Finite size effects in adsorption of helium mixtures by alkali substrates

M. Barranco, M. Guilleumas, E.S. Hernández, R. Mayol, M. Pi, and L. Szybisz

1Departament ECM, Facultat de Física, Universitat de Barcelona, E-08028 Barcelona, Spain
2Departamento de Física, Facultad de Ciencias Exactas y Naturales, Universidad de Buenos Aires, 1428 Buenos Aires, and Consejo Nacional de Investigaciones Científicas y Técnicas, Argentina
3Departamento de Física, CAC, Comisión Nacional de Energía Atómica, 1429 Buenos Aires, Argentina

(October 29, 2018)

Abstract

We investigate the behavior of mixed $^3\text{He}-^4\text{He}$ droplets on alkali surfaces at zero temperature, within the frame of Finite Range Density Functional theory. The properties of one single $^3\text{He}$ atom on $^4\text{He}_N$ droplets on different alkali surfaces are addressed, and the energetics and structure of $^4\text{He}_N + ^3\text{He}_N$ systems on Cs surfaces, for nanoscopic $^4\text{He}$ drops, are analyzed through the solutions of the mean field equations for varying number $N_3$ of $^3\text{He}$ atoms. We discuss the size effects on the single particle spectrum of $^3\text{He}$ atoms and on the shapes of both helium distributions.

PACS 67.60.-g, 67.70.+n,61.46.+w
I. INTRODUCTION

The physics of wetting by quantum fluids, particularly $^4$He, $^3$He and isotopic mixtures, received considerable attention from both experimentalists and theoreticians along the last decade, with experiments mostly pointing at measuring adsorption isotherms, determining the interfacial surface tensions, establishing the wetting temperature and constructing phase diagrams for the mixed systems. A geometrical, visible parameter that characterizes the wetting behavior of a liquid-substrate combination at temperature T is the contact angle of a macroscopic drop or wedge on a planar surface. The contact angle of liquid $^4$He on Cs has been measured by at least three different groups, and although the reported values do not coincide—a result which among other effects, could be attributed to the preparation of the surface—they are fully consistent with the helium inability to wet this weak adsorber. By contrast, $^3$He has been theoretically shown to be a universal wetting agent, a prediction which was verified shortly after, as isotherms of $^3$He on Cs were measured that display neat prewetting jumps down to temperatures around 0.2 K.

It is well-known that $^3$He atoms admixed into bulk liquid $^4$He or $^4$He films at low T’s populate a twodimensional (2D) homogeneous layer of Andreev states on the free surface (see also Ref. 9 and Refs. therein for a recent review). In this case, the Andreev state originates in the presence of a broad surface at the $^4$He liquid-vapor interface and in the mutual interaction between the isotopes, and corresponds to the ground state (gs) of a discrete spectrum of states which become progressively localized towards the interior of the film. This structure appears as well in density profiles of mixed clusters. It was earlier proposed that if $^4$He lies on a weak adsorber, Andreev-like states could be expected at the liquid-substrate interface. Theoretical anticipations of the wetting behavior of mixtures rely heavily on the expected reduction of the liquid-vapor surface tension of the mixture with respect to that of pure $^4$He, due to the Fermi pressure of the Andreev surface layer. This effect, actually observed in Ref. 15, reduces the contact angle at a given temperature, thus permitting lower wetting temperatures as the $^3$He concentration increases. On these grounds, Pettersen and Saam predicted reentrant wetting of helium mixtures on Cs, a phenomenon measured shortly after by Ketola et al. The complete phase diagram for wetting of helium mixtures on alkali metal substrates derived in Ref. 17 offers a wide gallery of phenomena which includes prewetting, isotopic separation, triple point dewetting and $\lambda$-transitions in the solution, in the full range of $^3$He concentrations. Subsequent experimental work discovered a rich pattern of wetting by mixtures, which includes, i.e., dewetting transitions near coexistence. More recently, detailed measurements of the contact angle for dilute helium mixtures have been reported; an analysis of the data showed that the large values of these angles are consistent with the presence of single-particle (sp) states of the $^3$He atoms, together with ripplons, at the liquid-solid interface.

