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Surface properties of liquid ^{3}He and ^{4}He: A density-functional approach

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We present a comparative study of the free surface of ^{3}He and ^{4}He at zero temperature using phenomenological density functionals. The functional for each fluid is adjusted on experimentally known properties; the surface profile is then obtained variationally. Simple analytical formulas are derived for the relevant quantities characterizing the surface. Our results are compared with other available theoretical calculations.

I. INTRODUCTION

Surface properties of quantum liquids have constituted a field for numerous interesting investigations, both experimental and theoretical.[1] In ^{3}He and ^{4}He, the surface tension is known over a range of temperatures (see, for example, Refs. 2 and 3 and references therein); however, the density profile of the free surface of liquid helium remains to be determined experimentally.

The free surface of liquid ^{4}He has been the object of several theoretical studies. Some attempts have been made to develop theories for the wave function of the ground state. The early relevant works in this direction are by Shih and Woo,[4] and Chang and Cohen.[5] Recently, more sophisticated approaches to inhomogeneous Bose systems have been proposed.[6-10] In particular, the results for the binding energy of ^{4}He clusters obtained in Ref. 10, carrying out variational Monte Carlo calculations, extrapolate well to the experimental values for the volume and surface energy of liquid ^{4}He. In the case of large clusters, the above calculations are then expected to provide a realistic description of the free-surface profile too. Alternative approaches to inhomogeneous Bose systems have been developed using density-functional methods.[11-16] Ebner and Saam (Ref. 14 and references therein) have stressed the importance of the surface zero-point motion in renormalizing the surface properties of liquid ^{4}He. These effects can be taken into account by including nonlocal components in the energy density functional as discussed in Ref. 13. It is interesting to note that when such renormalization effects are properly included in the density-functional approach, the resulting predictions for the surface properties turn out to be in quite good agreement with the ones given by microscopic calculations; in particular, values of 6–7 Å are predicted for the surface thickness.[12,13]

The free surface of liquid ^{3}He has received much less attention in the literature. This is due to the enormous difficulties in carrying out microscopic calculations in inhomogeneous Fermi systems. The first microscopic calculations using the correlated basis functions were performed by Buchan and Clark,[17] Mackie and Woo,[18] and Senbetu and Woo.[19] More recently, accurate variational Monte Carlo calculations of ^{3}He clusters containing up to 240 atoms have been made.[10,20,21] The surface tension extracted in these works, by liquid-drop fits to the calculated binding energies, agrees well with the experimental value. Density-functional approaches have been extensively used to investigate the surface properties of Fermi systems (metals,[22] and atomic nuclei[23]) However, only recently such methods have been applied to the study of the surface of liquid ^{3}He.[24,25] In particular, the results of Refs. 24 and 25 for ^{3}He clusters agree well with the findings of Ref. 10 concerning both the binding energies and the density profiles.

It is the purpose of the present work to develop a unified density-functional approach to treat the free surface of liquid ^{4}He and ^{3}He. The density functional we use is of phenomenological nature and has been already employed in an extensive way to investigate the properties of atomic nuclei.[23] The same approach can be generalized to the time-dependent case to investigate the propagation of collective phenomena. An application to the study of surface ripplons is given in Ref. 26.

The explicit form of the energy density functional is discussed in Sec. II. Our choice includes local as well as nonlocal components, the latter being crucial for a correct description of the surface properties. We also discuss the relations between the kinetic-energy density and the diagonal density used to investigate the surface of ^{3}He and ^{4}He. In Secs. III A and III B we derive Euler-type equations for the density of ^{3}He and ^{4}He, respectively. After having adjusted the surface-free parameters entering the energy functional to reproduce the surface tension, we determine the surface profile of both isotopes, and find useful relations between the surface thickness, the surface tension, and the bulk compressibility. Our results are compared with other recent theoretical calculations of the surface of quantum liquids. Finally we draw our conclusions in Sec. IV.

