

Inhomogeneous Liquid ^4He : A Density Functional Approach with a Finite-Range Interaction

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The properties of liquid helium-4 are investigated in a mean-field approximation using a finite-range effective interaction. The long-range part (i.e., $r > 2.556 \text{ \AA}$) coincides with the standard Lennard-Jones potential and the hard core is phenomenologically renormalized. The model reproduces the static polarizability of bulk liquid, as well as the actual surface energy of helium at zero temperature. The surface width is found to be 5.8 \AA . A comparison is made with the earlier work of Ebner and Saam.

1. INTRODUCTION

The successful use of density-functional methods in the context of nuclear physics has renewed the interest in similar studies on liquid helium. Various properties of the liquid ^3He , ^4He , and ^4He - ^3He mixtures have been investigated in a number of recent works, and the comparison with *ab initio* calculations, when possible has been found satisfactory. The general framework of these studies in the time-dependent Hartree-Fock theory, together with the use of effective interactions of Skyrme type, the parameters of which are fitted so as to reproduce various properties of the actual system under investigation.

Reference 1 presented a study of surface properties of liquid ^3He and liquid ^4He at zero temperature along the lines mentioned above. Although the density profile and the width of the surface was found in good agreement with the calculations of large clusters by Pandharipande *et al.*², it was pointed out that the average field did not have the correct $1/z^3$ asymptotic behavior above the surface. This feature has little consequence on the

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energetics of the clusters because the surface tension is one of the quantities used to fix the parameters of the functional and also because the density is vanishingly small in this region of space. However, the scattering of free atoms by the helium surface is extremely sensitive to the asymptotic behavior of the potential well and indeed less sensitive to the surface width, at least in the range of incident perpendicular momentum considered in Ref. 3. It was concluded in Ref. 1 that the density-functional approach developed in that paper could not be used to describe the scattering of free atoms by the surface.

This undesirable feature is due to the zero-range nature of the Skyrme interaction, which is also responsible for another shortcoming. The fact that the fluid has a characteristic microscopic length, namely, the size of the interatomic potential hardcore, is completely ignored. This makes doubtful its use in situations where the liquid is perturbed on a microscopic scale, close to an impurity for instance.

The aim of the present paper is to introduce for liquid helium a novel density functional with an effective interaction having a correct long-range behavior and reasonable short range characteristics and which matches actual properties of liquid helium.

Earlier attempts of this kind exist, with the description of the surface density profile as a goal.⁴ After the early works of Gross, Regge, Padmore, and Cole⁵⁻⁷ Ebner and Saam⁸ developed such an approach introducing a nonlocal term in the density functional. They obtained a surprisingly low unrenormalized surface tension ($0.002 \text{ K } \text{\AA}^{-2}$ the experimental value $\sigma = 0.274 \text{ K } \text{\AA}^{-2}$)⁴, which led them to conclude that the zero-point motion of the surface is an essential part of the surface energy and to compute it through a sophisticated renormalization procedure. In their approach, the nonlocal effective interaction was derived from an approximate theory of the fluid.⁹ Such an approach is indeed well known for classical fluids,¹⁰ for which various approximation schemes have been proposed.

Our approach is much simpler and more empirical: it consists in modeling the fluid with a simple effective interaction, provided it is able to reproduce actual properties of liquid helium. In some way, the model interpolates the known properties of helium in less known situations. Our results are different from those of Ref. 8 and lead to different conclusions about the origin of the surface tension of helium.

2. CHOOSING THE FINITE RANGE EFFECTIVE INTERACTION

Let us first briefly recall the structure of the Skyrme interaction. The energy density E_{sky} is the sum of three types of components: an attractive

term, a density-dependent repulsive term, and a quadratic gradient term with a constant coefficient

$$E_{\text{sky}} = \int d^3r \left[\frac{b}{2} \rho^2 + \frac{c}{2} \rho^{2+\gamma} + d(\nabla\rho)^2 \right] \quad (1)$$