The first calculation of density profiles of finite droplets of $^4$He on Cs, at zero temperature, was forwarded by Ancilotto et al. within the framework of Finite Range Density Functional Theory (FRDF), using the FRDF of Ref. 21. More recently, we have presented calculations of nanoscale mixed $^3$He-$^4$He droplets on Cs, (hereafter, referred to as I) using a previously derived FRDF for mixtures. An important prediction in this paper is the existence of edge states, the lowest-lying $^3$He sp bound states, which are essentially onedimensional (1D) and localize around the contact line of the $^4$He drop—in contrast to the 2D surface Andreev
The purpose of this work is to examine in more detail the behavior of mixed helium droplets on alkali surfaces, at T=0. For this sake, in Sec. II we shortly review the current FRDF formulation, and in Sec. III we analyze the energetics and structure of a single $^3$He atom on $^4$He$_N$ droplets on alkali planar surfaces, for varying number $N$ of $^4$He atoms. In Sec. IV we discuss the solutions of the mean field equations for mixed $^4$He$_N$+$^3$He$_N$ drops on Cs with varying number $N$ of $^3$He atoms. Emphasis here lies on the features of the spectrum of $^3$He atoms predicted by the solution of the Kohn-Sham (KS) equations derived from the FRDF, as well as on the shapes of both helium distributions. Our conclusions and perspectives are summarized in Sec. V.

II. DENSITY FUNCTIONAL DESCRIPTION OF PURE AND MIXED HELIUM CLUSTERS ON ADSORBING SUBSTRATES

The FRDF for helium mixtures adopted in this work is the same as in Ref. 23, of the general form

$$E = \int d\mathbf{r} \left[ U_3(\rho_3, \tau_3, \rho_4) + U_4(\rho_4, \tau_4) + U_{34}(\rho_3, \rho_4) \right],$$

where $\rho_i(\mathbf{r})$, $\tau_i(\mathbf{r})$ are respectively the particle and the kinetic energy densities for $i = 3, 4$. The detailed form of the FRDF and values of the force coefficients have been given in Ref. 10, with the only 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. 21, and on the other hand, in the choice of the suppressed Lennard-Jones core as proposed earlier. More specifically, for $^3$He-$^3$He, $^3$He-$^4$He and $^4$He-$^4$He interactions, the Lennard-Jones screened interaction is written as

$$V_{LJ}(r) = \begin{cases} 
4\epsilon_{ii} \left[ \left( \frac{\sigma_{ii}}{r} \right)^{12} - \left( \frac{\sigma_{ii}}{r} \right)^6 \right] & \text{if } r \geq h_i \\
V_0 \left( \frac{r}{h_i} \right)^4 & \text{if } r \leq h_i,
\end{cases}$$

with $\epsilon_{44} = \epsilon_{33} = \epsilon_{34} = 10.22$ K, $\sigma_{44} = \sigma_{33} = 2.556$ Å, $\sigma_{34} = 2.580$ Å, hard-core radii $h_4=2.359665$ Å, $h_3 = 2.356415$ Å and $h_{34} = 2.374775$ Å, and with $V_0$ the value of the corresponding 6-12 potential at $r = h_i$. These values have been fixed so that the volume integrals of the interactions $V_{LJ}$ coincide with the original ones in Refs. 10, 21.

In the current geometry, for axially symmetric droplets, we have:

$$\rho_3(r, z) = 2 \sum_{nl} |\Psi_{nl}(r, z)|^2,$$

$$\tau_3(r, z) = 2 \sum_{nl} |\nabla \Psi_{nl}(r, z)|^2,$$

$$\tau_4(r, z) = \frac{1}{4} \left| \nabla \rho_4(r, z) \right|^2,$$

where

$$\Psi_{nl}(r, z) = \psi_{nl}(r, z) \frac{e^{il\varphi}}{\sqrt{2\pi}},$$

and

$$\psi_{nl}(r, z) = \psi_{nl}(r, z) \frac{e^{il\varphi}}{\sqrt{2\pi}}.$$
denotes the sp wave functions (wf’s) of the $^3$He atoms, and $l$ denotes the projection of the
sp orbital angular momentum on the $z$ axis. Since we shall only address spin saturated
systems, the sp energy levels are either twofold ($l = 0$) or fourfold ($l \neq 0$) degenerate.

The Euler-Lagrange (EL) equations arising from functional differentiation of the density
functional in Eq. (1) give rise to an integrodifferential coupled set

$$
\left[-\frac{\hbar^2}{2m_4} \nabla^2 + V_4 (\rho_3, \tau_3, \rho_4)\right] \sqrt{\rho_4} = \mu_4 \sqrt{\rho_4} \tag{7}
$$

$$
-\nabla \left(\frac{\hbar^2}{2m_3} \nabla \Psi_{nl}\right) + V_3 (\rho_3, \tau_3, \rho_4) \Psi_{nl} = \varepsilon_{nl} \Psi_{nl} \tag{8}
$$

In Eq. (7), $\mu_4$ is the chemical potential that enforces particle number conservation in the
$^4$He drop, and expression (8) represents the Kohn-Sham (KS) equations for $^3$He atoms in
the presence of their mutual interaction, their coupling to the $^4$He cluster and the potential
$V_s(z)$ generated by the alkali substrate filling the the $z \leq 0$ half-space. For $z \geq 0$, the mean
fields $V_i(r, z)$ include $V_i(z)$ chosen as the Chizmeshya-Cole-Zaremba (CCZ) potentials.\textsuperscript{25} The
energies $\mathcal{E}_i$, $i=3, 4$, of one helium atom in the substrate, obtained with the CCZ potentials,
are shown in Table I.