II. DENSITY FUNCTIONALS

Our starting point lies in the assumption that the energy of the system can be written as a functional of the one-body density matrix. This is supported by a well-known theorem due to Hohenberg and Kohn,[22] which, however, does not provide a practical way of constructing the functional. In the present work, we shall use the following form for the expectation value of the energy in a time-reversal invariant state at zero temperature:

$$E=\int \mathcal{H}(\rho,\tau)d^3r$$
$$=\int\left[\frac{h^2}{2m^*(\rho)}\tau+\tfrac{1}{2}b\rho^2+\tfrac{1}{2}c\rho^{2+\gamma}+d(\nabla\rho)^2\right]d^3r\ , \tag{1}$$

where

$$\rho(r)=\left\langle 0\left|\sum_i\delta(r-r_i)\right|0\right\rangle\ ,$$
$$\tau(r)=-\tfrac{1}{2}\left\langle 0\left|\sum_i\nabla_i^2\delta(r-r_i)+\text{H.c.}\right|0\right\rangle$$

are the number density and kinetic-energy density, respectively, and $|0\rangle$ denotes the ground state of the system. Functionals of the form (1) have been extensively employed to describe properties of atomic nuclei and can be derived using a phenomenological interaction of Skyrme type.[23]

The coefficients entering Eq. (1) are determined by fitting a set of experimentally known quantities, and we shall now examine successively the different components of the functional. Concerning the effective mass, in the case of ^{3}He, we use the following form:

$$\frac{\hbar^2}{2m^*(\rho)}=\frac{\hbar^2}{2m}\left[1-\frac{\rho}{\rho_c}\right]^2\ , \tag{2}$$

with $\rho_c=0.041$ Å^{-3}; it was shown in Ref. 27 that Eq. (2) accounts very accurately for the variation of the specific heat with pressure. In the case of liquid ^{4}He, there is no sufficient clear experimental information on the value of the effective mass, so we have chosen $m^*=m$.

The kinetic-energy density $\tau(r)$ requires different treatments for the cases of ^{3}He and ^{4}He, due to the different statistics. For liquid ^{3}He, we use a Thomas-Fermi approximation extended to inhomogeneity terms,

$$\tau(r)=\alpha\rho^{5/3}+\beta\frac{(\nabla\rho)^2}{\rho}+\delta\nabla^2\rho\ , \tag{3}$$

with $\alpha=\frac{3}{5}(3\pi^2)^{2/3}$, $\beta=\frac{1}{18}$, and $\delta=\frac{1}{3}$.[28] Such a functional can be derived through an $\hbar$ expansion of the energy of a system of fermions moving in a smoothly varying local potential. The coefficient $\beta=\frac{1}{18}$ that we choose simulates higher-order corrections involving the higher-order density dependence of the effective mass;[29] due to the large value of m^*, the final results are rather insensitive to the precise value of β. A detailed comparison between quantum and semiclassical calculations of finite ^{3}He clusters is presented in Ref. 25; the agreement is found very satisfactory; in particular, neither shell effects nor Friedel oscillations are found in the quantum calculation, so that the density profiles turn out to be identical in the semiclassical and in the quantum calculations; this indicates that the same semiclassical approach should provide a reliable description of the semi-infinite medium.

In the case of ^{4}He, the system will be discussed as a Bose condensate; then the kinetic energy arises only from the inhomogeneity of the diagonal density and one has

$$\tau(r)=\frac{1}{4}\frac{(\nabla\rho)^2}{\rho}\ . \tag{4}$$

Let us now turn to the other terms entering Eq. (1); the term in b is attractive and reflects the attractive character of the interatomic force at large distances, i.e., at low density of the medium. The c term is repulsive, as is the interaction at short distance, and it dominates at high densities. The coefficients b, c, and γ are determined so as to reproduce the equilibrium density, the energy per particle, and the compressibility K of the uniform liquid.

The nonlocality term in $(\nabla\rho)^2$ reflects the repulsive interaction at the surface, and is chosen to reproduce the experimental value of the surface tension at zero temperature. Such a term, which has not always been considered in previous works on the free surface of ^{4}He, incorporates in a phenomenological way the contribution to the surface tension of the zero-point energies of the surface ripplons.[1,14] As we shall see, it plays a crucial role in the determination of the surface tension.

