Wetting of planar substrates of rubidium by liquid films of ⁴He

Leszek Szybisz*

Laboratorio TANDAR, Departamento de Física, Comisión Nacional de Energía Atómica, Av. del Libertador 8250,

RA-1429 Buenos Aires, Argentina

and Departamento de Física, Facultad de Ciencias Exactas y Naturales, Universidad de Buenos Aires, Ciudad Universitaria,

RA-1428 Buenos Aires, Argentina

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The wetting of planar solid surfaces of Rb by superfluid ⁴He films at T=0 K is examined theoretically. The calculations were carried out by (i) using the most elaborated nonlocal density functional known as the Orsay-Trento proposal and (ii) assuming that the helium atoms interact with the substrate via the potential recently worked out by Chizmeshya, Cole, and Zaremba. The asymptotic surface tension was evaluated by applying two different procedures. Our results indicate that films of ⁴He wet Rb at zero absolute temperature.

I. INTRODUCTION

The adsorption of helium films on solid planar substrates is a subject with a long tradition in low-temperature physics. However, the interest in this matter has been significantly increased during the last decade after the pioneering theoretical work of Cheng et al.¹ on properties of ⁴He films adsorbed on alkali metals at T=0 K. These authors have proposed the possibility of nonwetting of the heaviest elements of this sort Cs, Rb, and K. In this geometry, the helium system is translationally invariant on the x-y plane determined by an inert solid planar surface of the substrate, while symmetry is broken in the z direction by the action of the helium-substrate potential $U_{sub}(z)$, which gives rise to a density profile $\rho(z)$ for the distribution of helium atoms of mass m. In the framework of density functional (DF) theories the energy per particle is²

$$e = \frac{E_{\text{g.s.}}}{N} = \frac{1}{n_c} \left[\frac{\hbar^2}{2m} \int_0^\infty dz \left(\frac{d\sqrt{\rho(z)}}{dz} \right)^2 + \int_0^\infty dz \,\rho(z) e_c(z) + \int_0^\infty dz \,\rho(z) U_{\text{sub}}(z) \right].$$
(1.1)

Here n_c is the coverage defined as the number of particles N per unit area A

$$n_c = \frac{N}{A} = \int_0^\infty dz \,\rho(z), \qquad (1.2)$$

and $e_c(z)$ is the correlation energy per particle depending on the DF approach. The minimization of the energy given by Eq. (1.1) with the constrain of fixed n_c leads to

$$\left[-\frac{\hbar^2}{2m} \frac{d^2}{dz^2} + V_H(z) + U_{\rm sub}(z) \right] \sqrt{\rho(z)} = \mu \sqrt{\rho(z)}.$$
(1.3)

Here $V_H(z)$ is the Hartree mean-field potential

$$V_H(z) = \frac{\delta E_c[\rho]}{\delta \rho(z)} = \frac{\delta}{\delta \rho(z)} \int_0^\infty dz \, \rho(z) e_c(z). \quad (1.4)$$

By solving Eq. (1.3) one gets $\rho(z)$ as well as the chemical potential μ . The nonwetting phenomenon was argued in Ref. 1 on the

basis of results obtained by using (i) the $e_c(z)$ provided by the nonlocal density functional (NLDF) proposed by the Orsay-Paris (OP) Collaboration³ and (ii) the simple twoparameter "3-9" adsorption potential

$$U_{\rm sub}(z) = U_{3-9}(z) = \left(\frac{4 C_3^3}{27 D^2}\right) \frac{1}{z^9} - \frac{C_3}{z^3}, \qquad (1.5)$$

with the well depth D and the van der Waals (vdW) coefficient C_3 derived by Zaremba and Kohn.⁴ This fascinating feature is obtained because the well depths of the heliumsubstrate interactions are comparable to that of the heliumhelium interaction.⁵ Taborek and Rutledge⁶ and Hallock⁷ have reported instructive reviews of experiments done in the early nineties for checking this prediction. Meanwhile, progress in the theory has been achieved. An improved NLDF was proposed by the Orsay-Trento Collaboration (OT-NLDF).8 This approach accounts better than the OP-NLDF for properties of uniform and nonuniform helium systems.^{8,9} The adsorbate-substrate interaction has been revised by Chizmeshya, Cole, and Zaremba¹⁰ (CCZ) who proposed the expression (CCZ potential)

