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Adsorption of superfluid ^{4}He films on planar heavy-alkali metals studied with the Orsay-Trento density functional

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The wetting of planar surfaces of alkali metals (Cs, Rb, K, and Na) by superfluid ^{4}He films at $T=0$ K is theoretically studied. Calculations have been carried out by using the Orsay-Trento nonlocal density functional and the adsorption potential of Chizmeshya-Cole-Zaremba. Surface tensions and contact angles are determined for several approximations. We find that the conclusion on the wetting of an Rb substrate is sensible to the gradient-gradient term in the functional and the softness of the He-metal interaction.

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In a pioneering theoretical work, Cheng *et al.*[1] have claimed that ^{4}He does not wet planar surfaces of the heavy-alkali metals K, Rb, and Cs at $T=0$ K. This phenomenon was argued on the basis of calculations carried out by using the nonlocal density-functional (NLDF) theory formulated by Orsay-Paris (OP) Collaboration[2] and assuming that helium atoms and the metallic surface interact via the two-parameter (9,3) potential with parameters derived by Zaremba and Kohn.[3] Due to its extraordinary characteristic, the nonwetting to wetting transitions have been extensively studied. It was experimentally verified[4,5] that ^{4}He does not wet Cs at temperatures lower than the wetting temperature $T_w \simeq 2$ K. The contact angle θ_c defined by the Young relation

$$\cos\theta_c = (\sigma_{sv} - \sigma_{sl})/\sigma_{lv}, \tag{1}$$

where σ_{sv}, σ_{sl}, and σ_{lv} are the solid-vapor, solid-liquid, liquid-vapor surface tensions, respectively, was also measured.[6–8] The reported values of T_w and θ_c at $T=0$ are listed in Table I. On the contrary, experimental data[9] indicate that potassium is wetted by ^{4}He at $T=0$. The main features of the adsorption on K and Cs could be interpreted in recent theoretical works.[10–12] In these studies, the calculations have been carried out by utilizing the improved NLDF formulation proposed by the Orsay-Trento (OT) Collaboration[13] and the more recent adsorption potential derived by Chizmeshya, Cole, and Zaremba[14] (CCZ potential). It should be noticed that for a given substrate the CCZ potential is more attractive than the old (9,3) interaction.[3]

The situation for surfaces of Rb is still somewhat controversial. A number of measurements have been performed in order to determine the wetting behavior of the ^{4}He/Rb system. Several different experimental techniques have been applied,[9,15] the obtained wetting temperatures T_w are listed in Table I of Ref. 16. Although the authors claim that the preparation of the surface plays a crucial role, the most recent measurements indicate that planar Rb would be wetted by ^{4}He at $T=0$. The latter conclusion is supported by our calculations reported in Ref. 16. Just later on, Ancilotto, Faccin, and Toigo[12] (AFT) have extended the OT-NLDF to finite temperatures and examined the wetting of heavy-alkali metals by calculating separately σ_{sv}, σ_{sl}, and σ_{lv} as a function of temperature (this method requires determination of pressure). They found that in the $T=0$ limit ^{4}He wets K but not Rb. So, the authors of Ref. 12 by using essentially the same theoretical background as that of Ref. 16, but applying an alternative procedure for the analysis arrived at a different conclusion about wetting of Rb.

In order to shed some light on this issue, we shall complete previous studies of the adsorption of ^{4}He on alkali metals. In doing so, we shall examine a few different approaches within the OT-NLDF at $T=0$. The ground-state energy of ^{4}He confined by a potential $U_{\rm sub}(\mathbf{r})$ is

$$E_{\rm gs} = \frac{\hbar^2}{2m}\int d\mathbf{r}\,(\nabla\sqrt{\rho(\mathbf{r})})^2 + \int d\mathbf{r}\,\rho(\mathbf{r})\,e_{sc}(\mathbf{r}) + \int d\mathbf{r}\,\rho(\mathbf{r})\,U_{\rm sub}(\mathbf{r}), \tag{2}$$

where $e_{sc}(\mathbf{r})$ is the correlation energy per particle,[13]