Eqs. (7) and (8) have been discretized using 7-point formulae and solved on a 2D ($r, z$)
mesh. We have used sufficiently large boxes with spatial steps $\Delta r = \Delta z = h_4/12 \sim 0.197\,\text{Å}$. As indicated in some detail in Ref. 26, we have employed an imaginary time method to
find the solution of these equations written as imaginary time ($\tau$) diffusion equations. After
every $\tau$-step, the $^4$He density is normalized to $N_4$, whereas the $^3$He wf’s are orthonormalized
using a Gram-Schmidt scheme. To start the iteration procedure, we have used the halved
density of a $^4$He$_{2N_4}$ cluster obtained form a spherically symmetric FRDF code, and random
numbers to build the $^3$He wf’s $\psi(r, z)$. This leaves little room to introduce any bias in the
final results.

III. ONE $^3$HE ATOM ON $^4$HE CLUSTERS SPREADED ON ALKALI SURFACES

The first problem to consider is the sp spectrum for one single $^3$He atom in the field of a
$^4$He$_{N_4}$ cluster on an alkali surface. As an example, we solve the corresponding Schrödinger
equation for the $^3$He atom for $^4$He drops with $N_4 = 20$ and 100 on a Cs surface. In Fig. 1
we show the sp level scheme $\varepsilon_{nl}$ in terms of squared angular momentum $l$, up to $\mathcal{E}_3 = -3.13$
K (cf. Tab. I). For comparison, the lowest panel displays the sp spectrum obtained in I for
a cluster with $N_4 = 1000$. Localized states with energy higher than -3.13 K are meaningless,
as they would be an artifact of the calculation, carried out in a large but finite box. Indeed,
on any substrate, $^3$He atoms with energies higher than the corresponding $\mathcal{E}_3$, would prefer
to leave the $^4$He neighborhood and occupy the lowest lying sp state of the alkali surface
potential. One can see from Fig. 1 that whereas there are many $^3$He sp states below $\mathcal{E}_3$ for
$N_4 = 1000$ (notice that only the lowest lying ones are shown), their number decreases with
decreasing $N_4$. In particular, for $N_4 = 20$ only the states with $n = 1$ and $l = 0$ to 3 ($s, p, d,$
and $f$ states in spectroscopic notation), and the $2s$ state are bound to the $^4$He droplet.

In I we have shown that, for large $N_4$ values, states $(nl)$ with $n = 1$ and $l = 0, 1, 2, \ldots$
distributed into a rotational band. This means, on the one hand, that their sp energies
lie on a perfect straight line as functions of $l^2$ as seen in the bottom panel of Fig. 1, and on the other hand, that their probability distributions $|\psi_{l1}(r,z)|^2$ are sensibly identical to $|\psi_{l0}(r,z)|^2$. Depending on $N_4$, the same may happen, to some extent, to $(2l)$ states with $l = 0, 1, 2, \ldots$ (cf. Fig. 1). For $N_4 = 20$, the small size of the $^4\text{He}$ host cluster does not allow the energy levels to group into rotational bands, even for gs-based states $\psi_{l1}$. This is due to the fact that the wf’s $\psi_{n0}$ lie close enough to the $z$ axis to experience centrifugal distortion at any nonvanishing $l$. The rotational character of the gs band $\varepsilon_{l1}$ is recovered for $N_4 = 100$, as depicted in the middle panel of Fig. 1.