$$U_{\rm sub}(z) = U_{\rm CCZ}(z)$$

= $V_0(1 + \alpha z) \exp(-\alpha z)$
 $-f_2[\beta(z)(z - z_{\rm vdW})] \frac{C_{\rm vdW}}{(z - z_{\rm vdW})^3},$ (1.6)

where

and

 $f_2(x) = 1 - \left(1 + x + \frac{x^2}{2}\right) \exp(-x),$ (1.7)

$$\beta(z) = \frac{\alpha^2 z}{1 + \alpha z}.$$
(1.8)

All the parameters of these expressions are quoted in Table 1 of Ref. 10. For a given substrate the CCZ potential is more

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TABLE I. Wetting temperature T_w for the adsorption of ⁴He on planar rubidium substrates.

Experiment	Theory	T_w [K]	Ref.
Heat transport		0	15
Quartz microbalance		<1.6	16
Mass flow		0.31	17
Third sound		< 0.15	18
Quartz microbalance		0	13,19
	Simple model	1.3	10
	OT-NLDF	0	PW

^aResults corresponding to the CCZ potential of Ref. 10.

attractive than the "3-9" potential with D and C_3 taken from Ref. 4 (see discussion in Ref. 10).

At the present time, the wetting properties of the heaviest element Cs and the lightest one K of the triad put into consideration in Ref. 1 show a consistent picture. Experimentally it was verified⁶ that ⁴He does not wet Cs at temperatures lower than the wetting temperature $T_w \approx 2$ K and, in addition, the contact angle for this substrate was measured (see, e.g., Ref. 11). Both these properties were fairly well reproduced theoretically by using the CCZ potential.^{10,12} On the other hand, experimental data¹³ indicate that potassium is wetted by ⁴He at T=0 K. A simple model estimation¹⁰ as well as a very recent microscopic calculation carried out within NLDF formalisms¹⁴ successfully accounted for this feature by using the CCZ potential.

The situation of the adsorption of ⁴He on Rb is still not completely clear. In the following lines we shall summarize the state-of-the art concerning wetting in this case. Since the attraction of the adsorption potential increases for decreasing mass number of alkali metals, Rb should exhibit lower wetting temperatures than Cs. The question is whether it is zero as for a surface of K or takes a finite value. Several different kind of measurements including heat and mass transport, third sound, and quartz crystal microbalance have been made. By using an extrapolation scheme that accounts for the temperature dependence of data provided by heat transport measurements in the range 1.0-1.7 K,15 the authors have obtained $T_w = 0$ K. Early quartz microbalance data¹⁶ indicated that the wetting temperature T_w is well below the lowest examined temperature 1.6 K. From a mass flow study of ⁴He across a Rb ring, performed at temperatures between 0.1 and 0.4 K, it was determined $T_w = 0.31$ K.¹⁷ In the case of third sound, it was clearly seen that the behavior depends on the preparation of the Rb substrate, but nevertheless from the experiments it can be definitely stated that $T_w < 0.15$ K.¹⁸ Finally, a recent measurement using the quartz crystal microbalance technique has undoubtedly indicated that ⁴He wets Rb surfaces at T=0 K.^{13,19} So, with the exception of the result of Ref. 17, the experimental data quoted in Table I are consistent with $T_w = 0$ K. However, the recent theoretical evaluation of Ref. 10 performed by using the CCZ adsorption potential yielded $T_w = 1.3$ K. This wetting temperature is not only different from zero, but in fact it also exceeds significantly the sole non null experimental value.

