From nonwetting to prewetting: the asymptotic behavior of $^4\text{He}$ drops on alkali substrates

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Abstract

We investigate the spreading of $^4\text{He}$ droplets on alkali surfaces at zero temperature, within the frame of Finite Range Density Functional theory. The equilibrium configurations of several $^4\text{He}_N$ clusters and their asymptotic trend with increasing particle number $N$, which can be traced to the wetting behavior of the quantum fluid, are examined for nanoscopic droplets. We discuss the size effects, inferring that the asymptotic properties of large droplets correspond to those of the prewetting film.

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I. INTRODUCTION

Wetting phenomena\(^1\) of alkali and graphite-based surfaces by quantum fluids have been a burgeoning field of research in recent years, both from the experimental and the theoretical viewpoints. Early in the last decade, it was demonstrated that liquid \(^4\)He is not a universal wetting agent,\(^2−8\) since it fails to spread uniformly on cesium surfaces at temperatures below 1.95 K. From the theoretical viewpoint, Cheng et al\(^9\) predicted, in the frame of a nonlocal density functional model, the inability of helium to wet all heavy alkalis. More stringent theoretical studies\(^10−14\) showed that while for the majority of moderately thin to thick helium films on substrates, one or two layers of solid helium would be continuously wetted by the excess liquid, a different pattern was to be expected in the presence of weak adsorbers. In this case, either nonwetting, or wetting preceded by a first order prewetting transition,\(^15\) was most likely to appear. The prewetting transition undergone by \(^4\)He on Cs substrates was first observed by Hallock and coworkers\(^6\) and the complete phase diagram was presented in Ref. 7. The situation is not so clear for Rb surfaces, since although a number of experimental evidences\(^4,16\) and at least one theoretical calculation\(^17\) are consistent with a vanishing wetting temperature, a more recent experiment\(^18\) indicates that pristine Rb is nonwetted by helium up to a temperature above 300 mK, in agreement with earlier predictions.\(^19,20\)

So far, theoretical work on wetting properties of helium relies almost exclusively on the description of structure and energetics of films, uniformly extended on the plane perpendicular to the substrate, in spite that all these systems, even on wetted substrates, are thermodynamically unstable for the lowest areal coverages and should then appear as collections of puddles or clusters upon the adsorbing surface. In a prior work, Ancilotto et al\(^21\) have presented a calculation of density profiles of finite droplets of \(^4\)He on Cs, at zero temperature. These authors solve a nonlinear equation for the density profile, constructed by functional differentiation of a Finite Range Density Functional (FRDF) acknowledged as the Orsay-Trento (OT) density functional.\(^22\) Quite elaborated from the numerical viewpoint, this work develops an accurate theoretical instrument to investigate wetting patterns.
The purpose of this work is to perform a detailed investigation of the spreading of $^4\text{He}$ droplets on alkali surfaces at zero temperature in the framework of the FRDF formalism. Our systematic study permits us to relate the asymptotic trends of their energetics and structure, with increasing size, to the wetting properties of the fluid; in particular, we show that droplets as large as a few thousand $^4\text{He}$ atoms, on the strongest adsorbers, already exhibit the structural and thermodynamic characteristics of the prewetting film at zero temperature.

In Sec. II we shortly review the current FRDF formulation and give essential details of the method applied to solving the associated Euler-Lagrange (EL) equation to obtain the equilibrium configuration. In Sec. III we illustrate the predicted prewetting transition for films on alkali adsorbers, and subsequently, we present the numerical results for clusters. Our conclusions and perspectives are presented in Sec. IV.

II. DENSITY FUNCTIONAL DESCRIPTION OF $^4\text{He}$ CLUSTERS ON ADSORBING SUBSTRATES

The FRDF for $^4\text{He}$ adopted in this work is that of Ref. 23, which was originally developed to describe mixtures of helium isotopes, here restricted to the $^4\text{He}$ component. Its detailed form and values of the coefficients entering its definition have been given in Ref. 24, with the two changes reported in Ref. 23 which consist, on the one hand, in the neglect of the nonlocal gradient correction to the kinetic energy term in Ref. 22, and on the other, in the choice of the suppressed Lennard-Jones core as in Ref. 25, namely