Contour plots of the probability densities $[\psi_{n0}(x,z)]^2$ for $n = 1$ to 3, on the $(x,z)$ plane, together with those of the density $\rho_4(x,z)$, are shown in Fig. 2 for $N_4 = 100$. It is clear that the gs $\psi_{10}$ is localized on the circular contact line, revealing once again the edge state reported in I, where similar plots were presented for $N_4 = 1000$. In that work, we also showed that for $n > 1$, the probability densities display several fringes on the surface of the $^4\text{He}$ cluster; a similar pattern occurs for $N_4 = 100$. This is not the case if $N_4$ is as low as 20, since although the edge state is still present, the small host immediately pulls out the $^3\text{He}$ probability density into unbound configurations (cf. upper panel in Fig. 1). We show in Fig. 3 contour plots of the probability densities $[\psi_{l1}(x,z)]^2$ for $l = 0, 1, 2$ (hereafter all the contour plots figures are drawn on the $y = 0$ plane), together with those of the density $\rho_4(x,z)$, for $N_4 = 20$.

Effective masses $m_{n0}^*$, defined as the state averages of the parametrized local prefactor of the $^3\text{He}$ kinetic energy in the density functional Eq. (1)

$$\frac{1}{m_{n0}^*} = \int dr \frac{|\psi_{l1}(r,z)|^2}{m_3^*[\rho_4(r,z),\rho_3(r,z)]}$$

are displayed in Table II. The rotational character of the gs band $\varepsilon_{l1}$ if $N_4 = 100$, allows one to fit these sp energies to a law

$$\varepsilon_{l1} = \varepsilon_{l0} + \frac{\hbar^2l^2}{2m_{10}^*R_{10}^2}$$

with $\varepsilon_{l0} = -4.52$ K and regression unity. From $m_{10}^* = 1.18 m_3$, we obtain a gs radius $R_{10} = 15.5$ Å, which sensibly coincides with the geometrical radius of the droplet at a $^4\text{He}$ density of about $10^{-2}\text{Å}^{-3}$, as viewed in the bottom panel of Fig. 2.

The appearance of the $^3\text{He}$ level structure built on these spreaded droplets is quite distinct. Within an strictly independent particle model, one may see from Fig. 1 that a fairly large amount of $^3\text{He}$ is needed before $2l$ states start being occupied. For $N_4=100$, only after filling the $1h$ ($l = 5$) state, i.e., $N_3 = 22$, the state $2s$ becomes occupied, and for $N_4=1000$, only after filling the $1k$ ($l = 8$) state, i.e., $N_3 = 34$. For this reason, the KS results discussed in Sec. IV refer to rather small $^4\text{He}$ droplets. Otherwise, the number of $^3\text{He}$ sp orbitals to compute, in order to see a sizeable effect due to the presence of this isotope, becomes prohibitive.

It is also interesting to see how the number of $\psi_{n0}$ localized bound states, with energies below the $E_3$ value in Table I, varies with $N_4$ and the alkali substrate. In particular, for Na and Li we have found only one single $l = 0$ bound state, namely the edge state $\psi_{10}$, for $N_4$ values from 20 to 3000. This is at variance with the results found for K, for which, similarly to Cs, the droplet with $N_4=20$ can only sustain one $s$ state, whereas the drop with $N_4=100$...
and larger may sustain \( n = 1, 2 \) and \( 3 \) \( s \) states. We show in Fig 4 the contour plots of the probability densities \([\psi_{n0}(x, z)]^2\) for \( n = 1 \) to 3, together with those of the density \( \rho_4(x, z) \), for \( N_4 = 3000 \) on K. As in the case of Cs\(^{22}\), the probability densities of the excited states display peaks on the upper surface of the drop.

Contour plots of the probability density \([\psi_{10}(x, z)]^2\), together with those of the density \( \rho_4(x, z) \), for \( N_4 = 3000 \) on K. As in the case of Cs\(^{22}\), the probability densities of the excited states display peaks on the upper surface of the drop.

Figure 6 shows the energy \( \varepsilon_{1s} \) of the edge state, as a function of \( N_4 \) for Cs, K, Na, and Li substrates, with \( \varepsilon_{1s}(N_4 = 0) \equiv \mathcal{E}_3 \). It should be kept in mind that, as shown in Ref. 26, in the current FRDF description, Li, Na and K are wetted by \( ^4\)He, whereas Rb (not discussed here) and Cs are not. To establish a connection with the situation in films, for a given \( N_4 \) droplet we define

\[
n_4(r) = 2 \pi \int dz \rho_4(r, z)
\]  

(11)

and in view of the analysis carried out in Ref. 26, it is safe to consider that \( n_4(0) \) for the largest droplets \( N_4 = 3000 \) represents the prewetting coverage \( n_c \) of a \( ^4\)He film on the given adsorber. This yields \( n_c = 0.467 \text{Å}^{-2} \) for K, \( n_c = 0.140 \text{Å}^{-2} \) for Na, and \( n_c = 0.056 \text{Å}^{-2} \) for Li. We have obtained the structure of films in the vicinity of \( n_c \), working out the first three \( ^3\)He sp states as illustrated in Figs. 7 and 8.