It is interesting at this point to compare the energy per particle for liquid ^{3}He and ^{4}He using dimensionless coordinates, namely

$$\frac{E/N}{(E/N)_0}\ \text{and}\ \frac{\rho}{\rho_0}\ ,$$

where the indice 0 indicates zero-pressure (i.e., saturation) quantities. This is done in Fig. 1(a). One sees that both curves differ essentially at low density, where the kinetic energy for ^{3}He is dominant over the attractive force up to a ratio $\rho/\rho_0\simeq 0.15$. For higher densities the potential energy becomes dominant, also because the increase of the effective mass reduces the contribution of

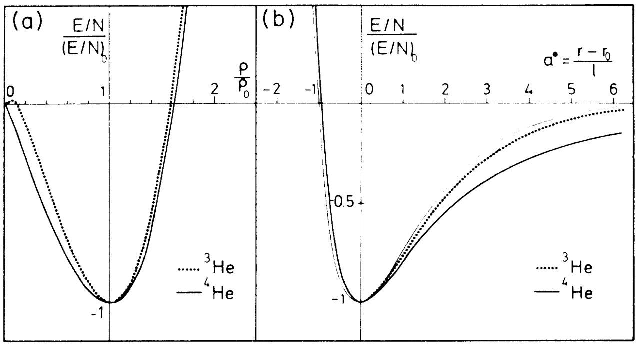

FIG. 1. (a) Plot of the energy per particle for liquid ^{3}He (dotted curve) and ^{4}He (solid curve) as a function of the density using dimensionless coordinates. The indice 0 refers to saturation quantities. (b) Plot of the energy per particle for liquid ^{3}He (dotted curve) and ^{4}He (solid curve) as a function of the mean distance between the atoms relative to the scaling length defined in Sec. II. The thin curve is taken from Fig. 1 of Ref. 30.

the kinetic energy to the total energy of liquid ^{3}He; indeed, for $\rho/\rho_0 > 1$ both curves are almost identical.

The differences between the two curves below saturation density persists if one uses a scaled abscissa, as proposed in Ref. 30, in order to exhibit a universal binding-energy–distance relation. Let us introduce the incompressibility of the liquid as

$$\frac{1}{K\rho} = \frac{\delta}{\delta\rho}\left[\rho^2\frac{\delta(E/N)}{\delta\rho}\right] \tag{5}$$

(it has the dimension of an energy); the authors of Ref. 30 introduce a scaled abscissa a^* by

$$a^* = \frac{r - r_0}{l} , \tag{6a}$$

where r is the interparticle distance at the density ρ, r_0 the equilibrium distance, and l a length scale defined as

$$l = r_0\left[\frac{(E/N)_0}{9/K\rho_0}\right]^{1/2} . \tag{6b}$$

The resulting curves are plotted in Fig. 1(b), where we also show for comparison the curve presented in Fig. 1 of Ref. 30, representing the binding-energy–distance relation for molecular and metallic systems. Although at short distances the scaling accounts satisfactorily for the small differences seen above saturation density in Fig. 1(a), the effect of the different statistics obeyed by liquid ^{3}He and ^{4}He shows up clearly for $a^* > 0$. The scaling length (6b) accounts for the fact that different physical systems have different compressibilities; however, the relative variation of K with density (related to the Grüneisen constant) may also be quite different for various systems, and indeed we think that this fact explains the differences seen in Fig. 1(b) between the curve of Ref. 30 and the curve that we obtain for ^{3}He. The value of the ratio of surface-to-volume energy is partly related to the same question. The value proposed in Ref. 30 ($\simeq 0.82$) does not apply for ^{3}He or ^{4}He, quantitatively. This ratio $\simeq 2.4$ for ^{4}He and $\simeq 3.4$ for ^{3}He ($\simeq 1.2$ for atomic nuclei). We shall indeed derive below a relation between the compressibility, the surface tension, and the surface thickness. Such relations have already been investigated in the literature.[28,31,32]

We now turn to the description of the semi-infinite medium. Starting with a finite system with spherical symmetry and taking the limit $N \to \infty$, we shall define in a general way the surface, curvature, etc. contributions to the total energy. Due to the saturating character of the interaction, one can isolate the contribution to the energy of the surface; one subtracts from the total energy a volume contribution equal to the energy the system would have if the energy density per particle kept a constant value equal to its value e in the center,

$$E = eN + \int (\mathcal{H} - e\rho)\, d^3r . \tag{7}$$

Let us introduce the sharp equivalent radius R_s, related to the central density by

$$N = \tfrac{4}{3}\pi R_s^3 \rho_c , \tag{8}$$

and a new variable $z = r - R_s$; Eq. (7) takes the form (for a spherical geometry)

$$\begin{aligned} E = eN &+ 4\pi R_s^2 \int_{-R_s}^{+\infty} (\mathcal{H} - e\rho)\, dz \\ &+ 8\pi R_s \int_{-R_s}^{+\infty} (\mathcal{H} - e\rho) z\, dz \\ &+ 4\pi \int_{-R_s}^{+\infty} (\mathcal{H} - e\rho) z^2 dz . \end{aligned} \tag{9}$$

The surface energy, the curvature energy, and the constant term (independent of R_s) appear as surface moments of the surface energy density $\mathcal{H} - e\rho$.

