations, corresponding to a wetting transition in the case of the first helium layer. For the second layer, this means that at coverages where $\mu$ has a negative slope, droplets of $^4$He form on a completed liquid first layer of $^4$He, i.e. the second layer phase separates.

**IV. DENSITY PROFILES**

The density profiles of $^4$He films adsorbed on substrate III are given in Fig. (9), which also shows the dynamic H$_2$ surface layer getting slightly compressed due to the $^4$He adsorption. This compression is small, because the dynamic H$_2$ layers is sitting on a frozen layer. Here, we can see the formation of a shoulder in the first $^4$He layer already for a coverage of $0.072\text{Å}^{-2}$.

Fig. (10) shows the mean square displacement of the H$_2$ molecules in the top layer (at the $^4$He-H$_2$ interface) and in the second layer as a function of $^4$He coverage at $T=0.5\text{K}$, which will be discussed in more detail below, in the context of $^4$He adsorption. In the case of the free H$_2$ surface, $r^2_{\text{rms}}$ is more than twice as large as the bulk value, indicated by the middle arrow, while the $z$-component is four times its bulk value. The lower arrow is the experimental bulk value at zero temperature$^{35}$. Obviously, at least the two H$_2$ layers nearest to the free surface are very different from bulk layers in that the molecules have much more
zero point motion. The components of $r_{rms}^2$ perpendicular to the H$_2$ surface are shown with dashed curves in Fig. (10). As one might expect, the fluctuation perpendicular to the free surface is significantly larger than parallel to it.

The top arrow shows the $z_{rms}$ squared value for a $^4$He atom in the zero coverage limit obtained by Annett et al.$^6$ using a continuum model for the substrate. Our results are given by the dashed rising curve in Fig. (10). Contrary to the hydrogen zero point motion, the $z_{rms}$ squared of the $^4$He atoms increases with $\sigma$, because they have to spend a higher fraction of the time on less favorable adsorption sites, due to interatomic repulsion.

The lowest two curves of Fig. (10) are the components parallel to the H$_2$ surface of $r_{rms}^2$ of the first and the second layer of H$_2$ upon $^4$He adsorption vs. $^4$He coverage. For the top layer of H$_2$, the compression effect of the adsorbed helium is visible, even for the zero point motion parallel to the $^4$He-H$_2$ interface. The next two curves are the square of $z_{rms}$ of the second and first layer of H$_2$. The reduction of the H$_2$ zero point motion perpendicular to the adsorption interface due to He adsorption is significant even in the second H$_2$ layer.

All values for the square displacement were obtained with substrate $I$, the corresponding density profiles for the various $^4$He coverage are overlayed in Fig. (11). In substrate $I$, which consists of four dynamic H$_2$ layers, the stabilization against buckling is achieved by the external potential alone - there are no frozen layers in that substrate. Because we adsorb $^4$He on both sides of these four hydrogen layers, substrate $I$ consists effectively of two H$_2$ top layers, one at each $^4$He-H$_2$ interface, and two neighboring “H$_2$ second layers”. Therefore, these “second layers” have another “second” layer as their neighbor in bulk direction, favoring large zero point motion. Consequently, the zero point motion of the second H$_2$ layer shown in Fig. (10) is possibly too large. The frozen layer of substrate $III$ inhibits the zero point motion perpendicular to it in an adjacent dynamic layer. This difference in relaxation of
the hydrogen layers is obvious when comparing Fig. (11) from substrate I with Fig. (9) from substrate III. A top H₂ layer resting on a frozen layer relaxes much less upon ⁴He adsorption than top H₂ layer on another dynamic H₂ layer. The true top H₂ layer relaxation should be between the two extreme cases depicted in Fig. (9) and Fig. (11). This issue is discussed further in the companion paper¹⁴.

The compression of two dynamic hydrogen layers of substrate I upon ⁴He adsorption at T=0.5K is demonstrated in Fig. (11), where the density profiles of five different simulations are overlayed. The two peaks at the low z values are the H₂ showing compression due to adsorption of five different ⁴He films of coverages 0.020Å⁻² to 0.100Å⁻² in increments of 0.020Å⁻². The ⁴He film of 0.100Å⁻² has formed a second layer.