$$\begin{aligned} e_{sc}^{\rm OT}(\mathbf{r}) = {} & \frac{1}{2}\int d\mathbf{r}'\,\rho(\mathbf{r}')\,V_l^{\rm OT}(|\mathbf{r}-\mathbf{r}'|) + \frac{c_4'}{2}[\overline{\rho}(\mathbf{r})]^2 \\ & + \frac{c_4''}{3}[\overline{\rho}(\mathbf{r})]^3 - \frac{\hbar^2}{4\,m}\alpha_s\int d\mathbf{r}'\,\rho(\mathbf{r}')F(|\mathbf{r}-\mathbf{r}'|) \\ & \times\left[1-\frac{\tilde{\rho}(\mathbf{r})}{\rho_{0s}}\right]\left(\frac{\boldsymbol{\nabla}\rho(\mathbf{r})}{\rho(\mathbf{r})}\right)\cdot\left(\frac{\boldsymbol{\nabla}\rho(\mathbf{r}')}{\rho(\mathbf{r}')}\right)\left[1-\frac{\tilde{\rho}(\mathbf{r}')}{\rho_{0s}}\right]. \end{aligned} \tag{3}$$

The two-body interaction is a truncated LJ potential, being $V_l^{\rm OT}(r) = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6]$ if $r \geqslant h_{\rm OT}$ and $V_l^{\rm OT}(r)=0$ otherwise. Quantity $\overline{\rho}(\mathbf{r})$ is the "coarse-grained density"

$$\overline{\rho}(\mathbf{r}) = \int d\mathbf{r}'\,\rho(\mathbf{r}')\,\mathcal{W}(|\mathbf{r}-\mathbf{r}'|) \tag{4}$$

with $\mathcal{W}(|\mathbf{r}-\mathbf{r}'|) = 3/(4\pi h_{\rm OT}^3)$ if $|\mathbf{r}-\mathbf{r}'| \leqslant h_{\rm OT}$ and zero otherwise. Furthermore, $\tilde{\rho}(\mathbf{r})$ is the weighted average

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TABLE I. Coefficients of the expansions for the energetics and contact angles at $T=0$ K, together with wetting temperatures.

System	NLDF Version	e_∞ [K]	σ_{sl} [K/Å^2]	σ_∞ [K/Å^2]	$\gamma_c\times10^3$ [K/Å^6]	$a_4\times10^3$ [K/Å^8]	$a_5\times10^3$ [K/Å^{10}]	θ_c $(T=0)$	T_w [K]	Ref.
^{4}He slab					$-1.28(\gamma_c^0)$					
Fit e	OT	-7.155		0.275	-1.38	-0.016	0.004			18
Slabs	AFT	-7.163		0.278						12
Fit e	$\alpha_s=0$	-7.165		0.279	-1.42	-0.026	0.004			PW [a]
^{4}He/Cs					$4.63(\gamma_c^0)$					
Expt.								48° ± 1°	2.00 ± 0.05	6
Expt.								25.5° ± 0.05°	1.90 ± 0.05	7
Expt.								21° ↔ 32°	1.75 ↔ 2.04	8
Fit e	OT	-7.151		0.012	4.49	-0.560	0.020	17°		PW
Slabs	AFT		-0.238	0.040				31°	2.1	12
Droplet								36°		11
Fit e	$\alpha_s=0$	-7.164		0.044	4.30	-0.537	0.019	33°		PW
Fit μ	$\alpha_s=0$	-7.162			3.73	-0.313	-0.003			PW
Eq. (15) [b]	$\alpha_s=0$			0.039				31°		PW
^{4}He/Rb					$4.72(\gamma_c^0)$					
Expt.								≃0°		9,15
Fit e	OT	-7.151		-0.010	4.71	-0.581	0.021	0°		16
Fit μ	OT	-7.152			4.27	-0.361	0.005	0°		16
Eq. (15) [b]	OT			-0.011				0°		16
Fit e	AFT	-7.164		-0.010	4.68	-0.576	0.021	0°		PW
Slabs	AFT		-0.260	0.018				21°	1.4	12
Fit e	$\alpha_s=0$	-7.165		0.022	4.54	-0.561	0.020	23°		PW
Fit μ	$\alpha_s=0$	-7.163			4.70	-0.540	0.013			PW
Eq. (15) [b]	$\alpha_s=0$			0.020				22°		PW
Fit e	(9,3) [c]	-7.151		0.063	4.06	-0.533	0.020	40°		PW
^{4}He/K					$4.81(\gamma_c^0)$					
Expt.								0°		9
Fit e	OT	-7.155		-0.056	4.48	-0.501	0.016	0°		10
Slabs	AFT		-0.306	-0.028				0°		12
Fit e	$\alpha_s=0$	-7.164		-0.027	4.81	-0.575	0.020	0°		PW
Fit μ	$\alpha_s=0$	-7.163			4.71	-0.475	0.007			PW
Eq. (15) [b]	$\alpha_s=0$			-0.027				0°		PW
Fit e	OP	-7.154		-0.013	4.46	-0.512	0.017	0°		10
^{4}He/Na					$5.60(\gamma_c^0)$					
Expt.								0°		24
Fit e	OT	-7.155		-0.361	6.30	-0.608	0.018	0°		PW