Indeed, the presence of the gradient term implies that the interaction is not really zero-range. In the density functional theory for classical fluids,¹⁰ it corresponds to a gradient expansion limited to quadratic terms, giving a Van der Waals theory of the interface. For a quantum fluid, one must add to E_{sky} an inhomogeneity correction to the quantum kinetic energy density

$$E_q = \int d^3r \frac{\hbar^2}{2m} |\nabla\phi|^2 \quad (2)$$

where $\phi(\mathbf{r}) = \sqrt{\rho(\mathbf{r})}$. The equation of state of the liquid and the surface tension are correctly reproduced with the set of parameters

$$b = -8.8881 \times 10^2 \text{ K } \text{\AA}^3 \quad (3a)$$

$$c = 1.04554 \times 10^7 \text{ K } \text{\AA}^{3(1+\gamma)} \quad (3b)$$

$$d = 2.383 \times 10^3 \text{ K } \text{\AA}^5 \quad (3c)$$

$$\gamma = 2.8 \quad (3d)$$

The density is obtained by minimizing the grand potential

$$\Omega = E_q + E_{\text{sky}} - \mu N \quad (4)$$

(μ is the chemical potential, N the number of particles; the integral is taken over a fixed volume, and only zero temperature is considered). This gives the equation

$$-\frac{\hbar^2}{2m} \Delta\phi + U_{\text{sky}}(\mathbf{r})\phi = \mu\phi \quad (5a)$$

where $U_{\text{sky}}(\mathbf{r})$ is the mean field

$$U_{\text{sky}}(\mathbf{r}) = b\rho + \left(1 + \frac{\gamma}{2}\right) c\rho^{1+\gamma} - 2d\Delta\rho \quad (5b)$$

From Eq. (5a) it is clear that the density decays exponentially above the surface. Hence, through self-consistency, the mean field (5b) has the same exponential behavior and cannot follow a power law.

There are many ways to construct a finite range effective interaction. For instance, one can replace the gradients $\nabla\rho$ in the Skyrme interaction by finite differences ($\rho(\mathbf{r}) - \rho(\mathbf{r}')$), weighted by an appropriate function. This is the way followed by Ebner and Saam in Ref. 8, where they relate the weighting function to statistical functions of the fluid using reasonable assumptions.

We take here a simpler, but in some sense more radical, point of view. We discard from the functional any term with a zero range. More precisely, the energy is now written as

$$E = E_q + E_b + E_c \quad (6)$$

where E_b and E_c are the nonlocal generalizations of the contributions of $b\rho^2/2$ and $c\rho^{2+\gamma}/2$ to E_{Sky} . E_b is now taken as a two-body interaction term

$$E_b = \frac{1}{2} \iint d^3r d^3r' \rho(\mathbf{r}) \rho(\mathbf{r}') V_l(|\mathbf{r} - \mathbf{r}'|) \quad (7)$$

It is indeed natural to take for V_l the standard Lennard-Jones potential describing the He-He interaction ($\varepsilon = 10.22$ K, $\alpha = 2.556$ Å),^{11*} screened in a simple way at distances shorter than a characteristic distance h

$$V_l(|\mathbf{r} - \mathbf{r}'|) = 4\varepsilon \left[\left(\frac{\alpha}{|\mathbf{r} - \mathbf{r}'|} \right)^{12} - \left(\frac{\alpha}{|\mathbf{r} - \mathbf{r}'|} \right)^6 \right] \quad \text{for } |\mathbf{r} - \mathbf{r}'| \geq h \quad (8a)$$

$$V_l(|\mathbf{r} - \mathbf{r}'|) = V_l(h) \left(\frac{|\mathbf{r} - \mathbf{r}'|}{h} \right)^4 \quad \text{for } |\mathbf{r} - \mathbf{r}'| < h \quad (8b)$$

The parameter h is adjusted so that the space integral of V_l is equal to b

$$h = 2.377 \text{ \AA} \quad (9)$$

Hence for smoothly varying densities, the expression (7) is equal to the contribution of the first term of the rhs of (1) to E_{Sky} , but the shape of the two-body interaction at long distance as well as at short distance is more realistic. The shape of the screened core potential (8b) has been arbitrarily taken as a power law. The choice of the fourth power is not critical and is justified below.