In view of the discrepancy between experimental results and the simple theoretical estimation, we found important to carry out new microscopic calculations in order to shed some light on wetting of Rb. It is the aim of the present paper to report our results obtained by applying the OT-NLDF formalism and using the novel CCZ adsorption potential. The corresponding expression for $V_H(z)$ is provided in a recent paper where several DF approaches are discussed.⁹ Section II is devoted to examine the calculated solutions putting a special emphasis in the analysis of the surface tension. The final conclusion is presented in Sec. III.

II. ANALYSIS OF NUMERICAL SOLUTIONS

Before beginning the data analysis we shall summarize the equations to be used and comment on their physical content. The solutions of Eq. (1.3) provide an equation of state *e* as a function of n_c , which must satisfy the basic laws of thermodynamics. At T=0 K the variation of the groundstate energy of a single-component system of *N* particles which presents flat interfaces of area *A* (notice that in this case the pressure is zero) is given by^{20,21}

$$dE_{\sigma_{S}} = \sigma_{A} \, dA + \mu \, dN, \qquad (2.1)$$

where σ_A is the total surface tension (i.e., the sum of the surface tensions at the solid-liquid and liquid-vacuum interfaces). The formal thermodynamic definitions of σ_A and μ yield

$$\sigma_A = \left(\frac{\partial E_{\text{g.s.}}}{\partial A}\right)_N = \left(\frac{\partial (E_{\text{g.s.}}/N)}{\partial (A/N)}\right)_N = -n_c^2 \frac{de}{dn_c} \qquad (2.2)$$

and

$$\mu = \left(\frac{\partial E_{\text{g.s.}}}{\partial N}\right)_{A} = \left(\frac{\partial (E_{\text{g.s.}}/A)}{\partial (N/A)}\right)_{A} = e + n_{c}\frac{de}{dn_{c}}.$$
 (2.3)

As pointed out in Ref. 21, a necessary condition for the stability of a system is to require a positive areal isothermal compressibility κ_s

$$\frac{1}{\kappa_s} = A \left(\frac{\partial \sigma_A}{\partial A} \right)_N = \frac{A}{N} \left(\frac{\partial \sigma_A}{\partial (A/N)} \right)_N = -n_c \frac{d \sigma_A}{dn_c} > 0. \quad (2.4)$$

From Eqs. (2.2) and (2.3) one gets the well-known relation

$$\sigma_A = n_c(e - \mu) = \frac{E_{\text{g.s.}} - \mu N}{A} = \frac{\Omega}{A}.$$
 (2.5)

Here Ω is the thermodynamic grand potential at T=0 K. By taking into account this relation, Eq. (2.1) may be cast into a Gibbs-Duhem form

$$d[\sigma_A A - (E_{g.s.} - \mu N)] = A \, d\sigma_A + N \, d\mu = 0.$$
 (2.6)

A straightforward integration

$$\int_{0}^{n_{c}} d\sigma_{A} = -\int_{0}^{n_{c}} n_{c}' d\mu(n_{c}') = -\mu(n_{c})n_{c} + \int_{0}^{n_{c}} \mu(n_{c}') dn_{c}'$$
(2.7)

allows us to compute the evolution of the surface tension in terms of an integral of the chemical potential over all the smaller films PRB <u>62</u>

$$\sigma_A(n_c) - \sigma_A(0) = \int_0^{n_c} [\mu(n'_c) - \mu(n_c)] dn'_c. \quad (2.8)$$

Wetting requires that in the limit of infinite coverage the grand potential per unit area, $\sigma_A(n_c \rightarrow \infty) = \sigma_\infty = \sigma_{SL} + \sigma_{LV}$ (here σ_{SL} and σ_{LV} are the asymptotic surface tensions at the solid-liquid and liquid-vacuum interfaces, respectively), be the absolute minimum.²²

Since wetting is governed by the asymptotic behavior of the energetics of planar films, it is convenient to examine e as a function of the inverse of coverage²³ writing the following polynomial expansion:^{9,14,24}

$$e = e_{\infty} + \frac{\sigma_{\infty}}{n_c} + \frac{\gamma_c}{n_c^3} + \frac{a_4}{n_c^4} + \frac{a_5}{n_c^5}.$$
 (2.9)