$$V_{LJ}(r) = \begin{cases} 
4\varepsilon \left[ (\sigma_4/r)^{12} - (\sigma_4/r)^{6} \right] & \text{if } r \geq h \\
V_0 r^4 & \text{if } r \leq h ,
\end{cases}$$

with $\varepsilon = 10.22 \text{K}$, $\sigma_4 = 2.556 \text{Å}$, and with hard-core radius $h = 2.359965 \text{Å}$. Notice that $V_0$ is the value of the 6-12 potential at $r = h$. The value of $h$ has been fixed so that the volume integral of the interaction $V_{LJ}$ coincides with the one in Refs. 22, 24. This form of the FRDF - we especially comment that dropping the nonlocal gradient correction present
in the OT density functional\textsuperscript{22} increases by around a 3\% the surface tension of $^4\text{He}$ with respect to the value there reported - has been introduced to render the numerical calculations less cumbersome.

The integrodifferential EL equation arising from functional differentiation of the FRDF is

\[
\left[ -\frac{\hbar^2}{2m_4} \nabla^2 + V(\rho) \right] \Psi(r) \equiv \mathcal{H}_H \Psi(r) = \mu \Psi(r) \tag{2}
\]

with \( \mu \) the chemical potential that enforces particle number \( N \) conservation in the drop, and with \( \Psi = \sqrt{\rho} \), where \( \rho(r) \) is the particle density. The mean field \( V(r) \) in the Hartree Hamiltonian \( \mathcal{H}_H \) includes the substrate potential \( V_s(z) \) chosen to be the appropriate Chizmeshya-Cole-Zaremba (CCZ) potential.\textsuperscript{19} To illustrate the adsorbing power of the alkali surfaces, in Table I we show the energies \( \mathcal{E}_4 \) of one $^4\text{He}$ atom in the substrate, obtained with the CCZ potentials.

In the current geometry, for axially symmetric droplets, we have \( \rho(r) = \rho(r, z) \). The EL equation is discretized using 7-point formulae and solved on a two dimensional \((r, z)\) mesh. We have used a sufficiently large box with spatial steps \( \Delta r = \Delta z \equiv \Delta = \hbar/12 \sim 0.197\text{\AA} \). The stability of our results against the increase of the number of mesh points, as well as with respect to the order of the formulae employed to discretize the partial derivatives, has been checked comparing the solutions for spherical drops with those obtained with a spherically symmetric code. We have employed an imaginary time method to find the forward solution of the imaginary time diffusion equation\textsuperscript{26}

\[
\frac{\partial \Psi}{\partial \tau} = -(\mathcal{H}_H - \mu) \Psi \tag{3}
\]

in other words,

\[
\Psi(\tau + \delta\tau) - \Psi(\tau) \equiv \Delta \Psi(\tau) = -\delta\tau(\mathcal{H}_H - \mu) \Psi(\tau), \tag{4}
\]

where \( \mu = \langle \Psi(\tau) | \mathcal{H}_H | \Psi(\tau) \rangle \).

To accelerate the self-consistent solution of this equation, we use the preconditioning smoothing operation described in Ref. 27. This means that \( \Delta \Psi(\tau) \) has been smoothed as
\[ \Delta \Psi(r, z; \tau) = \frac{1}{2} \{ \Delta \Psi(r, z; \tau) + \]

\[
\frac{1}{4} [\Delta \Psi(r - \Delta r, z; \tau) + \Delta \Psi(r + \Delta r, z; \tau) + \Delta \Psi(r, z - \Delta z; \tau) + \Delta \Psi(r, z + \Delta z; \tau)] \} .
\]

Finally, the performance of the code has been further improved by adding a ‘viscosity term’, i.e., Eq. (4) has been changed into

\[ \Psi(\tau + \delta \tau) = \Psi(\tau) + \Delta \overline{\Psi}(\tau) + \alpha_V [\Psi(\tau) - \Psi(\tau - \delta \tau)] . \]

The heuristic viscosity parameter \( \alpha_V \) is fixed to a value of 0.8.

We recall that as shown in Ref. 26, the maximum \( \tau \)-step, \( \delta \tau_m \), that can be used to produce a stable imaginary time evolution is \( \hbar^2 \delta \tau_m / (2m_4 \Delta^2) \lesssim 1/4 \). The combination of smoothing and viscosity allows one to use large values of \( \delta \tau \), typically up to \( \delta \tau \sim 0.5 \delta \tau_m \).