For the second term, E_c , we use a prescription similar to the ‘‘weighted density approximation’’ introduced in the studies of classical fluids by Tarazona¹²

$$E_c = \int d^3r \frac{c}{2} \rho(r) (\bar{\rho}_r)^{1+\gamma} \quad (10a)$$

where $\bar{\rho}_r$ is the ‘‘coarse grained density,’’ defined by averaging $\rho(\mathbf{r})$ over a sphere with a radius h

$$\bar{\rho}_r = \int d^3r' \rho(\mathbf{r}') \Pi_h(\mathbf{r} - \mathbf{r}') \quad (10b)$$

*Better potentials are available (for instance, Ref. 11 or Ref. 15). However this choice is not as critical as for *ab initio* methods, because the effective potential will be tuned to reproduce actual properties of helium. However, exponential potentials are not appropriate as discussed in Sec. 5.

with

$$\begin{aligned}\Pi_h(\mathbf{r}) &= \frac{3}{4\pi h^3}, & \text{for } r < h \\ \Pi_h(\mathbf{r}) &= 0, & \text{for } r > h\end{aligned}\quad (10c)$$

The term E_c is always positive, and accounts for the internal kinetic energy as well as for the increasing contribution of the hard core when the density is increased. When $\bar{\rho}_r$ is expressed in terms of $\rho(\mathbf{r})$ through (10b), it appears clearly that (10a) describes many-body correlations with a range of the order of h .

The energy of the inhomogeneous liquid now reads

$$E = \int d^3r \mathcal{H}(\mathbf{r}) \quad (11a)$$

where

$$\mathcal{H}(\mathbf{r}) = \frac{\hbar^2}{2m} |\nabla\phi|^2 + \frac{1}{2} \int d^3r' \rho(\mathbf{r})\rho(\mathbf{r}') V_r(|\mathbf{r}-\mathbf{r}'|) + \frac{c}{2} \rho(\mathbf{r})(\bar{\rho}_r)^{1+\gamma} \quad (11b)$$

It is a mean-field description of the fluid, incorporating finite-range interactions with a correct long-range behavior and incorporating correlation effects in a phenomenological way. There is no adjustable parameter left. Although some choices may appear arbitrary, they meet several essential requirements. First, the equation of state of bulk helium is correctly described over a wide range of densities by keeping the proved values of b , c , and γ . Second, we will show now that the density-density response function of the model fluid is very close to that of real helium around the equilibrium density ρ_0 , for wavevectors going from 0 to 4 \AA^{-1} .

3. LINEAR RESPONSE THEORY

The state of the fluid in an external time dependent potential $\mathcal{V}_{\text{ext}}(\mathbf{r}, t)$ is determined through the time-dependent Hartree theory. The wavefunction $\phi(\mathbf{r}, t)$ satisfies a nonlinear Schrödinger equation obtained by minimizing with respect to ϕ^* the action

$$A = \int_{t_0}^{t_1} \mathcal{L}(t) dt \quad (12a)$$

with

$$\mathcal{L}(t) = \int \left(\phi^* \frac{i\hbar}{2} \frac{\partial\phi}{\partial t} + c.c. \right) d^3r - E - \int d^3r (\mathcal{V}_{\text{ext}}(\mathbf{r}, t) - \mu) \phi^* \phi \quad (12b)$$

In Eq. (12b), E depends on ϕ^* through $\rho = \phi^* \phi$. One readily obtains the equation

$$i\hbar \frac{\partial \phi}{\partial t} = -\frac{\hbar^2}{2m} \Delta \phi + U(\mathbf{r}, t) \phi + (\mathcal{V}_{\text{ext}}(\mathbf{r}, t) - \mu) \phi \quad (13)$$

where $U(\mathbf{r}, t)$ is a mean field equal to the functional derivative of the effective potential energy with respect to $\rho(\mathbf{r}, t)$

$$U(\mathbf{r}, t) = \int d^3 r' \rho(\mathbf{r}', t) V_l(|\mathbf{r} - \mathbf{r}'|) + \frac{c}{2} (\bar{\rho}_r)^{\gamma+1} + \frac{c}{2} (1 + \gamma) \int d^3 r' \Pi_h(|\mathbf{r} - \mathbf{r}'|) \rho(r') (\bar{\rho}_r)^\gamma \quad (14)$$