Here e_{∞} may be identified with the saturation equilibrium value for bulk ⁴He, $e_B = -7.15$ K.²⁵ The use of Eqs. (2.2), (2.3), and (2.4) leads to

$$\sigma_A = -n_c^2 \frac{de}{dn_c} = \sigma_\infty + 3 \frac{\gamma_c}{n_c^2} + 4 \frac{a_4}{n_c^3} + 5 \frac{a_5}{n_c^4}, \quad (2.10)$$

$$\mu = e + n_c \frac{de}{dn_c} = e_{\infty} - 2 \frac{\gamma_c}{n_c^3} - 3 \frac{a_4}{n_c^4} - 4 \frac{a_5}{n_c^5}, \quad (2.11)$$

and

$$\frac{1}{n_c \kappa_s} = -\frac{d\sigma_A}{dn_c} = n_c \frac{d\mu}{dn_c} = m c_3^2 = 6 \frac{\gamma_c}{n_c^3} + 12 \frac{a_4}{n_c^4} + 20 \frac{a_5}{n_c^5}.$$
(2.12)

In fact, the stability condition of Eq. (2.4) was rewritten in terms of the incompressibility, which is related to the thirdsound velocity c_3 (see the second citation of Ref. 1). The coefficient γ_c drives the departure of σ_A and μ from their asymptotic values. It is mainly determined by the van der Waals tail $U_{\text{sub}}^{(\text{tail})}(z) \approx -C_{\text{tail}}/z^3$ of the adsorption potential. The simplest version of the Frenkel-Halsay-Hill (FHH) model without effects due to retardation [see, e.g., Eq. (5) in Ref. 26] yields

$$\gamma_c \simeq \gamma_c^0 = \frac{1}{2} \rho_0^3 \Gamma_{\text{FHH}} = \frac{1}{2} \rho_0^3 (C_{\text{tail}} - C_{\text{tail}}^{\text{He}}),$$
 (2.13)

where $\rho_0 = 0.021836 \text{ Å}^{-3}$ is the equilibrium bulk density²⁵ and $C_{\text{tail}}^{\text{He}} \approx 120 \text{ K} \text{ Å}^3$ is the *C* coefficient of a hypothetical ⁴He substrate.

The integrodifferential problem (1.3) has been solved for a large range of coverages up to $n_c^{\text{max}} = 0.44 \text{ Å}^{-2}$ corresponding to a film of about 5.5 nominal layers $l \ (l = \rho_0^{2/3} = 0.07812 \text{ Å}^{-2})$. This maximum coverage was adopted because for a weak adsorption potential further growth of the system occurs without noticeable new oscillations of $\rho(z)$ [see, e.g., Fig. 6(b) of Ref. 2] implying that the asymptotic trend was reached. Equation (1.3) has been discretized in a box of size enough to insure that the neglected $\sqrt{\rho(z)}$ be always smaller than $10^{-5} \text{ Å}^{-3/2}$. The consistency of solutions for μ was checked with a special care following the procedures developed in Ref. 14.

Figure 1(a) shows the behavior of e and μ as a function of n_c . In the limit $n_c \rightarrow 0$ quantities e and μ converge to the

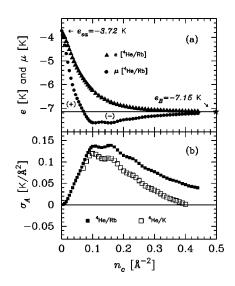


FIG. 1. (a) Energy per particle $e = E_{g.s.}/N$ and chemical potential μ as a function of the coverage n_c . The star is the bulk saturation value $e_B = -7.15$ K. The symbols (+) and (-) indicate the positive and negative contributions to σ_{∞} as given by Eq. (2.14). The region where *e* becomes negative is magnified in the inset. (b) Surface tension σ_A evaluated with Eq. (2.5). For comparison, the surface tension obtained in the case of a substrate of K in Ref. 14 is also displayed.