In the limit N (or R_s) $\to \infty$, the central density tends to the saturation density ρ_0, and e tends to the saturation energy of the uniform medium $(E/N)_0 = \mu$, which is stationary with respect to ρ_0. Hence, in this limit, minimizing the energy amounts to minimizing the surface tension, defined as

$$\sigma = \lim_{S \to \infty} (E - eN)/S = \int (\mathcal{H} - \mu\rho)\, dz . \tag{10}$$

The semi-infinite profile is thus determined by the one-dimensional Euler equation

$$\frac{\delta\mathcal{H}}{\delta\rho} = \mu . \tag{11}$$

In the $N \to \infty$ limit, the equivalent sharp radius R_s is such that

$$\int_{-\infty}^{R_s} [\rho_0 - \rho(z)]\, dz = \int_{R_s}^{+\infty} \rho(z)\, dz . \tag{12}$$

Introducing the radius constant r_0,

$$r_0 = \left[\frac{3}{4\pi\rho_0}\right]^{1/3} ,$$

the surface energy coefficient is given by

$$a_s = 4\pi r_0^2 \sigma . \tag{13}$$

From the knowledge of $\rho(z)$, one can calculate the curvature energy coefficient a_c and the constant term a_0 as

$$a_c = 8\pi r_0 \int_{-\infty}^{+\infty} (\mathcal{H} - e\rho) z\, dz , \tag{14}$$

$$a_0 = 4\pi \int_{-\infty}^{+\infty} (\mathcal{H} - e\rho) z^2 dz . \tag{15}$$

III. SURFACE PROPERTIES

A. Liquid ^{3}He

Inserting Eqs. (2) and (3) into (1), one gets for the energy density

$$\begin{aligned} \mathcal{H}(\rho, \nabla\rho) &= h(\rho) + \frac{\hbar^2}{2m}\beta\frac{(\nabla\rho)^2}{\rho} + d_1(\nabla\rho)^2 + d_2\rho(\nabla\rho)^2 , \\ h(\rho) &= \frac{\hbar^2}{2m}\left[1 - \frac{\rho}{\rho_c}\right]^2 \alpha\rho^{5/3} + \frac{b}{2}\rho^2 + \frac{c}{2}\rho^{(2+\gamma)} , \\ d_1 &= d - \frac{\hbar^2}{m}\frac{\beta - \delta}{\rho_c} , \\ d_2 &= \frac{\hbar^2}{2m}\frac{\beta - 2\delta}{\rho_c^2} . \end{aligned} \tag{16}$$

TABLE I. Values of the parameters entering the energy density functionals.

	^{3}He	^{4}He
b (K Å^3)	-6.8300×10^2	$-8.888\,10\times 10^2$
c (K Å$^{3(1+\gamma)}$)	$1.405\,057\times 10^6$	$1.045\,54\times 10^7$
γ	2.1	2.8
ρ_c (Å^{-3})	4.06×10^2	∞
d (K Å^5)	2.222×10^3	2.383×10^3

The Euler equation (11) reads as

$$\frac{dh}{d\rho}+\frac{\hbar^2}{2m}\beta\left[\frac{\rho'^2}{\rho^2}-2\frac{\rho''}{\rho}\right]-2d_1\rho''-d_2(\rho'^2+2\rho\rho'')=\mu\ . \tag{17}$$

This equation, after multiplying by ρ', can be integrated once to yield

$$h(\rho)-\frac{\hbar^2}{2m}\beta\frac{\rho'^2}{\rho}-d_1\rho'^2-d_2\rho\rho'^2=\mu\rho\ ,$$

or

$$\frac{d\rho}{dz}=-\left[\frac{h(\rho)-\mu}{(\hbar^2/2m)\beta+d_1+d_2\rho}\right]^{1/2}. \tag{18}$$

The integrability of Eq. (17) is due to the fact that the variable z does not appear explicitly in the functional; Eq. (18) can be interpreted using the following analogy: the functional which is being minimized is of the form

$$I\{\rho\}=\int\left[F(\rho)+G(\rho)\left[\frac{d\rho}{dz}\right]^2-\mu\rho\right]dz\ .$$

$I\{\rho\}$ can be considered as an action integral for a dynamical system of a point particle in a potential $\mu\rho-F(\rho)$, where ρ represents the position of the particle and z represents the time; the mass of the particle depends on its position and is given by $m(\rho)=\frac{1}{2}G(\rho)$. The Euler equation (11) is formally identical to the Lagrange equation obtained from $I\{\rho\}$, and Eq. (18) is just equivalent to the conservation of energy.