For a stationary uniform density, Eq. (13) gives the relation between the chemical potential μ and the density ρ

$$b\rho + \left(1 + \frac{\gamma}{2}\right) c\rho^{\gamma+1} = \mu \quad (15a)$$

as well as the pressure p

$$p = \frac{1}{2} b\rho^2 + \left(\frac{\gamma+1}{2}\right) c\rho^{\gamma+2} \quad (15b)$$

and the equilibrium density at zero pressure

$$\rho_0 = \left[\frac{-b}{c(1+\gamma)} \right]^{1/\gamma} \quad (15c)$$

The linear response of the system to the perturbation \mathcal{V}_{ext} is easily obtained by linearizing Eq. (13) around the equilibrium density ρ_0 . Taking $\phi_0 = \sqrt{\rho_0} + \delta\phi$, $\rho = \rho_0 + \delta\rho$ and using (15c), one obtains to first order in $\delta\phi$, $\delta\rho = \sqrt{\rho_0}(\delta\phi + \delta\phi^*)$ and \mathcal{V}_{ext}

$$i\hbar \frac{\partial \delta\phi}{\partial t} = -\frac{\hbar^2}{2m} \Delta \delta\phi + \sqrt{\rho_0} \mathcal{V}_{\text{ext}}(\mathbf{r}, t) + \sqrt{\rho_0} \int [V_l(|\mathbf{r} - \mathbf{r}'|) + V_c(|\mathbf{r} - \mathbf{r}'|, \rho_0)] d^3 r' \delta\rho(\mathbf{r}', t) \quad (16a)$$

where

$$V_c(|\mathbf{r} - \mathbf{r}'|, [\rho]) = \frac{c}{2} (1 + \gamma) (\bar{\rho}_r^\gamma + \bar{\rho}_r'^\gamma) \Pi_h(\mathbf{r} - \mathbf{r}') + \frac{c}{2} \gamma (1 + \gamma) \times \int d^3 r_1 \Pi_h(\mathbf{r} - \mathbf{r}_1) \Pi_h(\mathbf{r} - \mathbf{r}') \rho(r_1) (\bar{\rho}_{r_1})^{\gamma-1} \quad (16b)$$

The last integral on the rhs of Eq. (16a) results from the first-order departure of $U(\mathbf{r}, t)$ from the bulk static mean field. A Fourier transform brings Eq.

(16a) into an algebraic equation. We denote $\hat{f}(\mathbf{q}, \omega)$ the Fourier transform of $f(\mathbf{r}, t)$, defined according to

$$\hat{f}(\mathbf{q}, \omega) = \int f(\mathbf{r}, t) e^{-i(\mathbf{q}\cdot\mathbf{r} - \omega t)} d^3r dt \quad (17)$$

The density-density response function

$$\chi(\mathbf{q}, \omega) = \frac{\delta\hat{\rho}(\mathbf{q}, \omega)}{\hat{\varphi}_{\text{ext}}(\mathbf{q}, \omega)} \quad (18)$$

is found to be given by the expression

$$\chi^{-1}(\mathbf{q}, \omega) = \frac{m\omega^2}{\rho_0 q^2} - \frac{\hbar^2 q^2}{4m\rho_0} - \hat{V}_i(\mathbf{q}) - c(1+\gamma)\rho_0^2 \hat{\Pi}_h(\mathbf{q}) - \frac{c}{2} \gamma(1+\gamma)\rho_0^2 \hat{\Pi}_h^2(q) \quad (19)$$

One verifies that the response of the fluid for large momenta goes to that of the free particles. In the following, we are essentially interested in static properties of the fluid obtained by letting ω go to zero in (19). Figure 1

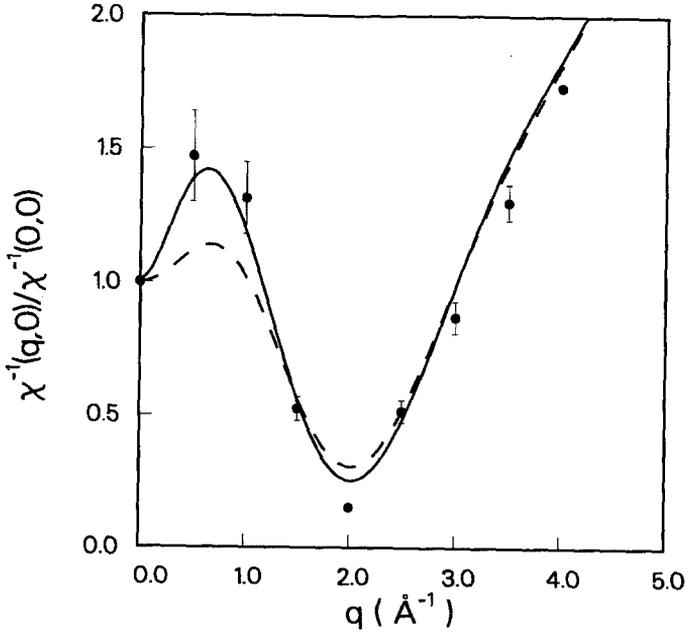


Fig. 1. Calculated static polarizability of helium (solid line). The experimental points are those of Cowley and Woods.¹³ The dashed line is obtained using an exponential potential instead of the Lennard-Jones as explained in Sec. 5.