single-atom binding energy in the $U_{CCZ}(z)$ potential, i.e., $e_{SA} = -3.72$ K. According to this drawing when n_c grows continuously from a certain minimum coverage towards ∞ then μ approaches the asymptotic value e_{∞} from below. This behavior guarantees that large films always satisfy the stability condition of Eq. (2.12). Figure 1(b) shows that σ_A given by Eq. (2.5) is, for large films, a decreasing function of n_c . For the sake of comparison, we also plotted σ_A for K evaluated with data of Ref. 14. Since $\sigma(0)=0$, to ensure wetting of Rb it must be demonstrated that σ_{∞} is negative. There are two ways of determining this value from the calculated data. One way consists of simply fitting e to Eq. (2.9). The other is based on the evaluation of Eq. (2.8) for $n_c \rightarrow \infty$.

Figure 2 shows the behavior of e, μ , and σ_A as a function of $\nu = 1/n_c$. Two different fits were performed. In one of them, the energy data covering the regime $0 \le \nu \le 10 \text{ Å}^2$ were fitted to the polynomial (2.9). In the other, values of μ were fitted to Eq. (2.11) which is independent of σ_{∞} . In the latter case, the fit was restricted to the range $0 \le \nu \le 5$ Å² where both μ and σ_A exhibit their final trend towards the asymptotic values. By following the procedure outlined in Ref. 24 the energy per particle for bulk liquid e_B = -7.15 K was included in the fits. The extracted values of e_{∞} , σ_{∞} , γ_c , a_4 , and a_5 are listed in Table II. As expected, the results for e_{∞} are consistent with e_B . The solid curves in Fig. 2 show the good quality of the fit of *e* and indicate that the results for σ_A and μ evaluated with Eqs. (2.10) and (2.11) reproduce quite well the displayed data. Furthermore, since the obtained σ_{∞} is negative and γ_c is positive (in practice equal to the FHH estimation γ_c^0) both conditions of Eqs. (2.6) and (2.12) required for wetting are satisfied. The inset in Fig. 2(a) indicates the crossing of e and μ which produces the change of sign of σ_A at about $n_c \simeq 1 \text{ Å}^{-2}$. The dashed curve in Fig. 2(a) shows that the fit of μ to Eq. (2.11) is good in the adjusted region.

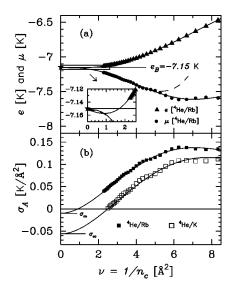


FIG. 2. Same data as in Fig. 1 but as a function of the inverse of coverage $\nu = 1/n_c$. The continuous curves are *e*, μ , and σ_A evaluated with the parameters obtained form the fit of *e* to Eq. (2.9). The dashed curve represents μ calculated with parameters determined from its fit to Eq. (2.11).

When evaluating σ_{∞} with Eq. (2.8), the task is reduced to the computation of the contributing integral. This procedure takes into account the whole spectrum of μ and the result is determined by the balance of areas indicated in Fig. 1(a). Since the positive and negative contributions present a strong cancellation effect, in a first step, we checked whether there is an important limitation in accuracy due to possible systematic numerical deviations of the calculated solutions. For this purpose we computed the surface tension at n_c^{max} =0.44 Å⁻² obtaining σ_A =0.0380 K/Å², which differs less than 5% from the value $\sigma_A(n_c^{\text{max}})$ =0.0398 K/Å² given by Eq. (2.5) and plotted in Fig. 1(b). For computing σ_{∞} , the integral was split into two parts. One running over the solutions for μ displayed in Fig. 1(a) and the other over the asymptotic regime where μ was approximated by Eq. (2.11)

$$\sigma_{\infty} = \int_{0}^{\infty} \left[\mu(n_{c}) - e_{\infty} \right] dn_{c} = \int_{0}^{n_{c}^{\max}} \left[\mu(n_{c}) - e_{\infty} \right] dn_{c} - \gamma_{c} \left(\frac{1}{n_{c}^{\max}} \right)^{2} - a_{4} \left(\frac{1}{n_{c}^{\max}} \right)^{3} - a_{5} \left(\frac{1}{n_{c}^{\max}} \right)^{4}.$$
 (2.14)