Fig. (12) shows the center of mass distance between the top layer and second layer of H₂, and that between the second and third H₂ layer vs. ⁴He coverage at T=0.5K. The zero temperature bulk layer separation in (1,1,1) direction is 3.09Å. The layer H₂ layer spacing in (1,1,1) direction is at the free surface 3.48Å, compared to 3.09Å in bulk. This causes a lower H₂ volume density near the free surface compared to deep inside.

V. THE EFFECTIVE MASS OF A PARTICLE

The mass, m*, quantifies how freely a particle can move. In the case of ⁴He films, the ⁴He motion can be impeded by substrate potential corrugations or by ⁴He-⁴He interactions. We estimate this using imaginary time correlation functions. Define the intermediate scattering function in imaginary time:

\[ F_\rho(t) = \frac{1}{NZ} Tr \left[ \rho_\rho e^{-tH} \rho_{-\rho} e^{-(\beta-t)H} \right] , \]  

where Z is the partition function, and the Fourier transform of the particle density is:

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\[ \rho_\vec{k} = \sum_{j=1}^{N} e^{i \vec{k} \cdot \vec{r}_j}. \]  

(6)

Let \( \beta \to \infty \) and \( t \to \infty \). At low temperature, only one state will contribute to the sum:

\[ F_\vec{k}(t) \to N^{-1} \left| \left< 0 \mid \rho_{-\vec{k}} \mid \vec{k} \right> \right|^2 e^{-t E_\vec{k}}. \]  

(7)

Now expand Eq. (5) in terms of particle coordinates:

\[ F_\vec{k}(t) = \sum_{j=1}^{N} \left< e^{i \vec{k} \cdot (\vec{r}_j(t) - \vec{r}_i(0))} \right>. \]  

(8)

At low coverages only \( j = 1 \) will contribute, and let \( \vec{\Delta}(t) = \vec{r}(t) - \vec{r}(0) \).

\[ F_\vec{k}(t) = \left< e^{i \vec{k} \cdot \vec{\Delta}(t)} \right> \]  

(9)

Assuming \( \vec{\Delta}(t) \) is a Gaussian variate (true always at low \( \vec{k} \)):

\[ F_\vec{k}(t) = e^{-\frac{1}{2D} \lambda k^2 \langle \Delta^2(t) \rangle} \]  

(10)

But the effective mass is defined in terms of the “band energy”

\[ E_\vec{k} = \left( \frac{m}{m^*} \right) \lambda k^2, \text{ as } k \to 0 \]  

(11)

with

\[ \lambda \equiv \frac{\hbar^2}{2m}. \]  

(12)

Equating terms in the exponents of Eq. (7) and Eq. (10) (and generalizing to finite temperature) gives:

\[ \frac{m}{m^*(t)} = \frac{\beta \langle \Delta^2(t) \rangle}{2D \lambda (\beta - t)}. \]  

(13)

Fig. (13) shows the inverse effective mass obtained with Eq. (13) for three \(^4\)He coverages vs. imaginary time at \( T=0.5 \)K. Fig. (14) shows our results for \( m/m^* \) of \(^4\)He atoms on the \( \text{H}_2 \)
surface at $T=0.5K$ using eq. (13) as a function of $^4\text{He}$ coverage for $0.020\,\text{Å}^{-2} \leq \sigma \leq 0.221\,\text{Å}^{-2}$ on the frozen substrate II, and $0.003\,\text{Å}^{-2} \leq \sigma \leq 0.097\,\text{Å}^{-2}$ on the dynamic substrate III and evaluated at $t = \beta/2$. $m/m^*$ = 1 corresponds to a free particle, whereas $m/m^*$ = 0 indicates localization. The $^4\text{He}$ atoms can move more easily on the dynamic substrate, witnessed by the smaller $m^*$ there, i.e. the $\text{H}_2$ molecules can, to some extent, get out of the way, facilitating the $^4\text{He}$'s diffusion. The frozen substrate II, on the other hand, can not respond and appears more rigid to the $^4\text{He}$. This effect will be visible in the speed of third sound, calculated in section VIII.

Obviously, a dynamic substrate model is important for the study of $^4\text{He}$ on hydrogen in the submonolayer region, i.e. when the $^4\text{He}$ is in direct contact with the substrate. The other important point is that the effective mass of $^4\text{He}$ atoms moving parallel the adsorption interface grows with the $^4\text{He}$ coverage up to about one monolayer.