After every \( \tau \)-step, the \(^4\text{He} \) density is normalized to \( N \). The iteration procedure starts on the halved density of a \(^4\text{He}_2 N \) cluster calculated with the spherically symmetric code. Most of the computational time is spent in the evaluation of the mean field \( V(r, z) \) by folding the helium density with the screened Lennard-Jones potential. For this reason, the mean field is updated only every ten \( \tau \)-iterations.

III. EQUILIBRIUM CONFIGURATIONS

A. Films

The thermodynamic criterium for wetting requests that the surface grandpotential, namely at zero temperature \( \sigma = (E - \mu N) / A \), where \( A \) is the area of the surface, regarded as a function of coverage \( n = N / A \), displays its absolute minimum when \( n \) approaches infinity.\(^{10,13} \) For films of \(^4\text{He} \), and within the density functional frame,\(^{17} \) this criterion can be formulated in terms of the expansion coefficients of the total energy of the film in powers of \( 1/n \). In particular, it has been shown that the OT density functional plus the CCZ potential yields, for \(^4\text{He} \) films on alkali substrates, wetting of \( \text{Rb} \)\(^{17} \) and \( \text{K} \),\(^{28} \) and nonwetting
of a Cs surface. In Figs. 1 and 2, we respectively plot the chemical potential and the surface grandpotential of $^4$He on different alkali surfaces, as predicted by the current FRDF. The energy per particle $e = E/N$ that corresponds to the equilibrium density for a given coverage is also shown in Fig. 1 in dashed lines for Cs, Na and Li.

It is worthwhile to recall here that from the thermodynamic relations

$$\mu = e + n \frac{\partial e}{\partial n}$$

(one can establish the condition $\mu = e$ to localize the prewetting jump. This condition corresponds to vanishing areal grandpotential

$$\frac{\partial e}{\partial n} = \sigma = 0$$

from where the Maxwell construction (8) defines the chemical potential and coverage at the prewetting first order transition. The prewetting jumps are indicated in Fig. 1 as horizontal segments, for the stronger adsorbers Na and Li. Although K is wetted by helium in the current FRDF description (see Fig. 3), the prewetting jump lies too far to the right in the scale here displayed. In the present calculation, $^4$He does not wet Rb and behaves very much like Cs; this can be appreciated in Fig. 3 where the surface grandpotential of the film is plotted as a function of the inverse coverage. Indeed, it has been already pointed out that this substrate represents a delicate limiting case, largely sensitive to the details of the calculation, of the density functional and of the adsorbing potential adopted.$^{17,20,29}$

**B. Clusters**

We concentrate on droplet sizes below $N = 3000$, which illustrate the spreading trend on adsorbers of different strengths and provide a good representation of the asymptotic behavior as shown below. Addressing larger drops requires larger mesh sizes, which is possible at the
obvious price of making the calculations more cumbersome and time consuming. In Fig. 4 we show the contour plots of the particle densities in the \((x, z)\) plane for a cluster with \(N = 1000\), on substrates Cs, K, Na and Li -Rb looks very much as Cs, and for this reason we do not discuss it here- (hereafter, lengths are given in \(\text{Å}\)). The profiles \(\rho(r, z_{\text{min}})\) of these drops, at the minimum \(z_{\text{min}}\) of the CCZ potential, and \(\rho(0, z)\) along the symmetry axis are plotted in Fig. 5 for the same substrates. As we see, increasing attractiveness provokes a change of shape in two complementary manners, which becomes most noticeable for the clusters on Na and Li, i.e., flattening along the vertical coordinate, and sizeable radial spreading. We also visualize a smoothing of the structure, since the oscillations in \(\rho(0, z)\) evolve from practically three peaks on Cs and K, and two on Na, to just one on Li. This feature does reflect the wetting behavior obtained for \(^4\text{He}\) on these alkalis, namely nonwetting for Cs, and prewetting with a jump of above three layers on K, two on Na and one on Li, respectively. In fact, submonolayer wetting occurs for the strongest adsorber Li.\(^{13,14,30}\)