In Fig. 7 we plot the \( ^4\)He density as a function of the perpendicular distance \( z \), for films on K, Na and Li, computed at the corresponding \( n_c \), together with the mean field for one \( ^3\)He atom, the gs wave function \( \psi_0(z) \) and the first excited state wave function \( \psi_1(z) \). In Fig. 8 we display the energies \( \varepsilon_n \), \( n = 0 \) to 2 (see also Fig. 7) for \( ^4\)He coverages around \( n_c \) on the same three substrates, with the gs energy \( \mathcal{E}_3 \) of the \( ^3\)He atom on the substrate shown as a dotted line. The heavy dot indicates their corresponding \( n_c \).\(^{29}\) As seen in Fig. 7, the wave functions follow the trend anticipated in Ref. 30: the gs of a \( ^3\)He impurity in a \( ^4\)He film remains mostly concentrated inside the Li monolayer, and is localized at the liquid-vapor interface if the film thickness is above one layer (K and Na).

The energetics of the impurity shows an interesting evolution across the prewetting transition. In the case of K, the weakest adsorber of this series, for the range of \( n_4 \) values here considered, the film spectrum is essentially constant with coverage, and for all three adsorbers the first excited state appears above the gs of the \( ^3\)He atom on the substrate. In contradistinction, at the prewetting density \( n_c \) the energy of the Andreev state is below \( \mathcal{E}_3 \) for K, and above it for Na and Li. We appreciate then that an important finite size effect for \( ^4\)He clusters on these later two adsorbers is the generation of the edge state for the \( ^3\)He impurity, which is favored with respect to binding to the substrate, thus permitting the existence of mixed drops of both helium isotopes on these adsorbers. It may also be inferred from Fig. 8 the different behaviour of adsorbers like K and weaker, as compared to Na adsorbers and stronger. Whereas in the former case, after filling the edge-like states, \( ^3\)He atoms start populating Andreev-like states covering the cup of the \( ^4\)He droplet, in the latter, once the edge-like states are filled, \( ^3\)He spreads on the alkali surface.
IV. MIXED CLUSTERS ON CS

We now solve the coupled EL/KS equations (7) and (8) for \( N_4 = 20 \) and 100, and for several \( N_3 \) values, on a Cs substrate. Fig. 9 displays the variation of the lowest KS sp energies \( \varepsilon_{nl} \) as functions of \( N_3 \) for the case \( N_4 = 20 \), with heavy dots indicating the Fermi energy/chemical potential \( \mu_3 \) of the given configuration. As seen in this figure, the chemical potential \( \mu_3 \) is a -nonmonotonically- increasing function of the system size. This trend coincides with that encountered in \(^3\)He, either pure\(^6\) or dissolved in thick \(^4\)He films.\(^{12,13}\) Figure 9 also shows that for \( N_4=20 \), values of \( N_3 = 6, 10, 16, \) and 30 yield strong shell closures, and they are thus the magic numbers for the fermion component of a \(^4\)He\(_{20}^{}\)+\(^3\)He\(_{N_3}^{}\) droplet on Cs -these numbers depend on the alkali substrate and on \( N_4 \). Weaker shell closures also appear for \( N_3 = 2, 20, 24, 34, \) and 38.

Contour plots of the particle densities \( \rho_3(x,z) \) and \( \rho_4(x,z) \) are shown in Fig. 10 for \( N_4 = 20 \), and in Fig. 11 for \( N_4 = 100 \), and two values of \( N_3 \). As we analyze these patterns, several observations may be put forward. On the one hand, we verify the spreading tendency of the density profiles \( \rho_3 \) with growing \( N_3 \), as well as a tendency to cover the horizontal base and show some slight dilution inside the bulk of the host. It is also apparent from these figures that \(^3\)He atoms in Andreev surface states do coat the \(^4\)He cap. On the other hand, the shape of the \(^4\)He droplet is rather insensitive to the presence of the intruder atoms, whose outwards spreading does not induce splashing of the \(^4\)He cluster on the substrate surface as well.

The behavior of the \(^3\)He density should be interpreted together with examination of the KS level scheme in Fig. 9: an energetically based criterion for spreading of mixtures of \((N_3,N_4)\) atoms on any substrate is given by the constraint \( \mu_3(N_3c,N_4) = E_3 \), which defines a critical size \( N_3c(N_4) \) above which it is energetically more convenient, for an extra \(^3\)He atom, to adhere to the flat substrate than to the \(^4\)He drop. As seen in Fig. 9, for \( N_4=20 \), \( N_3c \) is about 44.