VI. STRUCTURE FACTOR AND FIRST $^4\text{HE}$ LAYER LIQUIDITY

We define static structure factor in a layer as

$$S(\vec{k}) \equiv N^{-1} \left \langle \rho_{\vec{k}} \rho_{-\vec{k}} \right \rangle = F_{\vec{k}}(0).$$

(14)

Fig. (15) shows our results for the first $^4\text{He}$ layer at $T=0.5K$ for coverages $0.020\,\text{Å}^{-2}, 0.030\,\text{Å}^{-2}, 0.040\,\text{Å}^{-2},$ and $0.221\,\text{Å}^{-2}$ on substrate II. The first adlayer of helium stays liquid for up to 3 monolayers even on the frozen substrate. The frozen substrate will aid the crystallization of the first $^4\text{He}$ layer on the hydrogen surface, but was always found liquid in our calculations. Therefore, we think $^4\text{He}$ never crystallizes on $\text{H}_2$. The bending upward of $S(k)$ as $k \to 0$ for the lower three $^4\text{He}$ coverages in Fig. (15) indicates droplet formation on the length scale of our simulation box, i.e. we observe liquid-gas coexistence.
VII. SUPERFLUIDITY

Pollock and Ceperley\textsuperscript{27} derived in the PIMC formalism an expression for the effect of moving boundary walls on the many-body density matrix. Changing the reference frame led to a relation between the superfluid fraction $\rho_s/\rho$ and the flow of liquid $^4$He past stationary walls. In PIMC superfluidity corresponds to large ring polymers winding around the simulation cell in periodic boundary conditions. These long polymers come from permuting particles connecting up their paths. The superfluid fraction of $N$ particles in two dimensions can be computed from

$$\frac{\sigma_s}{\sigma} = \frac{m}{\hbar^2} \frac{\langle \vec{W}^2 \rangle}{2N\beta},$$

where $\vec{W}$ is the winding number, which is a multiple of the box length. For further details on the computation of superfluid densities in $2D$ and $3D$, see Refs. [29,27].

Fig. (16) shows our results for the superfluid coverage vs. $^4$He coverage at $T=0.5K$ from substrate II. The arrows show the completion of the first and second $^4$He layer, respectively. Thus, our results indicate the absence of a “dead layer” of helium on a hydrogen substrate. The dashed curve in Fig. (16) shows the hypothetical case of such an inert $^4$He layer. These results are for our system in periodic boundary conditions of $15.2 \text{Å} \times 13.1 \text{Å}$ and have not been scaled to the infinite system. Experience in $2D$ films suggests that scaling is not necessary below $T_c$ in the KT phase transition, but the lowest two points probably scale to zero. The continuous vertical line at $\sigma = 0.026 \text{Å}^{-2}$ shows an estimate of the onset of superfluidity for the infinite system at $T=0.5K$. Note that roughly 20% of the atoms do not participate in the superfluid phase for less than one monolayer. The two coverages bracketing the second layer completion in Fig. (16) exhibit minima in $\sigma_s/\sigma$, consistent with a decreased mobility of the $^4$He atoms in a filled layer. However, the statistical errors are large.
VIII. THIRD SOUND

One of the chief methods for the investigation of superfluid $^4$He films is third sound. One can think third sound or “ripplons” as long wavelength, low amplitude ripples propagating on a shallow liquid at the free interface. The restoring force is the substrate attraction. The oscillations are carried out by the superfluid component of the film, which carries no entropy and is thus at zero temperature. Therefore, one can view third sound alternatively as an entropy wave or a heat wave. In the long wavelength limit, the third sound velocity for a $^4$He film is

$$ c_3 = \left( \frac{\sigma_s \partial \mu}{m \partial \sigma} \right)^{1/2}, \quad (16) $$

where $\mu$ is the chemical potential.