represents the comparison of $\chi^{-1}(q, \omega = 0)$ with the measurements of Cowley and Woods.¹³ The agreement is satisfactory.

It is worth pointing out that the response $\chi^{-1}(q, 0)$ depends only slightly on the power used to define V_i in the interval $r=0$ to $r=h$. The fourth power used gives a slightly better agreement than lower powers near the minimum about $q = 2 \text{ \AA}^{-1}$. On the other hand, higher powers do not make much difference.

At this stage, it appears that a simple density-functional description of liquid helium exists that reproduces the correct static response function for the fluid (around its equilibrium density), the pressure-density relation (over a wide range of pressure and at zero temperature), using a two-body effective interaction which is correct at long distance and reasonable at short distance. To our knowledge, such a result was not yet clearly established.^{14,*} It is indeed essential to have such a description to deal with inhomogeneous situations where the density may vary on a small scale. It is also clear that we have not treated precisely correlations between atoms at short distances. Processes such as ordering are not evidently at hand.

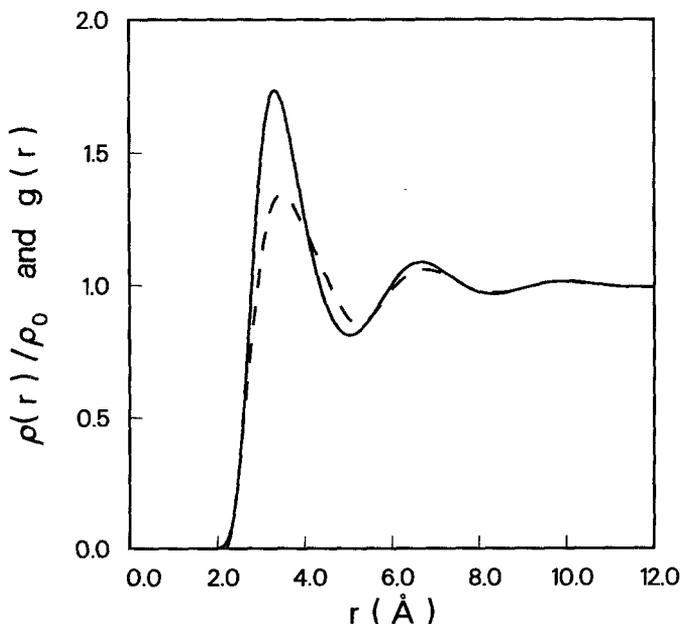


Fig. 2. Calculated radial density profile around a helium atom with infinite mass. The dashed line represents the radial distribution of real helium.¹⁷

*Let us mention nevertheless the work of Ref. 14, in which the linear response function is modeled directly, without specifying an explicit Hamiltonian for the fluid.

In order to test the ability of our model fluid to behave like liquid helium, even outside the linear response regime, we solved Eq. (13) taking for \mathcal{V}_{ext} the He-He potential.¹⁵ This gives the density distribution around a fictitious helium atom with infinite mass.^{16,*} We expect to obtain a radial density profile close to the radial distribution function $g(r)$ of liquid helium. The result is shown in Fig. 2, together with $g(r)$.¹⁷ The profiles are very similar. In particular, the model reproduces the oscillations of $g(r)$. They are more pronounced than in real helium. Note however that neither the symmetrization principle nor the zero-point motion of the fictitious helium atom producing the potential have been taken into account.