Finally, a word of caution should be spoken with respect to the contact angle, for whose determination in large, macroscopic droplets, a procedure has been developed in Ref. 20. As discussed in I, due to the fact that nanoscopic \(^4\)He droplet densities present large stratification near the substrate, the contact angle cannot be established by ‘visual inspection’ of the equidensity lines. Yet, even at a qualitative level of description, it is clear from the results here presented that addition of \(^3\)He atoms affects the contact angle. To illustrate this assertion further, in Fig. 12 we show the contour plot of the total \(^4\)He+\(^3\)He density for \(^4\)He equidensity lines about 0.011 \( \text{Å}^{-3} \), roughly half the bulk \(^4\)He density, for \( N_4 = 20 \) and 100 and several \( N_3 \) values. Although the small size of these droplets prevents us from drawing quantitative conclusions, this figure indicates that the height of the cap changes less than the radius of its base, thus suggesting overall flattening of the combined density and a decrease of the contact angle, as expected according to both theory and experiment carried upon macroscopic samples.

V. SUMMARY

In our previous work here denoted as I, we have shown that in addition to the well known Andreev states, nanoscopic \(^4\)He drops on Cs substrates host a new class of \(^3\)He sp states,
edge states that generate a 1D ring of $^3$He atoms along the contact line. In this work we have carried a deeper investigation of size effects in mixed droplets on planar alkali surfaces, that enables us to assert that these edge states are a definite consequence of the particular combination of the confinement provoked by the adsorbing field, selfsaturation of the helium system and mutual interaction between isotopes. In fact, these localized states appear in all alkali substrates and for all $^4$He clusters, although their 1D nature becomes more evident either above nanoscopic host sizes or for the strongest confining potentials. We are able to compare the energy spectrum of single $^3$He impurities in $^4$He clusters with similar spectra on films, and found that while the evolution of the lowest lying sp states with film coverage near the prewetting transition may prevent solvation of a $^3$He atom in a film on a strong adsorber such as Na or Li, its gs energy being unfavorable compared with binding to the substrate, the breaking of translational symmetry introduced by the finite size of clusters does change this trend, giving rise to an energetically favored edge state which remains localized around the drop. We also show that increasing $N_3/N_4$ ratios enlarge the tendency of the mixed cluster to spread outwards on the surface; explicit KS calculations of the energy spectrum of added $^3$He atoms and examination of the Fermi energy as a function of $N_3$ support the quantummechanical interpretation for the onset of wetting by mixtures of helium fluids, as driven by the energetic benefit of abandoning the $^4$He and adhering instead to the substrate.

ACKNOWLEDGMENTS

This work has been performed under grants BFM2002-01868 from DGI, Spain, 2001SGR00064 from Generalitat of Catalonia, EX-103 from University of Buenos Aires, and PICT2000-03-08450 from ANPCYT, Argentina. E.S.H. has been also funded by M.E.C.D. (Spain) on sabbatical leave.
REFERENCES