[a] PW stands for results obtained in the present work.

[b] Using coefficients from the fit of μ to Eq. (11).

[c] Using complete OT-NLDF and the new (9,3) potential with CCZ parameters (see text).

$$\tilde{\rho}(\mathbf{r})=\int d\mathbf{r}'\,\rho(\mathbf{r}')F(|\mathbf{r}-\mathbf{r}'|), \qquad (5)$$

where $F(|\mathbf{r}-\mathbf{r}'|)$ is the Gaussian function

$$F(|\mathbf{r}-\mathbf{r}'|)=\frac{1}{\pi^{3/2}\,\ell^3}e^{-|\mathbf{r}-\mathbf{r}'|^2/\ell^2}. \qquad (6)$$

Dalfovo *et al.*[13] obtained $h_{\mathrm{OT}}=2.190\,323$ Å, $c_4'=-24\,118.57$ K Å^6, and $c_4''=1\,858\,496$ K Å^9 by adjusting the energy per particle $e_B=-7.15$ K, the incompressibility $\mathcal{K}=27.2$ K, the equilibrium density $\rho_0=0.021836$ Å^{-3} of saturated bulk liquid ^{4}He at zero pressure,[17] while $\alpha_s=54.31$ Å^3, $\rho_{0s}=0.04$ Å^{-3}, and $\ell=1$ Å were fixed to reproduce the static response function (see Fig. 1 in Ref. 13). The authors of Ref. 12 got slightly different values $h_{\mathrm{OT}}=2.190\,35$ Å, $c_4'=-24\,258.88$ K Å^6, and $c_4''=1\,865\,257$ K Å^9 by fitting $e_B=-7.163$ K. In their extension to finite temperature a contribution due to a Bose vapor was added. The coefficients h_{OT}, c_4', and c_4'' as a function of T are listed in Table I of Ref. 12.

In the case of planar geometry, the system is translationally invariant in the x-y plane determined by the solid surface and symmetry is broken in the $z>0$ direction giving rise to a $\rho(z)$ for helium atoms yielded by

$$\left[-\frac{\hbar^2}{2\,m}\frac{d^2}{dz^2}+V_H(z)+U_{\mathrm{sub}}(z)\right]\sqrt{\rho(z)}=\mu\sqrt{\rho(z)}, \quad (7)$$

which also provides the chemical potential μ. The Hartree mean-field potential $V_H(z)$ is given elsewhere.[18]

The use of density functionals like that given by Eq. (3) for describing planar helium films is sometimes criticized in the literature. The main objection is that the parameters are fixed to give correctly the saturation properties of bulk ^{4}He, but the corresponding straightforward two-dimensional (2D) limit does not reproduce results specifically calculated for 2D homogeneous ^{4}He.[19] However, it should be noticed that density profiles yielded by the OT-NLDF agree well with those obtained from path integral Monte Carlo simulations. This feature is illustrated in Ref. 20, where $\rho(z)$ for ^{4}He/Li systems exhibiting pronounced layered structures are shown.

In practice, Eq. (7) is solved for a fixed coverage n_c defined as the number of particles N per unit area A,

$$n_c=\frac{N}{A}=\int_0^\infty dz\rho(z). \quad (8)$$

The integrodifferential problem (7) has been solved for coverages up to a sufficiently large $n_c^{\max}$. The precision and consistency of solutions for $\rho(z)$ and μ were checked by utilizing methods developed in Refs. 10 and 16.