4. THE FREE SURFACE

We now turn to the problem of determining the surface tension and profile of the free surface. It corresponds to solving Eq. (13) in its static form ($\partial\phi/\partial t=0$) and without external field ($\mathcal{V}_{\text{ext}}=0$) for the specific geometry we are interested in: the interface is taken as flat, parallel to the x - y plane, the liquid being in the region of negative z . The convolution integral appearing in the mean field $U(\mathbf{r})$ of Eq. (14) can be performed analytically over the variables x and y . Equation (13) then reads as

$$-\frac{\hbar^2}{2m} \frac{d^2\phi(z)}{dz^2} + U(z)\phi(z) = \mu\phi(z) \quad (20)$$

where the self-consistent field $U(z)$ is given by

$$\begin{aligned} U(z) = & 4\pi\epsilon\alpha^2 \int_{z+h}^{\infty} \rho(z') dz' \left(\frac{1}{5} \left(\frac{\alpha}{z-z'} \right)^6 - \frac{1}{2} \right) \left(\frac{\alpha}{z-z'} \right)^4 \\ & + 4\pi\epsilon\alpha^2 \int_{z-h}^{z+h} \rho(z') dz' \left(\left(\frac{8}{15} \left(\frac{\alpha}{h} \right)^6 - \frac{5}{6} \right) - \frac{1}{3} \left(\left(\frac{\alpha}{h} \right)^6 - 1 \right) \left(\frac{z-z'}{h} \right)^6 \right) \\ & \times \left(\frac{\alpha}{h} \right)^4 + 4\pi\epsilon\alpha^2 \int_{-\infty}^{z-h} \rho(z') dz' \left(\frac{1}{5} \left(\frac{\alpha}{z-z'} \right)^6 - \frac{1}{2} \right) \left(\frac{\alpha}{z-z'} \right)^4 \\ & + \frac{3c}{8h} (1+\gamma) \int_{z-h}^{z+h} dz' \left(1 - \left(\frac{z-z'}{h} \right)^2 \right) \rho(z') (\bar{\rho}_z)^\gamma + \frac{c}{2} (\bar{\rho}_z)^{1+\gamma} \end{aligned} \quad (21)$$

Note that for large and positive values of z , $\rho(z) \approx 0$ and only the third term of Eq. (21) is nonzero. The mean field is

$$U(z) \approx -\frac{2\pi\epsilon\alpha^6\rho_0}{3z^3} \quad (22)$$

It has the correct asymptotic behavior.

*This is a usual way of computing $g(r)$ for classical fluids.

Equation (20) is solved using the “imaginary time-step method,” well suited for the numerical determination of a ground state. Given an arbitrary wavefunction φ , applying the evolution operator $\exp(-i/\hbar Ht)$ for an imaginary time $t = -i\hbar\tau$ gives for large τ

$$\lim_{\tau \rightarrow \infty} e^{-H\tau}|\varphi\rangle \sim e^{-E_0\tau}|\psi_0\rangle \quad (23)$$

where E_0 and $|\psi_0\rangle$ denote the energy and the wavefunction of the ground state we are looking for. The algorithm is thus the following. We start with an initial guess $|\varphi^{(0)}\rangle$. One chooses a time step $\Delta\tau$ and iterates the recursion relation

$$|\varphi^{(n+1)}\rangle = e^{-H\Delta\tau}|\varphi^{(n)}\rangle \simeq \left(1 - \Delta\tau \left(-\frac{\hbar^2}{2m}\nabla^2 + U\right)\right)|\varphi^{(n)}\rangle \quad (24)$$

The ground-state energy E_0 is the limit of the series of numbers E_n , defined as

$$E_n = \frac{\langle\varphi^{(n)}|H|\varphi^{(n)}\rangle}{\langle\varphi^{(n)}|\varphi^{(n)}\rangle} \simeq \frac{1}{\Delta\tau} \left(1 - \frac{\langle\varphi^{(n)}|\varphi^{(n+1)}\rangle}{\langle\varphi^{(n)}|\varphi^{(n)}\rangle}\right) \quad (25)$$

The time step $\Delta\tau$ is related to the mesh in r space. One can show that $\Delta\tau$ should be smaller than the inverse of the largest eigenvalue of the discretized Hamiltonian. By considering the kinetic operator only, one gets the upper limit

$$\Delta\tau_{\text{sup}} \simeq \frac{(\Delta x)^2}{4} \frac{2m}{\hbar^2} \quad (26)$$

where Δx is the discretization interval. A value of $\Delta\tau \simeq 0.05\Delta\tau_{\text{sup}}$ gives a good convergence.