1. J. Klier, P. Stefani and A. F. G. Wyatt, Phys. Rev. Lett. 75, 3709 (1995).
2. E. Rolley and C. Guthmann, J. Low Temp. Phys. 108, 1 (1997).
3. D. Ross, P. Taborek and J. E. Rutledge, J. Low Temp. Phys. 111, 1 (1998).
4. J. Klier and A. F. G. Wyatt, J. Low Temp. Phys. 110, 919 (1998).
5. L. Szybisz, Phys. Rev. B 67, 132505 (2003).
6. L. Pricaupenko and J. Treiner, Phys. Rev. Lett. 72, 2215 (1994).
7. D. Ross, J. A. Phillips, J. E. Rutledge and P. Taborek, J. Low Temp. Phys. 106, 81 (1997).
8. D. O. Edwards and W. F. Saam, Progress in Low Temperature Physics, ed. D. F. Brewer, North Holland, Amsterdam (1978) p. 283.
9. R. B. Hallock, J. Low Temp. Phys. 101, 31 (1995).
10. M. Barranco, M. Pi, S. M. Gatica, E. S. Hernández, and J. Navarro, Phys. Rev. B 56, 8997 (1997).
11. M. Pi, R. Mayol, and M. Barranco, Phys. Rev. Lett. 82, 3093 (1999).
12. N. Pavloff and J. Treiner, J. Low Temp. Phys. 83, 331 (1991).
13. J. Treiner, J. Low Temp. Phys. 92, 1 (1993); L. Pricaupenko and J. Treiner, J. Low Temp. Phys. 101, 349 (1995).
14. M. S. Pettersen and W. F. Saam, J. Low Temp. Phys. 90, 159 (1993).
15. D. Ross, J. E. Rutledge, and P. Taborek, Phys. Rev. Lett. 74, 4483 (1995).
16. K. S. Ketola and R. B. Hallock, Phys. Rev. Lett. 71, 3295 (1993); K. S. Ketola, T. A. Moreau, and R. B. Hallock, J. Low Temp. Phys. 101, 343 (1995).
17. M. S. Pettersen and W. F. Saam, Phys. Rev. B 51, 15369 (1995); W. F. Saam and M. S. Pettersen, J. Low Temp. Phys. 101, 355 (1995).
18. D. Ross, J. E. Rutledge, and P. Taborek, Phys. Rev. Lett. 76, 2350 (1996).
19. J. Klier and A. F. G. Wyatt, J. Low Temp. Phys. 116, 61 (1999).
20. F. Ancilotto, A. M. Sartori, and F. Toigo, Phys. Rev. B 58, 5085 (1998).
21. F. Dalfovo, A. Lastri, L. Pricaupenko, S. Stringari, and J. Treiner, Phys. Rev. B 52, 1193 (1995).
22. R. Mayol, M. Barranco, E. S. Hernández, M. Pi, and M. Guilleumas, Phys. Rev. Lett. 90, 185301 (2003).
23. R. Mayol, M. Pi, M. Barranco, and F. Dalfovo, Phys. Rev. Lett. 87, 145301 (2001).
24. J. Dupont-Roc, M. Himbert, N. Pavloff and J. Treiner, J. Low Temp. Phys. 81, 31 (1990).
25. A. Chizmeshya, M. W. Cole and E. Zaremba, J. Low Temp. Phys. 110, 677 (1998).
26. M. Barranco, M. Guilleumas, E. S. Hernández, R. Mayol, M. Pi, and L. Szybisz, eprint cond-mat/0303500, submitted to Phys. Rev. B (2003).
27. J. M. Luttinger, J. Math. Phys. 4, 1154 (1962).
28. F. D. M. Haldane, Phys. Rev. Lett. 47, 1840 (1981); J. Chem. Phys. 14, 2585 (1981).
29. Actually, in the case of K, we have not reached $n_c$ in the calculations. This is not crucial, since for this alkali, the spectrum is insensitive to the actual value of the coverage for values above $\sim 0.40 \text{Å}^{-2}$, see for example Ref. 12.
30. N. Pavloff and J. Treiner, J. Low Temp. Phys. 83, 15 (1991).
TABLE I. Energies $\mathcal{E}_3$ and $\mathcal{E}_4$ (K) of one $^3$He and one $^4$He atom on different alkali substrates.

|       | Li  | Na  | K   | Rb  | Cs  |
|-------|-----|-----|-----|-----|-----|
| $\mathcal{E}_3$ | -9.81 | -6.41 | -3.74 | -3.31 | -3.13 |
| $\mathcal{E}_4$ | -10.70 | -7.08 | -4.20 | -3.72 | -3.53 |

TABLE II. Effective masses $m_{n0}^*$ (in units of $m_3$) for single $^3$He atoms in He$_{N_4}$ clusters on Cs.

| $N_4$ | 20 | 100 | 500 | 1000 | 2000 | 3000 |
|-------|----|-----|-----|------|------|------|
| $m_{10}^*$ | 1.13 | 1.18 | 1.20 | 1.21 | 1.22 | 1.22 |
| $m_{20}^*$ | 1.10 | 1.26 | 1.29 | 1.30 | 1.31 | 1.31 |
| $m_{30}^*$ | 1.05 | 1.24 | 1.30 | 1.30 | 1.30 | 1.30 |
FIGURES

FIG. 1. Single-particle energies $\varepsilon_{nl}$ of one $^3$He atom in a $^4$He$_{N_4}$ droplet on Cs, as functions of squared angular momentum projection, for $N_4 = 20, 100$ and $1000$. The dotted lines have been drawn to guide the eye. In each panel, from bottom to top, the states have principal quantum numbers $n = 1, 2, 3, \ldots$, and the dot-dashed line represents the gs energy $E_{3}$ of one $^3$He atom on the Cs substrate.