Using Eq. (18), the surface tension can be written as

$$\sigma=2\int_0^{\rho_0}\left[\frac{\hbar^2}{2m}\beta+d_1\rho+d_2\rho^2\right]^{1/2}\left[\frac{h(\rho)}{\rho}-\mu\right]^{1/2}d\rho\ . \tag{19}$$

Equation (19) does not require the knowledge of the density profile $\rho(z)$. It can thus be used to fix the still undetermined coefficient d, so as to reproduce the experimental value of the surface tension (see Tables I–II).

TABLE II. Bulk and surface properties at saturation, obtained using the parameters of Table I.

	^{3}He	^{4}He
ρ_0 (Å^{-3})	1.6347×10^2	2.1836×10^2
E/N (K)	-2.49	-7.15
$1/K\rho_0$ (K)	12.1	27.2
σ (K Å^{-2})	0.113	0.274

TABLE III. Density of semi-infinite medium, relative to saturation density ρ_0, as function of distance across the surface. The origin is taken at half-density.

z (Å)	^{3}He	^{4}He
-11	0.990	0.997
-10	0.985	0.994
-9	0.978	0.991
-8	0.967	0.985
-7	0.952	0.976
-6	0.930	0.960
-5	0.898	0.936
-4	0.853	0.897
-3	0.793	0.839
-2	0.713	0.755
-1	0.615	0.640
0	0.500	0.500
1	0.376	0.346
2	0.256	0.201
3	0.150	0.089
4	0.071	0.025
5	0.023	0.004
6	0.004	0.001

Equation (18) can now be solved numerically. The result is given in Table III and shown in Fig. 2, where we also plot the mean field

$$U(z)=\frac{\delta\mathcal{H}}{\delta\rho}=\frac{d}{d\rho}\left[\frac{\hbar^2}{2m^*(\rho)}\right]\tau(\rho)+b\rho+\frac{2+\gamma}{2}c\rho^{(1+\gamma)}$$

$$-2d_1\rho''-d_2(\rho'^2+2\rho\rho'')\ . \tag{20}$$

whose asymptotic value is just

$$U(-\infty)=\mu-\frac{\hbar^2}{2m^*}\frac{d\tau}{d\rho}=\mu-\frac{\hbar^2}{2m}\left[1-\frac{\rho_0}{\rho_c}\right]\tfrac{5}{3}\alpha\rho_0^{2/3}\ . \tag{21}$$

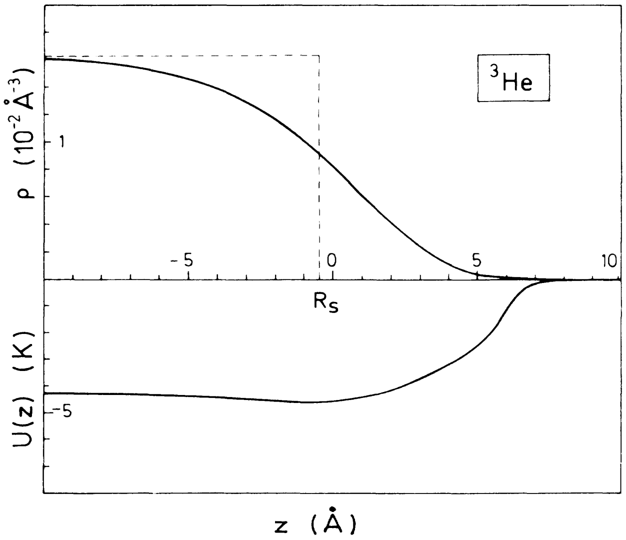

FIG. 2. Surface profile of liquid ^{3}He and the corresponding mean field [Eq. (20)]. The origin is taken at half bulk density. The equivalent sharp radius R_s is -0.43 Å.