How can we obtain $c_3$ from our ab initio particle simulations? There are two ways. The straightforward substitution of $\sigma_s$ and $\mu$ calculated above into Eq. (16) is possible, but the statistical error, particularly of $\sigma_s$, suggests to take another route. Fortunately, the second alternative to compute the third sound speed in PIMC has already been almost completed in the derivation of the expression for the effective mass in the previous section. The intermediate scattering function $F_k(t)$ defined in Eq. (7) for the decay of excitations in imaginary time was used to derive an expression the effective mass in PIMC. However, for small $k$, $F_k(t)$ also affords an expression for $c_3$ when we associate the energy $E_k$ with a ripplon:

$$ E_k = \hbar k c_3, \quad (17) $$

At low temperatures the intermediate scattering function of Eq. (5) can be expanded in terms of excited states:
\[ F_k(t) = \sum_{j=1}^{J} S_k^{(j)} \left[ e^{-t E_k^{(j)}} + e^{-(\beta - t) E_k^{(j)}} \right], \quad (18) \]

where

\[ S_k^{(j)} = a^{(j)} \left| \langle 0 \mid \rho_{-k} \mid \bar{k} \rangle \right|^2, \quad (19) \]

with \( a^{(j)} \) a positive coefficient. We compute \( F_k(t) \) in imaginary time and fit it to a sum of decaying exponentials where the summation is over modes. We obtained the best \( \chi^2 \) for two or three modes, depending on the \(^4\text{He} \) coverage and on whether the frozen or dynamic substrate was used. The actual variation of \( S_k^{(j)} \) and \( E_k^{(j)} \) was done using the downhill simplex algorithm by Nelder and Mead\(^{37,38} \). Our fitting routine further varied the number of included imaginary time steps to avoid fitting the noise at long \( t \).

Fig. (17) shows for two coverages on the dynamic substrate III the lowest energy mode vs. wave vector. The continuous line is the zero temperature variational upper bound for excitation energies in liquid \(^4\text{He} \) given by the Feynman-Cohen formula:

\[ \epsilon_k \leq \frac{\hbar^2}{2m} \frac{k^2}{S(k)}. \quad (20) \]

Fig. (18) shows the lowest energy mode for the three smallest wave vectors on the frozen substrate II vs. coverage. Fig. (19) shows our results of the speed of third sound at \( T=0.5 \text{K} \) obtained with eq. (17), on substrate II. The arrows indicate completion of the first and second \(^4\text{He} \) layer respectively. Overlayed are recent experimental data (upper curve) from Chen and Mochel\(^{36} \), and results obtained with density functional theory (lower curve) by Cheng et al.\(^{39} \). For comparison with our results, we assumed 3.60 Å for one layer. The experimental determination of the substrate area and thus of the coverages is difficult, due the sample geometry, i.e. compressed powder. This may cause some uncertainty in the location of maxima and minima.
The agreement with the most recent experimental data from Chen and Mochel\textsuperscript{35} is not as close as it was with previous measurements by Shirron and Mochel's\textsuperscript{11}, but still satisfactory, given that our simulation box was on the order of 15 Å wide. Shirron and Mochel's\textsuperscript{11} determined the first two maxima of 79 m/s and 71 m/s, while we calculate 82 and 68 m/s, respectively. Chen and Mochel\textsuperscript{35} obtain now 86 and 79 m/s, shown in the upper curve of Fig. (19). Shirron and Mochel's\textsuperscript{11} first minima are 58 and 56 m/s, whereas we obtain 53 and 58 m/s. Most likely, our second minimum is too high. Chen and Mochel's first two minima are 66 and 56 m/s. The agreement of our with Cheng \textit{et al.}\textsuperscript{39} is better. They calculate 85 and 63 m/s for the first two maxima, and 49 and 45 m/s for their first two minima.

However, we can not easily calculate our statistical error in $c_3$. There are inherent statistical uncertainties in fitting our correlation functions to several exponentials. Also, our calculated $c_3$ could be systematically underestimated, because we used data from the frozen substrate II to obtain $c_3$ to avoid the “noise” from the responding dynamic substrate. In Fig. (14) we found the effective mass of $^4\text{He}$ to be consistently smaller on dynamic substrate than on the frozen one, as discussed above. Thus, especially in the monolayer coverage regime, third sound should propagate faster on the “softer” dynamic substrate. In Fig. (20), we show the third sound speed for the respective two smallest wave vectors on both, the frozen and the dynamic substrate, overlayed with the same experimental\textsuperscript{36} and theoretical\textsuperscript{39} results as in Fig. (19).

Matching our third sound results in Fig. (19) with the pertinent density profiles shown in Fig. (5) indicates clearly that the minima of $c$ coincide with layer completion. Our second maximum lies at half filling of the second $^4\text{He}$ layer. Our first maximum occurs at more than half filling of the first layer, presumably because the superfluid density vanishes for small finite $\sigma$ rather than zero coverage. If the coverage published by Shirron and Mochel\textsuperscript{11} would
be scaled by a factor 1.8 then the location of their third sound minima and maxima are in good agreement with our calculations.