We report a summary of new and previous results for adsorption on Cs, Rb, K, and Na. The energy per particle evaluated with the obtained solutions has been fitted to a polynomial expansion in terms of the inverse of coverage[18,21,22]

$$e=\frac{E_{\mathrm{gs}}}{N}=e_\infty+\frac{\sigma_\infty}{n_c}+\frac{\gamma_c}{n_c^3}+\frac{a_4}{n_c^4}+\frac{a_5}{n_c^5}. \quad (9)$$

In this approach the surface energy σ_A, the chemical potential μ, and the grand potential Ω obey[18,21,22]

$$\sigma_A=\left(\frac{\partial E_{\mathrm{gs}}}{\partial A}\right)_N=\sigma_\infty+3\frac{\gamma_c}{n_c^2}+4\frac{a_4}{n_c^3}+5\frac{a_5}{n_c^4}, \quad (10)$$

$$\mu=\left(\frac{\partial E_{\mathrm{gs}}}{\partial N}\right)_A=e_\infty-2\frac{\gamma_c}{n_c^3}-3\frac{a_4}{n_c^4}-4\frac{a_5}{n_c^5}, \quad (11)$$

$$\sigma_A=n_c(e-\mu)=(E_{\mathrm{gs}}-\mu\,N)/A=\Omega/A. \quad (12)$$

Here e_∞ may be identified with e_B and σ_∞ is the asymptotic total surface energy $\sigma_A(n_c\to\infty)=\sigma_{sl}+\sigma_{lv}$ (where σ_{sl} and σ_{lv} are also asymptotic surface tensions). The coefficient γ_c is mainly determined by the van der Waals tail $U_{\mathrm{sub}}^{(\mathrm{tail})}(z)\simeq -C_{\mathrm{tail}}/z^3$ of the adsorption potential. In the simplest approximation [see, e.g., Eq. (2.10) Ref. 22] yields $\gamma_c\simeq\gamma_c^0=\frac{1}{2}\rho_0^3(C_{\mathrm{tail}}-C_{\mathrm{tail}}^{\mathrm{He}})$, where $C_{\mathrm{tail}}^{\mathrm{He}}$ corresponds to a hypothetical ^{4}He substrate. Wetting requires that $\sigma_\infty=\Omega_\infty/A$ be the absolute minimum.[23] Furthermore, to ensure wetting the asymptotic areal isothermal compressibility κ_s must be positive,[21]

$$\frac{1}{\kappa_s}=A\left(\frac{\partial\sigma_A}{\partial A}\right)_N=-n_c\frac{d\sigma_A}{dn_c}=n_c^2\frac{d\mu}{dn_c}>0. \quad (13)$$

By following the procedure outlined in Ref. 22 the value of e_B was included in the fits of calculated e to Eq. (9). The extracted values of e_∞, σ_∞, γ_c, a_4, and a_5 are listed in Table I.

The result for the test ^{4}He/Na system yields wetting in agreement with experimental evidence[24] and previous calculations.[1,22,25] As expected, nonwetting was obtained for ^{4}He/Cs. Since at low temperatures σ_{sv} is negligibly small (see Table II in Ref. 12) Eq. (1) can be approximated by $\cos\theta_c=1-\sigma_\infty/\sigma_{lv}$. In the case of the complete original version of the OT-NLDF approach, this expression yields a value of θ_c which is slightly smaller than the lowest experimental limit.

A glance at Table I shows that the values $\sigma_\infty=\sigma_{sl}+\sigma_{lv}$ evaluated with data from Ref. 12 lie systematically above ours for all heavy substrates. However, it should be noticed that the differences are small when compared with the size of the absolute values of both σ_{lv} and σ_{sl}. A further analysis indicates that the differences $\sigma_{sl}(^4\mathrm{He/Cs})-\sigma_{sl}(^4\mathrm{He/K})\simeq 0.068$ K/Å^2 and $\sigma_{sl}(^4\mathrm{He/Rb})-\sigma_{sl}(^4\mathrm{He/K})\simeq 0.047$ K/Å^2 are almost identical to our $\sigma_\infty(^4\mathrm{He/Cs})-\sigma_\infty(^4\mathrm{He/K})$ and $\sigma_\infty(^4\mathrm{He/Rb})-\sigma_\infty(^4\mathrm{He/K})$. These results indicate that there is some sort of common perturbative difference.