FIG. 2. Contour plots of the probability densities $[\psi_{n0}(x, z)]^2$ for a single $^3$He atom in a $^4$He$_{100}$ droplet on Cs, together with the density $\rho_4(x, z)$ (bottom panel). In the case of $^4$He, the equidensity lines correspond to $10^{-4}, 10^{-3}, 5 \times 10^{-3}, 2 \times 10^{-2}$, and $2.5 \times 10^{-2}$ Å$^{-3}$. In the case of $^3$He, the equiprobability lines start at $10^{-4}$ Å$^{-3}$ and increase in $10^{-4}$ Å$^{-3}$ steps.

FIG. 3. Contour plots of the probability densities $[\psi_{1l}(x, z)]^2$ for a single $^3$He atom in a $^4$He$_{20}$ droplet on Cs, together with the density $\rho_4(x, z)$ (bottom panel). The equidensity and equiprobability lines are as in Fig. 2.

FIG. 4. Contour plots of the probability densities $[\psi_{n0}(x, z)]^2$ for a single $^3$He atom in a $^4$He$_{3000}$ droplet on K, together with the density $\rho_4(x, z)$ (bottom panel). The equidensity lines are as in Fig. 2, and the equiprobability lines start at $10^{-5}$ Å$^{-3}$ and increase in $10^{-5}$ Å$^{-3}$ steps.

FIG. 5. Contour plots of the probability density $[\psi_{10}(x, z)]^2$ for a single $^3$He atom in a $^4$He$_{3000}$ droplet on Li, together with the density $\rho_4(x, z)$ (bottom panels), and on Na (top panels). In the case of $^4$He, the equidensity lines are as in Fig. 2, and in the case of $^3$He, the equiprobability lines correspond to $10^{-8}, 10^{-7}, 10^{-6}, 10^{-5}, 2 \times 10^{-5}, 2.5 \times 10^{-5}$ and $5 \times 10^{-5}$ Å$^{-3}$.

FIG. 6. Energy $\varepsilon_{10}$ of the edge state (K) as a function of $N_4$. The dotted lines have been drawn to guide the eye. The $y$ axis is broken below $\sim -7.5$ K.

FIG. 7. The $^4$He density at the prewetting coverage $n_c$ on Na and Li substrates, and at $n_4 = 0.40$ Å$^{-2}$ for K, together with the mean field $V_3$ experienced by a $^3$He impurity, and the gs wave function $\psi_0(z)$ and first excited wf $\psi_1(z)$ in that field.

FIG. 8. The sp energies $\varepsilon_{nl}$, $n = 0, 1, 2$, for a $^3$He impurity in $^4$He films at coverages around $n_c$ on K, Na and Li. The dotted line indicates the gs energy $E_3$ of one $^3$He atom on the corresponding substrate, and the heavy dot points to the coverage at the prewetting jump.$^{29}$

FIG. 9. The Kohn-Sham sp energies $\varepsilon_{nl}$ for $^3$He$_{N_3}$+$^4$He$_{20}$ mixed drops on Cs as functions of $N_3$. The horizontal line indicates the energy $E_3$ of one $^3$He atom in the Cs adsorbing field. Heavy dots indicate the corresponding Fermi energy. The lines connecting the sp energies of $(nl)$ states have been drawn to guide the eye.
FIG. 10. Contour plots of the density profiles $\rho_3(x, z)$ (top panels) and $\rho_4(x, z)$ (bottom panels) of $^4\text{He}_{20} + ^3\text{He}_{N_3}$ mixed drops on Cs, for $N_3 = 20$ (left panels) and 40 (right panels). In the case of $^4\text{He}$, the equidensity lines are as in Fig. 2, and in the case of $^3\text{He}$, they correspond to $10^{-4}, 2.5 \times 10^{-4}, 5 \times 10^{-4}, 7.5 \times 10^{-4}, 10^{-3}, 2.5 \times 10^{-3}, 5 \times 10^{-3}, 7.5 \times 10^{-3}$, and $10^{-2}\text{Å}^{-3}$.

FIG. 11. Same as Fig. 10 for $N_4 = 100$, and $N_3 = 46$ (left panels), and $N_3 = 86$ (right panels). The equidensity lines are as in Fig. 10.

FIG. 12. Contour plots of the total $^4\text{He} + ^3\text{He}$ density corresponding to $0.011 \text{ Å}^{-3}$ for the case $N_4 = 20$, $N_3 = 20$ and 40 (top panel), and $N_4 = 100$, $N_3 = 46$ and 86 (bottom panel). The dashed lines correspond to the pure $^4\text{He}_{N_4}$ drop.
This figure "fig1.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig2.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig3.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig4.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig5.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig7.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig8.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig9.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig10.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig11.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1
This figure "fig12.png" is available in "png" format from:

http://arxiv.org/ps/cond-mat/0305590v1