The energetics as a function of cluster size is illustrated in Fig. 6, where we plot the chemical potential of the \(^4\text{He}\) atoms, and the grandpotential per particle \(\omega = E/N - \mu\) as functions of \(N\) for the above alkalis. From this figure, we realize that for wetted adsorbers, there is a tendency -very clear for the strongest substrates Na and Li- to saturate \(\mu\) at a finite value below the bulk value -7.15 K, and \(\omega\) at zero value. In the case of Cs, the limiting chemical potential points towards the bulk figure, in agreement with the nonwetting behavior of \(^4\text{He}\); notice that although \(^4\text{He}\) wets K, as seen in Fig. 3, within the current scale we do not reach values of \(\mu\) lower than -7.15 K, since this crossing takes place at a much larger number of atoms, likely several tens of thousands.

We now select Na as a test case to examine the evolution of the density profiles with particle number; as a reference, in Fig. 7 we depict the contour plots of the particle densities in the \((x, z)\) plane for several values of \(N\) varying between 100 and 3000, where we already observe a rather flat profile with two ridges, corresponding to density oscillations parallel to the substrate. Figure 8 displays \(\rho(r, z_{\text{min}})\) and \(\rho(0, z)\) for the above particle numbers. We appreciate a definite inclination to saturate the vertical density profiles with two shells.
and a limiting height, as well as a tendency to flatten the radial dependence into a wedge-like shape, splashing outwards any extra material. These effects are more pronounced in the case Li, as can be seen from Fig. 9 where the submonolayer profiles become manifest; nevertheless, it should be kept in mind that as mentioned in Ref. 30, DF theory predicts a prewetting jump at a larger coverage than i.e., path-integral Monte Carlo calculations.

These patterns suggest the existence of an asymptotic coverage at the center of large clusters; in fact, as we define

\[ n(r) = 2\pi \int dz \rho(r, z) \]

and plot \( n(0) \) as a function of \( N \) for all substrates under consideration as shown in Fig. 10, the shapes of these curves indicate that for wetted adsorbers, such an asymptotic value exists. Our data indicate that these limiting numbers are \((\mu_{Na}, n_{Na}(0)) = (-8.27 \text{ K}, 0.14 \text{ Å}^{-2})\) and \((\mu_{Li}, n_{Li}(0)) = (-11.15 \text{ K}, 0.06 \text{ Å}^{-2})\). These values coincide with the coordinates of the respective prewetting points shown on the curves for the chemical potentials displayed in Fig. 1.

**IV. SUMMARY AND OUTLOOK**

In this work we have described the spreading of \( ^4 \text{He} \) droplets on alkali substrates as they grow in size. One outcome of this investigation is that for wetted substrates, the large size limit is not the bulk liquid but, interestingly, the minimum stable film. In other words, the deposited cluster can grow towards the thermodynamic limit along the directions permitted by the geometrical constraint, namely those parallel to the planar substrate. The transverse confinement just fixes the maximum height of the sample so as to yield the prewetting coverage.

Along this work we have shown density profiles of clusters on alkali adsorbers, and seen that for wetted substrates, droplets of \(^4\text{He} \) atoms are present in the nonwetting, subspinodal regime -i.e., \( \partial \mu / \partial n < 0 \)- and seem to present a contact angle in spite that, in the light of the
results discussed in the previous section, the asymptotic limit of these large systems is the prewetting film, that is associated to vanishing contact angle. This apparent inconsistency stems from the impossibility to define a contact angle for nanoscopic droplets in which the particle density displays a non-negligible surface width, and is highly stratified near the substrate, which render the contact angle an ill defined quantity. It also illustrates that a determination of the contact angle by ‘visual inspection’ of the equidensity lines is fraught with danger and may lead to either a crude estimate or even to a qualitatively wrong result. As we have commented, Ancilotto et al\textsuperscript{21} presented density profiles of $^4$He droplets on Cs, at zero temperature, for the OT density functional.\textsuperscript{22} In this work, emphasis lay on the determination of the contact angle of macroscopic nonwetting samples of $^4$He on Cs, and a rather complex procedure was developed that permitted to compute this angle extrapolating the results for nanoscopic drops to the macroscopic ones for which a contact angle can be sensibly defined. We want to point out that since the FRDF here employed differs only slightly from the OT -we have verified that no visible discrepancies show up in the structural properties of clusters on Cs- applying the method of Ref. 21 would yield in our case a similar contact angle for $^4$He on Cs (about $36^\circ$, cf. Table I in Ref. 29).