Seeking for small effects we solved the controversial ^{4}He/Rb system by using the complete OT-NLDF with the parameters determined by AFT (Ref. 12) for $T=0$. As can be observed in Table I, this alternative choice did not introduce any sizeable effect.

Next, we studied the adsorption of ^{4}He onto all heavy substrates by using the AFT parameters[12] for $T=0$ but assuming $\alpha_s=0$. This corresponds to eliminate the gradient-gradient contribution which leads to the correct static response function for bulk ^{4}He.[13] The results quoted in Table I show that all these new values of σ_∞ move upwards and lie surprisingly close to the sum $\sigma_{sl}+\sigma_{lv}$ estimated with data of Ref. 12. The most important qualitative change is that in this approach ^{4}He does not wet rubidium because σ_∞ is positive. Furthermore, the contact angle evaluated for the ^{4}He/Cs system now agrees with the values of Refs. 11 and 12. The consistency of results obtained in this approximation was checked by evaluating σ_∞ with an expression derived from the Gibbs-Duhem relation[16]

$$\sigma_A(n_c)-\sigma_A(0)=\int_0^{n_c}[\mu(n_c')-\mu(n_c)]dn_c'. \quad (14)$$

Since $\sigma_A(0)=0$, in the limit $n_c\to\infty$, one gets

$$\sigma_\infty=\int_0^\infty[\mu(n_c)-e_\infty]dn_c=\int_0^{n_c^{\max}}[\mu(n_c)-e_\infty]dn_c -\gamma_c\left(\frac{1}{n_c^{\max}}\right)^2-a_4\left(\frac{1}{n_c^{\max}}\right)^3-a_5\left(\frac{1}{n_c^{\max}}\right)^4. \quad (15)$$

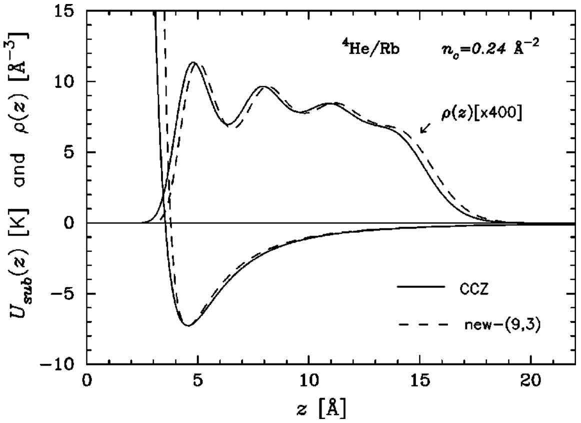

FIG. 1. Adsorption potential $U_{\mathrm{sub}}(z)$ and density profile $\rho(z)$ as a function of the distance from the substrate. Results for the CCZ and new (9,3) potentials are displayed.

In Table I we included results computed with sets of parameters e_∞, γ_c, a_4, and a_5 extracted from fitting μ to Eq. (11). Note that the latter equation does not have explicit information on σ_∞. These new values are quite similar to that determined by fitting the energy to Eq. (9) supporting consistency. Moreover, as one would expect, when setting $\alpha_s = 0$ the solutions move towards those yielded by the OP-NLDF. This is illustrated by the values of σ_∞ for the ^{4}He/K case listed in Table I.

For the sake of completeness, the ^{4}He/Rb system was solved by adopting a new (9,3) potential evaluated with parameters from CCZ. Such a potential has been already used for studying adsorption on alkali metals in Refs. 11 and 20. Figure 1 shows that both the new (9,3) and the complete CCZ potentials have the same depth and long-range behavior, however, the (9,3) exhibits a steeper hard core and a narrower well. Calculations performed within the original OT-NLDF indicate nonwetting for this (9,3) interaction (see Table I). Since a steeper hard core pushes the helium film more outwards, see Fig. 1, the integral of $\rho(z)U_{\mathrm{sub}}(z)$ evaluated with the (9,3) potential collects less negative energy than in the CCZ case. This feature explains the trend of the value of σ_∞.

In summary, a detailed comparison between present results for wetting of alkali metals by ^{4}He and those of Ref. 12 is performed. It was found that in the critical case of the adsorption on an Rb substrate, the gradient-gradient term in the NLDF and the softness of $U_{\mathrm{sub}}(z)$ play crucial roles.

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