Ripplon dispersion and finite-range effects in the quantum-liquid surface

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We derive within the linear-response theory for inhomogeneous Bose liquids the dispersion relation of a surface excitation (ripplon). It is shown that, in an infinite half space, the ripplon dispersion relation is $\omega_r(q_{||}) \sim q_{||}^{3/2}$, where $q_{||}$ is the momentum parallel to the surface. This dispersion relation holds only if the system is not in an external field. For the special case of a Skyrme interaction with a gradient coupling term, we relate the coefficient of proportionality to the surface energy and thus reproduce the hydrodynamic prediction from linear-response theory. The ripplon-dispersion relation and the shape of the collective excitations is evaluated and discussed for a nonlocal, phenomenological Skyrme interaction and a microscopic interaction based on variational wave functions.

I. INTRODUCTION

The advent of accurate microscopic theories of inhomogeneous quantum liquids and advances in experimental technology have opened new perspectives for studying the surfaces of quantum liquids. Among those, the prospects of directly measuring the surface response function[1] and the layer structure of adsorbed films of ^{4}He by electron-mobility measurements[2] promise a most exciting field of experimental and theoretical research. The present paper addresses a number of problems related to the excitation spectrum of liquid surfaces, which should lead to both a deeper understanding of the theory of surface excitations and to a better interpretation of experimental evidence.

Surface excitations have been historically studied mostly within hydrodynamic models (for a review, see Ref. 3). These studies lead to a dispersion relation for the surface excitation ("ripplon") of the form

$$\omega^2(q_{||}) = \frac{\sigma}{m\rho_\infty} q_{||}^3 , \qquad (1.1)$$

where σ is the surface energy, $q_{||}$ the wave number parallel to the surface, and ρ_∞ is the asymptotic density. A derivation of the dispersion law (1.1) from linear-response theory does not exist according to our knowledge. The formulation of linear-response theory in the surface is rather straightforward,[4-6] but we will see that some delicate considerations are necessary for a rigorous proof of Eq. (1.1).

The hydrodynamic dispersion law is rigorously valid only in the long-wavelength limit. At shorter wavelengths, curvature and finite-range effects must be considered, and finally one expects a level crossing with the bulk zero-sound mode.[3]

This paper is devoted to a microscopic study of the properties of surface excitations. In order to cover a wide range of cases, we employ two different theories for a liquid in an infinite half-space: The first theory is phenomenological. We start from an energy functional of the form

$$E \equiv T + E_c = \int d^3 r \epsilon[\rho_1(\mathbf{r}), \nabla \rho_1(\mathbf{r})] , \qquad (1.2)$$

where $\epsilon(\mathbf{r}) = t(\mathbf{r}) + \epsilon_c(\mathbf{r})$ is the energy density consisting of the kinetic-energy density

$$t(\mathbf{r}) = \frac{\hbar^2}{2m} |\nabla \sqrt{\rho_1(\mathbf{r})}|^2 \qquad (1.3)$$

and the correlation-energy density $\epsilon_c(\mathbf{r})$. The explicit form assumed in Ref. 7 for the correlation-energy density is

$$\epsilon_c(\mathbf{r}) = \tfrac{1}{2} b \rho_1(\mathbf{r})^2 + \tfrac{1}{2} c \rho_1(\mathbf{r})^{2+\gamma} + d |\nabla \rho_1(\mathbf{r})|^2 . \qquad (1.4)$$

In (1.4), the term proportional to b corresponds to an attractive two-body contact force, the term proportional to c to a repulsive, density-dependent contact interaction, and finally the term proportional to d to a repulsive surface interaction. The surface structure of liquid ^{4}He has been obtained[7] by minimizing the energy (1.2) with respect to the one-body density for a fixed particle number $N = \int d^3 r \rho_1(\mathbf{r})$, i.e., by solving

$$\frac{\delta(E - \mu N)}{\delta \rho_1(\mathbf{r})} = 0 , \qquad (1.5)$$

where μ is the chemical potential. Equation (1.5) can be written as a Hartree equation

$$-\frac{\hbar^2}{2m} \nabla^2 \sqrt{\rho_1(\mathbf{r})} + V_H(\mathbf{r}) \sqrt{\rho_1(\mathbf{r})} = \mu \sqrt{\rho_1(\mathbf{r})} . \qquad (1.6)$$

V_H is the local Hartree potential, which is given for the model Hamiltonian (1.4) by

$$V_H(\mathbf{r}) = b \rho_1(\mathbf{r}) + \frac{2+\gamma}{2} c \rho_1^{1+\gamma}(\mathbf{r}) - 2d \nabla^2 \rho_1(\mathbf{r}) . \qquad (1.7)$$

Our second approach is based on a variational *ansatz*

for the many-body wave function. In that theory, one starts with the Feenberg form for the ground state

$$\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_N)=\exp\left[\frac{1}{2}\left[\sum_i u_1(\mathbf{r}_i)+\sum_{i<j}u_2(\mathbf{r}_i,\mathbf{r}_j)\right.\right.$$
$$\left.\left.+\sum_{i<j<k}u_3(\mathbf{r}_i,\mathbf{r}_j,\mathbf{r}_k)+\cdots\right]\right]$$