IX. VORTICITY-VORTICITY CORRELATION FUNCTION

The existence of two sound modes found by Chen et al.\textsuperscript{23} is bounded from above by the KT\textsuperscript{18} transition temperature, and from below by a new $T_C$ that also shows KT-like behavior, and about 0.2K below the KT line. It was recently proposed by Zhang\textsuperscript{24} that the new sound mode discovered by Chen et al.\textsuperscript{23} could be a transverse phonon mode in a vortex anti-vortex lattice. To test this, we have computed the wave vector dependent normal fluid fraction $\sigma_n(\vec{k})/\sigma$ from the vorticity-vorticity correlation function $\vec{G}(\vec{r})$ defined in Ref. \textsuperscript{29}, for four different coverages. The vorticity-vorticity correlation function $\vec{G}(\vec{r})$ is the probability of finding a vortex at position $\vec{r}$ given a vortex at the origin, and $\vec{G}(\vec{k})$ of Eq. (22) is the spatial Fourier transform of this, and should show some structure, if there were a vortex lattice. Following Ref. \textsuperscript{29}, we have

$$\sigma_n(\vec{k}) = \beta \left[ Tr(\vec{G}(\vec{k})) - \frac{\vec{k} \cdot \vec{G}(\vec{k}) \cdot \vec{k}}{k^2} \right], \quad (21)$$

with

$$\vec{G}(\vec{k}) = \frac{-N}{2 \lambda A} \left< \vec{\nabla}_k \otimes \vec{\nabla}_k^* \right>, \quad (22)$$

where $A$ is the simulation cell area, and the Fourier transform of the velocity in imaginary time defined as:

$$\vec{\nabla}_k = \frac{1}{\beta} \int_0^\beta dt \frac{1}{N} \sum_{j=1}^N \frac{d\vec{r}_j(t)}{dt} e^{i \vec{k} \cdot \vec{r}_j(t)} . \quad (23)$$

We have observed no structure in $\vec{G}(\vec{k})$, and see thus no indication of a pattern of vortices in our simulation, as shown in Fig. (21).
X. CONCLUSIONS

The first layer of $^4$He on a H$_2$ (1,1,1) surface at T=0.5 K is liquid for all $^4$He coverages we simulated, 0.008 Å$^{-2}$ to 0.221 Å$^{-2}$. The binding energy of $^4$He to the H$_2$ substrate has a minimum of 16.6 ± 0.4K per atom at $\sigma_0 \approx 0.046 \pm 0.002$ Å$^{-2}$. The experimental value for $^4$He on liquid H$_2$ is 16 ± 2K. The existence of this minimum confirms the prediction of a prewetting transition of $^4$He on hydrogen by Cheng et al.$^{15}$ and a suggestion of a similar transition in the second layer. We see no evidence of the vortex – anti vortex phase proposed by Shou-Cheng Zhang$^{24}$ to explain this coexistence of two sound modes. Third sound has been computed from an ab initio particle method in reasonable agreement with experiment. The minima in the third sound speed correspond to $^4$He layer completion, where the mobility of $^4$He atoms is smallest, due to collisions with other $^4$He atoms in the same layer. So far, we can not explain the coexistence of two sound modes in the $^4$He on H$_2$ system, experimentally observed by Chen, Roesler, and Mochel.$^{23}$
XI. ACKNOWLEDGEMENTS

We wish thank Ming-Tang Chen and Jack M. Mochel for stimulating discussions about their experimental finding of two coexisting sound modes in $^4$He on H$_2$, and for the communication of their results prior to publication. This work was done at the National Center for Supercomputing Applications in Urbana, Illinois. The computations were performed using NCSA’s Cray Y-MP and IBM RS6000/530 work stations. We gratefully acknowledge funding through NSF grant No. DMR–91–17822; and the support of the Department of Physics of the University of Illinois at Urbana-Champaign.
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FIGURE CAPTIONS

Fig. 1. Pair potentials [K] vs. interparticle distance [Å] for $^4$He-$^4$He$^{25}$ (upper continuous curve), $^4$He-H$_2^8$ (middle dashed curve), and H$_2$-H$_2^{26}$ (lower continuous curve).