Quantity σ_{∞} was evaluated with two different sets of parameters e_{∞} , γ_c , a_4 , and a_5 , namely, those extracted from fits of e and of μ , yielding the results quoted in Table II. Both these values are negative and similar to that determined by the fit of e to Eq. (2.9), being much closer to zero than the result obtained for potassium. It should be stressed that the information used by the procedures based on Eqs. (2.9) and (2.14) does not cover the same regime of n_c , this fact enforces the conclusion pointing out the wetting of the Rb substrate.

III. FINAL REMARKS

In summary, the fundamental output of this investigation is that calculations carried out by using the OT-NLDF

TABLE II. Coefficients of the expansions for the energetics of adsorbed films of liquid 4 He.

Data source	e_{∞}	σ_{∞}	10	$a_4 \times 10^3$	0
	[K]	$[K/Å^2]$	[K/Å ⁶]	$[K/Å^8]$	$[K/Å^{10}]$
Bulk ⁴ He	-7.15 ^a				
Rb Substrate					
FHH ^b (γ_c^0)			4.72		
Fitting e	-7.151	-0.0101	4.71	-0.581	0.0210
Eq. (2.14)		-0.0122 ^c			
Fitting μ	-7.152		4.27	-0.361	0.0045
Eq. 2.14)		-0.0114 ^d			
K Substrate ^e					
FHH ^b (γ_c^0)			4.81		
Fitting e	-7.155	-0.0561	4.48	-0.501	0.0164

^aBulk energy per particle e_B quoted in Table II of Ref. 25 and used for fixing the parameters of the OT-NLDF.

^bCoefficient γ_c estimated by using the FHH approach.

^cUsing coefficients from the fit of e to Eq. (2.9).

^dUsing coefficients from the fit of μ to Eq. (2.11).

^eResults taken from Ref. 14.

approach⁸ and the CCZ adsorption potential¹⁰ indicate the wetting of planar surfaces of Rb by films of ⁴He at T = 0 K in agreement with the majority of the experimental data listed in Table I. However, the almost complete cancellation effect shown by the quantity $\sigma_{\infty} = \sigma_{SL} + \sigma_{LV}$ points out that rubidium lies at the borderline between wetting and non-wetting at zero temperature.

In addition, it recently came to our knowledge that there was a preprint by Ancilotto, Faccin, and Toigo,²⁷ where wetting properties of heavy-alkali metals are analyzed at several temperatures. For their evaluations these authors adopted essentially the same theoretical framework as that used in the present work, but the study was performed in a different way. They calculated $\sigma_{\rm SL}$ and $\sigma_{\rm LV}$ separately finding that in the T=0 K limit ⁴He wets a potassium substrate but not a rubidium one. In the latter case they obtained $\sigma_{\infty} = \sigma_{SL}$ $+\sigma_{\rm LV} \simeq 0.017 \ {\rm K/\AA^2}$ in contrast to our results σ_{∞} $\simeq -0.011 \text{ K/Å}^2$ listed in Table II. However, it should be noticed that this difference is small when compared with the size of the absolute values of σ_{LV} and σ_{SL} . Furthermore, the data plotted in Fig. 4 of Ref. 27 yield $\sigma_{\rm SL}$ [potassium] $-\sigma_{\rm SL}$ [rubidium] ≈ -0.047 K/Å² which is in excellent agreement with the results for σ_{∞} [potassium] – σ_{∞} [rubidium] given by the values quoted in the present Table II. All these facts confirm that the wetting of rubidium surfaces by helium is a very cumbersome problem.

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