We find that the surface thickness t_{10-90}, defined as the distance separating the points where the density has decreased by 10% and 90%, is equal to 8.3 Å. This value is in excellent agreement with the calculation of Ref. 10 on large clusters of ^{3}He using the interatomic Aziz potential with correlated wave functions. We also find that the skewness of the density profile is large. This property can be understood as follows.[28] From Eq. (16), it appears that the density reaches exponentially its asymptotic bulk value,

$$\frac{\rho(z)-\rho_0}{\rho_0} \propto \exp(z/a_{\rm in}) , \tag{22}$$

with

$$a_{\rm in}=\left[2K\rho_0\left[\frac{\hbar^2}{2m}\beta+d_1\rho_0+d_2\rho_0^2\right]\right]^{1/2}$$

$$\simeq(2Kd_1)^{1/2}\rho_0 . \tag{23}$$

The outward exponential falloff of the density is governed by the chemical potential μ,

$$\rho(z)\propto\exp\left[\frac{z}{a_{\rm out}}\right] , \tag{24}$$

$$a_{\rm out}=\left[-\frac{\hbar^2}{2m}\frac{\beta}{\mu}\right]^{1/2} . \tag{25}$$

Numerically, one finds $a_{\rm in}=2.37$ Å (which would correspond to a surface thickness of $\simeq 10$ Å) and $a_{\rm out}=0.42$ Å. The surface profile is thus highly nonsymmetric around the inflexion point of the density distribution. [The true exponential decay of the density is asymptotically governed by Eq. (25) with $\beta=\frac{1}{4}$, rather than with the value $\beta=\frac{1}{18}$ used in the present work. However, this is true in the outermost region of the tail, which is not relevant for the calculation of the surface thickness.]

The above results suggest the following approximate shape for the density profile in liquid ^{3}He:

$$\rho(z)=\frac{\rho_0}{[1+\exp(z/a_{\rm in})]^{\nu}} . \tag{26}$$

The surface thickness t_{10-90} resulting from Eq. (26) is given by

$$t_{10-90}=a_{\rm in}\ln\left[\frac{10^{1/\nu}-1}{(\frac{10}{9})^{1/\nu}-1}\right] . \tag{27}$$

We find that with the exact value of $a_{\rm in}$ and a value of $\nu=2.8$, Eq. (26) reproduces well the self-consistent result. Equation (26) clearly shows that due to the large value of ν, the surface thickness is merely determined by the value of $a_{\rm in}$.

A useful approximate expression for σ can be derived if one uses a cubic expansion of $h(\rho)/\rho$ around saturation density. The result is

$$\sigma\simeq\tfrac{8}{15}\rho_0\left[\frac{d_1}{2K}\right]^{1/2}(1-\tfrac{2}{7}\Gamma) , \tag{28}$$

where Γ is related to the Grüneisen-like constant G,

$$G=\frac{\rho}{v_s}\frac{dv_s}{d\rho} \tag{29}$$

[v_s denotes the sound velocity $\sqrt{1/(mK\rho)}$] by

$$\Gamma=(2G-4)/3 . \tag{30}$$

By use of Eq. (23) one gets a simple relation between σ, K, $a_{\rm in}$, and Γ,

$$\sigma=\frac{4}{15}\frac{a_{\rm in}}{K}(1-\tfrac{2}{7}\Gamma) . \tag{31}$$

Next we have calculated the curvature coefficient a_c, which we find equal to 5.5 K. Collecting the different coefficients, we can write a mass formula for He clusters as a function of the number of atoms N,

$$\begin{aligned} E&=a_vN+a_sN^{2/3}+a_c^*N^{1/3}+a_0^* , \\ a_c^*&=a_c-\tfrac{1}{9}K\rho_0a_s^2 , \end{aligned} \tag{32}$$

where $a_v=(E/N)_0$ represents a volume energy.

The constant term a_0^* is the sum of a_0, which we find equal to 8.6 K, and of several corrections from compressibility effects. These corrections cannot be calculated with enough accuracy, and it is more reliable to adjust a_0^* to calculated clusters. The value $a_0^*=-19.8$ K was found in Ref. 25.

Finally, in order to evaluate the importance of the zero-point surface motion, we have set to zero the coefficient d in Eq. (1). The resulting value for the surface tension is $\sigma=0.0308$ K Å^{-2}, and for the surface thickness it is $t=2.15$ Å; hence both quantities are reduced by a factor of almost 4. The curvature energy is reduced to 0.58 K (instead of 5.5 K).