Fig. 2. Density profiles of substrate III perpendicular to the $^4$He-H$_2$ interface at T=0.5K for $^4$He coverages $\sigma = 0.016\text{Å}^{-2}$, $0.043\text{Å}^{-2}$, $0.097\text{Å}^{-2}$ overlayed with the used “explicit” Gaussian layers to represent them.

Fig. 3. Binding energy per $^4$He atom [K] to the H$_2$ surface at T=0.5K vs. $^4$He coverage [Å$^{-2}$]. The circles are the data obtained with the dynamic substrate III, the squares are from the frozen substrate II. The error bars to the squares are smaller than the symbols. The continuous curve through the values from substrate II is the fit to Eq. (2). Open arrows indicate the coverages at completion of the 1st and 2nd $^4$He layer. The upper filled arrow shows the calculated binding energy in the zero coverage limit from Ref. [7]. The lower filled arrow shows the measured binding energy of $^4$He to the surface liquid H$_2$ at T=15K from Ref. [34].

Fig. 4. Free energy (circles) and chemical potential (squares) on the frozen substrate II vs. $^4$He coverage.

Fig. 5. Density profiles perpendicular to the $^4$He-H$_2$ interface at T=0.5K on substrate II. Only the z>0 of the simulated system is shown. The curve with z<8 Å is the density of
two frozen H₂ layers. The other curves show the ⁴He density for coverages from 0.020Å⁻² to 0.221Å⁻² in intervals of 0.020Å⁻².

Fig. 6. fcc-stacking in (1,1,1) direction. A are the surface sites, B and C are in the second and third layer.

Fig. 7. ⁴He density profile in two primitive surface unit cells of substrate III for an average ⁴He coverage of 0.0804Å⁻². The darkest color corresponds to σ = 0.0963Å⁻², and the lightest color to 0.0572Å⁻², coinciding with A-sites.

Fig. 8. ⁴He coverage in Å⁻² vs. average coverage on substrate III on A-sites (empty circles), B-sites (empty squares), and C-sites (filled triangles). The lines between points of like sites for different coverages are drawn to guide the eye.

Fig. 9. Density profiles of ⁴He films adsorbed on substrate III at T=0.5K for ⁴He coverages 0.027Å⁻², and 0.008Å⁻² to 0.097Å⁻² in increments of 0.008Å⁻². All 13 profiles of each, the ⁴He adsorbate, and the dynamic H₂ top layer are overlayed.

Fig. 10. Square displacements of H₂ molecules and ⁴He atoms at the ⁴He-H₂ interface vs. ⁴He coverage at T=0.5K. All except bulk values were obtained with substrate I. All lines are to guide the eye, only. Only the z-components have dashed line styles. “Plus” symbols, rising curve: ⁴He atoms. All other curves H₂ molecules. Empty symbols H₂ top layer,
filled symbols H$_2$ second layer. Circles: 3D square displacement. 1D components of the square displacement: Triangles: component perpendicular to interface. Squares: component parallel to interface. Upper arrow: z-component of single $^4$He adatom at T=0K. Ref. [6]. Middle arrow: bulk H$_2$ molecule at T=0.5K, present work. Lower arrow: bulk H$_2$ molecule at T=0K, experiment. Ref. [35].

Fig. 11. Overlaid density profiles parallel to the $^4$He H$_2$ interface of five simulations, each showing two dynamic H$_2$ layers of substrate I, left two sets of peaks, and the adsorbed $^4$He film at T=0.5 K. The $^4$He coverages are 0.020Å$^{-2}$ to 0.100Å$^{-2}$ in increments of 0.020Å$^{-2}$.

Fig. 12. Distance between the center of mass of H$_2$ layers in (1,1,1) direction vs. $^4$He coverage, obtained from substrate I. The estimate of the standard error is 0.04Å. 3.09Å is the corresponding H$_2$ bulk layer separation. Upper curve (empty circles): distance between top layer and 2nd layer; lower curve (filled squares): distance between 2nd layer and 3rd layer.

Fig. 13. Inverse effective mass obtained with Eq. (13) for three $^4$He coverages on substrate III vs. imaginary time at T=0.5K. By time reversal symmetry, $\frac{m}{m^*}(t)$ is symmetric w.r.t. $t = \beta/2 = 1/ K$. The $^4$He coverages are: circles: 0.0161Å$^{-2}$, triangles: 0.0429Å$^{-2}$, squares: 0.0643Å$^{-2}$.