An interesting extension of this work is the investigation of the spreading of mixed $^3$He-$^4$He clusters on planar substrates. Such a project is feasible within the FRDF formalism, and in fact we have recently shown\textsuperscript{31} that one or few $^3$He atoms added to a large -yet nanoscopic- $^4$He drop on Cs, localize on a onedimensional ring around the contact line. This is a new feature of helium mixtures, which may contribute to interpreting the very complex phase diagram of such systems.\textsuperscript{32} A detailed systematics of the structure and energetics of mixed clusters is presently in progress and will be reported elsewhere.

Finally, we would like to mention that the FRDF formalism may also be used to shed light on the nucleation of wetting layers in the case of a first-order wetting transition, very much as it has been successfully used to address nucleation or cavitation in bulk liquid helium.\textsuperscript{33} A major advantage of the formalism is that it may circumvent some of the approximations made in other approaches. In particular, it avoids the use of an experimentally unknown
line tension,\textsuperscript{34} and the use of a separation between surface and line contributions to the free energy,\textsuperscript{35} which becomes doubtful when the radius of the critical droplet or bubble is comparable to its surface width.

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TABLES

TABLE I. Energy $\mathcal{E}_4$ (K) of one $^4\text{He}$ atom on different alkali substrates.

|      | Li  | Na  | K   | Rb  | Cs  |
|------|-----|-----|-----|-----|-----|
| -10.70 | -7.08 | -4.20 | -3.72 | -3.53 |
FIGURES

FIG. 1. Chemical potential $\mu$ (K) of $^4$He atoms in films upon Cs, Rb, K, Na and Li substrates as functions of areal coverage $n$ ($\text{Å}^{-2}$), predicted by the FRDF employed in this work. Also shown is the energy per particle $e$ (K) for Cs, Na, and Li, and the energy per particle in bulk $^4$He $e_B$ as a straight line.

FIG. 2. Same as Fig. 1 for the surface grandpotential.

FIG. 3. Surface grandpotential as a function of inverse coverage. The short horizontal line near $1/n = 0$ represents the value of the surface grandpotential in the limit of infinite coverage.

FIG. 4. Contour plot of the density of a $^4$He$_{1000}$ droplet on Cs, K, Na and Li, from top to bottom. The lowest equidensity lines correspond to $\rho = 10^{-4}\text{Å}^{-3}$, and the highest ones to $2.6 \times 10^{-2}\text{Å}^{-3}$ (Cs and K), and to $3 \times 10^{-2}\text{Å}^{-3}$ (Na and Li).

FIG. 5. Density profiles $\rho(r, z_{\text{min}})$ (upper panel) and $\rho(0, z)$ (lower panel) of a $^4$He$_{1000}$ cluster on different alkali substrates.

FIG. 6. Chemical potential (left panel) and grandpotential per particle (right panel) of $^4$He droplets on alkali adsorbers as a function of $N$. The energy per particle in bulk $^4$He $e_B$ is also shown in the left panel. The lines have been drawn to guide the eye.

FIG. 7. Contour plots of the density of a $^4$He$_N$ droplet on Na for $N = 100, 1000, 2000, 3000$. The lowest equidensity lines correspond to $\rho = 10^{-4}\text{Å}^{-3}$, and the highest ones to $3 \times 10^{-2}\text{Å}^{-3}$.

FIG. 8. Density profiles $\rho(r, z_{\text{min}})$ (upper panel) and $\rho(0, z)$ (lower panel) of $^4$He$_N$ clusters on Na.

FIG. 9. Contour plots of the density of a $^4$He$_N$ droplet on Li for $N = 100, 1000, 2000, 3000$. The lowest equidensity lines correspond to $\rho = 10^{-4}\text{Å}^{-3}$, and the highest ones to $2.5 \times 10^{-2}\text{Å}^{-3}$. 
FIG. 10. Coverage $n(r = 0)$ for $^4\text{He}_N$ droplets on the various alkali substrates as a function of atom number. The lines have been drawn to guide the eye.
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