B. Liquid ^{4}He

In this case, the energy density reads

$$\begin{aligned} \mathcal{H}(\rho,\nabla\rho)&=h(\rho)+\frac{\hbar^2}{2m}\frac{1}{4}\frac{(\nabla\rho)^2}{\rho}+d(\nabla\rho)^2 , \\ h(\rho)&=\frac{b}{2}\rho^2+\frac{c}{2}\rho^{(2+\gamma)} . \end{aligned} \tag{33}$$

The Euler equation, integrated once as in Eq. (18), reads

$$h(\rho)-\frac{\hbar^2}{2m}\frac{1}{4}\frac{\rho'^2}{\rho}-d\rho'^2=\mu , \tag{34}$$

and the surface tension can be written as

$$\sigma=2\int_0^{\rho_0}\left[\frac{1}{4}\frac{\hbar^2}{2m}+d\rho\right]^{1/2}\left[\frac{h(\rho)}{\rho}-\mu\right]^{1/2}d\rho . \tag{35}$$

The value of d which allows us to reproduce the experimental surface tension is given in Table I. Equation (22) holds also in the present case, with d_1 being replaced by d.

The surface profile, obtained by solving numerically Eq. (34), is given in Table III and shown in Fig. 3, together with the mean field,

$$U(z)=b\rho+\frac{2+\gamma}{2}c\rho^{(1+\gamma)}-2d\nabla^2\rho \ . \tag{36}$$

The surface thickness t_{10-90} is found to be 7 Å. This value agrees with all previous calculations which try to incorporate correlation effects in the surface;[10,13,14] it is significantly larger than that found by Shih and Woo,[4] partly because these authors impose a profile with zero skewness; indeed, as in the case of ^{3}He, the skewness of the surface profile is large, and most of the surface thickness is related to the inward surface diffuseness $a_{\rm in}$,

$$a_{\rm in}=\left[2K\rho_0\left[\frac{\hbar^2}{2m}\frac{1}{4}+d\rho_0\right]\right]^{1/2}\simeq(2Kd)^{1/2}\rho_0 \ . \tag{37}$$

The outward diffuseness is fixed by the chemical potential μ

$$a_{\rm out}=\left[-\frac{\hbar^2}{2m}\frac{1}{4\mu}\right]^{1/2} . \tag{38}$$

Numerically, one finds $a_{\rm in}=1.96$ Å and $a_{\rm out}=0.46$ Å; the profile is well produced by the simple parametrization (26), if one uses the exact value of $a_{\rm in}$ with $\nu=2.5$.

Of course Eq. (31), which relates σ, K, $a_{\rm in}$, and Γ, holds also for liquid ^{4}He. The term in Γ represents a correction to the leading term; by neglecting it, one can easily relate, as we shall now see, surface properties of ^{3}He to those of ^{4}He.

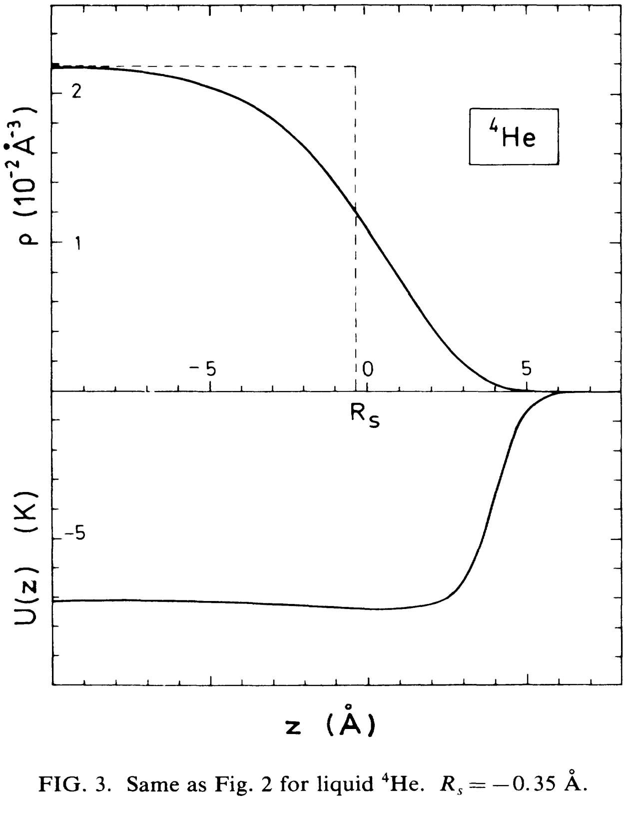

FIG. 3. Same as Fig. 2 for liquid ^{4}He. $R_s=-0.35$ Å.