The above procedure solves Eq. (20) for a given potential $U(z)$. We have yet to ensure self-consistency. This is done, as usual, by another cycle of iterations: start with a trial density, calculate the field $U(z)$, solve for $\varphi(z)$ as described above, calculate the new $U(z)$ etc.

The surface energy is¹

$$\sigma = \int dz (\mathcal{H}(\rho) - \mu\rho) \quad (27)$$

and is found to be equal to

$$\sigma = 0.277 \text{ K } \text{\AA}^{-2} \quad (28)$$

Remarkably, this is in excellent agreement with the value quoted in section 1 for the measured surface tension of helium extrapolated at zero temperature.⁴ The density profile and the mean field $U(z)$ is shown in Fig. 3. The thickness t_{10-90} is 5.8 \text{\AA}.

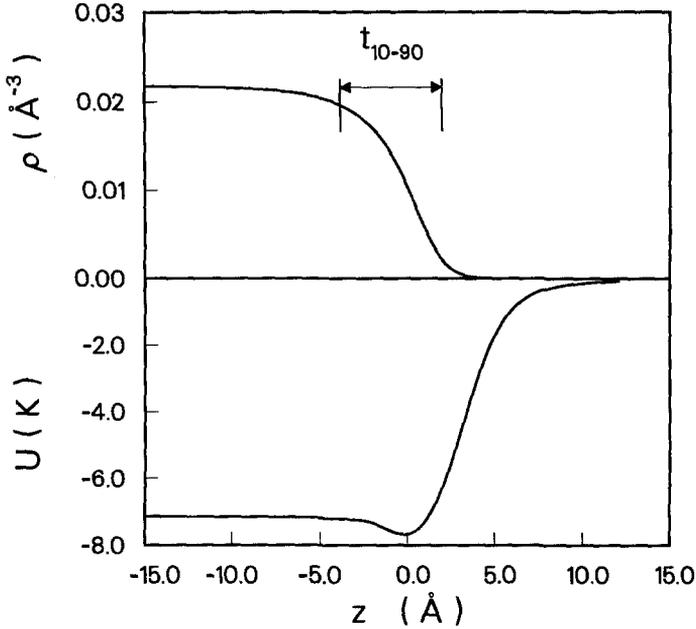


Fig. 3. The calculated density profile at the free surface and the corresponding mean field.

The value (28) is not very sensitive to details of the model used for the fluid, *provided* that it meets the criteria enumerated at the end of Sec. 2. For instance, changing from 1 to 10 the power law of the two-body effective potential between 0 and \hbar changes the value of σ by only $\pm 1\%$. The weighting function (10c) can be replaced by a gaussian distribution (such that its Fourier transform matches $\hat{\Pi}_h(\mathbf{q})$ to reproduce (19): the appropriate width is 1.07 \AA). We have tested also other types of effective potentials, physically less sensible (having for instance an attractive δ -function part at zero range) but also giving the right response function. We also got a value for σ close to the experimental one. Hence, it is felt that the value of the surface tension follows necessarily from three properties of bulk liquid: the equation of state, the density-density response function, and the Van der Waals nature of the two-body interaction.

From this result, one can get hints about the origin of the surface energy of liquid helium. The contribution of the quantum kinetic energy E_q to σ is found to be only 6%. The essential part of σ comes from the contribution of $E_b + E_c - \mu N$. More precisely, we have plotted in Fig. 4 $\mathcal{H}(\rho) - \mu\rho$ as a function of z , as well as $\hbar^2|\nabla\phi|^2/2m$, which is the contribution to the quantum kinetic energy associated with the surface. Whereas the