Fig. 14. Inverse effective $^4$He mass in units of free particle masses vs. $^4$He coverage $\sigma$ in Å$^{-2}$ at T=0.5K. Filled squares: values obtained on (frozen) substrate II; empty circles: values obtained on (dynamic) substrate III.
Fig. 15. Static structure factor of the first $^4\text{He}$ layer on the the $^4\text{He}-\text{H}_2$ interface at $T=0.5K$ on substrate II for four $^4\text{He}$ coverages overlayed: filled triangles: $0.020\text{Å}^{-2}$; empty squares: $0.030\text{Å}^{-2}$; filled circles: $0.040\text{Å}^{-2}$; plus symbols: $0.221\text{Å}^{-2}$.

Fig. 16. Superfluid fraction of $^4\text{He}$ on the frozen $\text{H}_2$ substrate II at $T=0.5K$. The continuous vertical line at $\sigma=0.026\text{Å}^{-2}$ shows the onset of superfluidity for the infinite system. Arrows indicate completion of the 1st and 2nd $^4\text{He}$ layer. The dashed line shows the hypothetical case of one inert layer with all $^4\text{He}$ above it being superfluid.

Fig. 17. The lowest energy mode for two $^4\text{He}$ coverages at $T=0.5K$ on the dynamic substrate III vs. wave vector. Crosses: $\sigma=0.043\text{Å}^{-2}$; squares: $\sigma=0.097\text{Å}^{-2}$. The continuous curve is given by the Feynman-Cohen formula (20).

Fig. 18. The lowest energy mode for the three smallest wave vectors on the frozen substrate II vs. $^4\text{He}$ coverage at $T=0.5K$. Squares: $k=0.414\text{Å}^{-1}$; triangles: $k=0.479\text{Å}^{-1}$; crosses: $k=0.633\text{Å}^{-1}$.

Fig. 19. Speed of third sound [$m/s$] vs. $^4\text{He}$ coverage $\sigma$ [$\text{Å}^{-2}$] at $T=0.5K$ on substrate II. The empty squares (upper curve) represent data from recent measurements by Chen and Mochel $^{36}$, and the plus symbols (lower curve) are results obtained with density functional theory by Cheng et al. $^{39}$. The arrows indicate completion of the first and second $^4\text{He}$ layer.
respectively. The filled circles are our results for the lowest energy mode of the smallest wave vector, \( q=0.414\AA^{-1} \). No error bars are determined.

Fig. 20. Speed of third sound \([m/s]\) vs. \(^4\)He coverage \(\sigma [\AA^{-2}]\) at \(T=0.5K\) on substrate II. The dashed line (upper curve) are experimental data from Chen and Mochel\(^{36}\), and the continuous line (lower curve) are theoretical results from Cheng et al.\(^{39}\). We show our results for the lowest two wave vectors for both, the frozen Substrate II and dynamic Substrate III. Filled circles: frozen substrate, \(q=0.414\AA^{-1}\); filled triangles: frozen substrate, \(q=0.479\AA^{-1}\); empty circles: dynamic substrate, \(q=0.319\AA^{-1}\); empty triangles: dynamic substrate, \(q=0.332\AA^{-1}\).

Fig. 21. \(\sigma_n(\mathbf{k})/\sigma\) from the vorticity correlation function defined Ref. [29] at \(T=0.5K\) for four different coverages on the frozen substrate: circles: 0.024\AA^{-2}; triangles: 0.043\AA^{-2}; squares: 0.064\AA^{-2}; crosses: 0.080\AA^{-2}.
TABLES

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Table I. Parameters of potential between bulk substrate of density $\rho$ and adsorbed particle. $D$ and $C_3$ are our potential depth and van der Waals constant. The following three columns, $C_3$(B), $C_3$(P), and $C_3$(S), show the experimental value of $C_3$ in K×Å³, from Brisson et al.\textsuperscript{12}, Paalanen and Iye\textsuperscript{9}, and from Shirron and Mochel\textsuperscript{11}, where we used 1 ‘layer’ = 3.58 Å for the conversion. The rightmost column, $C_3$(C), is from Cheng et al.\textsuperscript{33}.