Since $a_{\rm in}$ is much larger than $a_{\rm out}$, the ratio of the surface thicknesses of liquid ^{3}He and ^{4}He is close to the ratio of the *inward* diffusenesses $a_{\rm in}$, which in turn can be estimated from Eq. (31),

$$\frac{t(^3{\rm He})}{t(^4{\rm He})}\simeq\frac{a_{\rm in}(^3{\rm He})}{a_{\rm in}(^4{\rm He})}\simeq\frac{\sigma(^3{\rm He})}{\sigma(^4{\rm He})}\frac{K(^3{\rm He})}{K(^4{\rm He})}\simeq1.2 \ . \tag{39}$$

This result agrees well with the exact calculation of $t(^3{\rm He})/t(^4{\rm He})$.

Similar arguments allow one to understand the value found for the curvature energy coefficient of liquid ^{4}He, $a_c=10.9$ K; from Sec. (4.1) of Ref. 28, we see that

$$a_c\propto r_0\rho_0^2 d \ . \tag{40}$$

Together with Eqs. (31) and (37), this implies

$$a_c\propto\frac{r_0 a_{\rm in}^2}{K}\propto\sigma r_0 a_{\rm in} \ , \tag{41}$$

or

$$\frac{a_c(^4{\rm He})}{a_c(^3{\rm He})}\simeq1.83 \ , \tag{42}$$

which is in fair agreement with the ratio of the calculated values.

Next, a mass formula similar to Eq. (32) can be used to calculate the binding energy of ^{4}He clusters. The constant term a_0 is equal to 14.6 K, and here also, the constant term a_0^* to be used in the mass formula includes additional corrections from compressibility effects. The value found in Ref. 25 was $a_0^*=-29$ K.

Finally, as in the case of ^{3}He, in order to characterize the importance of the zero-point surface motion, we have repeated our calculations with a value of $d=0$ in Eq. (1). The result for the surface tension is $\sigma=0.0801$ K Å^{-2}, and for the surface thickness it is $t=1.75$ Å. Here also, the values are reduced by a factor of almost 4. The curvature energy is now equal to 1.45 K (instead of 10.9 K). These results are in agreement with those of Atkins[34] who first pointed out the importance of zero-point energies to the surface tension of liquid ^{4}He at zero temperature; our results also agree with those of Ebner and Saam (see, in particular, Sec. V of Ref. 14).

To summarize this section, we note that the results obtained in the framework of the energy density formalism show an overall agreement with the more fundamental calculations of Pandharipande *et al.*[10] One should, however, notice that the asymptotic behavior of our self-consistent one-body potential (36) is wrong; if the interatomic potential is proportional to $1/z^6$, the mean field should go to zero as an inverse power law $\sim -1/z^3$, whereas it has in our case an exponential behavior. Thus our one-body potential cannot be used to analyze the scattering of ^{4}He atoms by the free surface at low energy. Indeed, as described in Ref. 35, the scattering is essentially sensitive to this asymptotic behavior.

IV. CONCLUSION

In the present work we have developed a density-functional formalism for investigating the surface properties of liquid ^{3}He and ^{4}He. The energy density is of phenomenological nature and contains local as well as nonlocal components which account for the renormalization effects of the zero-point motion of the surface. These effects are found to be large both in liquid ^{3}He and ^{4}He. The free parameters of the model have been adjusted to reproduce relevant bulk properties known from experiment as well as the experimental value for the surface tension. Similar forms for the energy density have been extensively employed to investigate the properties of other quantum systems (atomic nuclei).

Using suitable relations between the kinetic-energy density and the diagonal density we have derived Euler-type equations and have explicitly investigated the density profile of ^{3}He and ^{4}He. We have found that the surface thickness (10–90% density) is $\simeq 7$ Å in the case of ^{4}He and 8.3 Å in ^{3}He, in good agreement with the recent result of Ref. 10, where clusters of ^{3}He and ^{4}He have been investigated using a variational Monte Carlo method. We have also derived useful relations between the surface thickness, the surface tension, and the bulk compressibility. Similar relations have already been discussed in the literature in the context of different quantum systems (metals, nuclei).

A major advantage of the present method is its simplicity. It can be extended to investigate surface effects in ^{3}He and ^{4}He mixtures[33] and helium films. It can also be extended to analyze the propagation of surface excitations.[26]

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