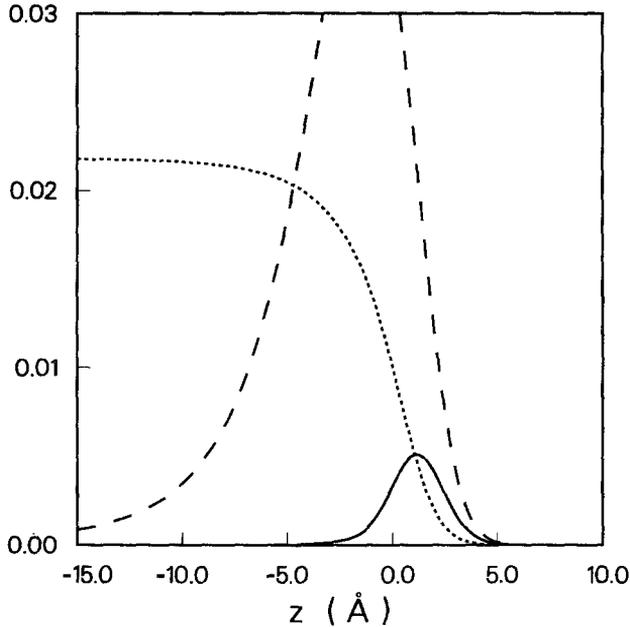


Fig. 4. Free energy density $\mathcal{H}[\rho] - \mu\rho$ (dashed line) and quantum kinetic energy density $\hbar^2|\nabla\phi|^2/2m$ (solid line) in $\text{K } \text{\AA}^{-3}$, as a function of z . Dotted line: $\rho(z)$ in \AA^{-3} .

quantum energy is indeed located near the steepest part of the density profile, the main contribution to the surface energy is located inside the fluid and decays only slowly with the distance from the interface as $1/z^3$. This is quite analogous to the situation in classical Van der Waals fluids: the surface energy originates essentially from the lack of Van der Waals attraction due to the missing half-space of liquid above the surface.

5. COMPARISON WITH PREVIOUS WORKS

The comparison with the calculations of Stringari and Treiner is straightforward. The same equation of state for bulk helium is used here, so that the only difference is the replacement of the Skyrme interaction by a finite-range potential. The surface thickness obtained in the present work is slightly smaller than was found in Ref. 1: 5.8 \AA rather than 7 \AA . The difference in the profiles lies essentially in the outward region of space, which shows larger gradients with the finite-range interaction than with a zero-range one. The latter appears, so to speak, to be more rigid. Indeed, in k -space, a Skyrme interaction has a k^2 -dependence unrealistically large

for large k 's, whereas the Fourier transform of a finite range interaction obviously goes to zero. If one thinks of a development in powers of k , the k^2 -component generating the $|\nabla\rho|^2$ term in the density functional is repulsive and opposes large gradients; on the contrary, the k^4 -components, generating higher order derivatives, would be attractive.

The comparison with the work of Ebner and Saam is not so simple, because their values of the energy density of the fluid $\varepsilon(\rho)$ as well as the effective interaction are determined indirectly through the theory of Mihara and Puff from the bare interatomic potential. For this last quantity, they have taken an exponential potential:

$$V(r) = E_0 \frac{a}{r} (e^{-r/a} - \gamma e^{-\beta r/a}) \quad (29)$$

with $E_0 = 8.95 \times 10^5$ K, $\gamma = 0.2560$, $\beta = 0.8000$, and $a = 0.376$ Å, which is similar to the Lennard-Jones only in the well region. In particular, the $1/r^6$ long-range tail is missing in this potential. In order to evaluate the consequences of such a choice, one can either repeat the Ebner-Saam procedure with the Lennard-Jones potential or repeat our calculations with the exponential potential of Eq. (29) in place of the Lennard-Jones potential. In fact, Ebner and Saam avoided the Lennard-Jones potential because it brings computational difficulties, so that the second method of comparison is much more simple. The appropriate value for h is then 2.40 Å. The response function is shown in Fig. 1 (dashed line). It is clearly not so good and the corresponding value of σ is only $0.19 \text{ K } \text{Å}^{-2}$. Hence, we are led to suspect that the use of the potential (29) tends to lower significantly the value of the bare surface energy of Ref. 8. It would be of interest to reconsider the approach of Ebner and Saam, which is, in some way, more fundamental than ours, with a more appropriate interatomic potential.

6. CONCLUSIONS

It appears possible to describe liquid helium by a density functional based on a nonlocal interaction having a correct $1/r^6$ behavior on the long range and a simple short-range shape. The effective interaction build on this prescription gives a correct response function in the bulk and the right surface tension. This modelization of helium is likely to be a useful tool to extrapolate actual properties of helium over a wide range of conditions, even on nearly microscopic scale. It could be used to study several interesting problems such as surface states of ^3He atoms, scattering of ^3He or ^4He atoms by the surface, clusters, thin films, and atomic impurities in liquid helium.

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