(1.8)

and a microscopic Hamiltonian

$$H=-\sum_i\frac{\hbar^2}{2m}\nabla_i^2+\sum_{i<j}v(\,|\,\mathbf{r}_i-\mathbf{r}_j\,|\,)\;.$$

(1.9)

For given correlation functions $u_1(\mathbf{r})$, $u_2(\mathbf{r}_i,\mathbf{r}_j)$, and $u_3(\mathbf{r}_i,\mathbf{r}_j,\mathbf{r}_k)$, the energy expectation value corresponding to the wave function (1.8) can be evaluated exactly by Monte-Carlo methods.[8] Due to the complication of choosing the "best" correlation functions for a Monte-Carlo calculation, only very simple choices have been investigated. While it is well known that the energy is only little affected by improved correlations, it is equally well known[9] that the spectrum of the low-lying collective excitations depends sensitively on the correct inclusion of long-ranged correlations. One expects therefore that one must also in the case of surface excitation take the "best possible" Feenberg function to obtain meaningful results for the excitation spectrum.

A route that addresses this problem[10] is to evaluate the energy approximately, using the hypernetted-chain (HNC) theory. In that case, the "best possible" correlation functions $u_n(\mathbf{r}_1,\ldots,\mathbf{r}_n)$ can easily be obtained by minimizing the ground-state energy

$$H_{00}=\frac{\int d^3r_1,\ldots,d^3r_N\Psi_0(\mathbf{r}_1,\ldots,r_N)H\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_N)}{\int d^3r_1,\ldots,d^3r_N\,|\,\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_N)\,|^2}$$

(1.10)

by means of Euler-Lagrange equations

$$\frac{\delta H_{00}}{\delta u_n(\mathbf{r}_1,\ldots,\mathbf{r}_n)}=0\;.$$

(1.11)

The method has the important advantage that one obtains automatically the correct long-ranged correlations and the spectrum of collective excitations.[6] The Euler equation for the one-body function $u_1(\mathbf{r})$ may be rewritten as an equation for the one-body density $\rho_1(\mathbf{r})$ which is structurally identical to Eq. (1.6); of course the definition of the Hartree-potential is more complicated. The disadvantage of the approach is that, once the wave function (1.8), the microscopic Hamiltonian, and the approximation (e.g., HNC) are chosen, there is no way to add information on the macroscopic properties of the system. This is especially a problem in ⁴He since approximation methods like HNC converge poorly in this system due to its high density.[9] The only way to improve upon the microscopic description is to add "elementary diagram" contributions and three-body correlations. The formal ingredients for such improvements are well understood,[11,12] but the pro-

gram has so far been carried out only for bulk ⁴He.

In the problem of describing the propagation of ripplons in the surface of liquid ⁴He we are, therefore, in the situation that there is essentially no theory that is entirely satisfactory from a microscopic point of view: The effective-interaction model of Ref. 7 allows for accurate predictions of the static surface properties, but it is of phenomenological nature and only partially accounts for finite-range effects. The Monte Carlo method has difficulties in describing details of the correlations like long-ranged effects and anisotropies, and estimates for the ripplon-dispersion relation based on these results[13] are questionable. HNC and Euler-Lagrange methods obviate both of the above problems, but can presently due to the poor convergence in ⁴He, provide only qualitative information.

In this work we utilize the phenomenological theory of Ref. 7 and the microscopic HNC theory of Ref. 10 to investigate the collective excitations in the surface of liquid ⁴He. The aim of our study is on the one hand to obtain information on the range of validity of the phenomenological Skyrme interaction, and on the other hand to get estimates for the importance of finite-range effects. To this end, we have studied two different parametrizations of the Skyrme interaction (1.4): One that reproduces the *experimental* properties of ⁴He and one that reproduces the *calculated* properties of ⁴He in the HNC approximation. The second model is compared with the prediction of the HNC approximation of Ref. 10.

Our paper is organized as follows. In the next section, we review the linear-response theory of inhomogeneous Bose systems and draw the connection to the sum-rule approach. This discussion follows the general line of Ref. 6, which is closely related to the algorithms used in the numerical part of our work. To make contact with the formulation of Chang and Cohen,[14] we formulate variational principles for the lowest normal mode.

Section III specializes on the semi-infinite system. We prove that, in a self-bound system *without an external field*, the lowest lying excitation corresponds, in the long-wavelength limit, to a rigid displacement of the surface. The dispersion relation in the limit $q_{||}\to0$, where $q_{||}$ is the wave number parallel to the surface, is obtained by first-order perturbation theory. We find, in agreement with the prediction of hydrodynamic models,[3] the dispersion relation (1.1). The fourth section presents our numerical results and discusses the importance of finite-range effects.

II. LINEAR-RESPONSE THEORY AND SUM RULES

The collective excitations of the system are given by the normal modes of the response function $X(\mathbf{r},\mathbf{r}';\omega)$, i.e., by the solutions of

$$\int d^3r_2X^{-1}(\mathbf{r}_1,\mathbf{r}_2;\omega)\delta\rho_1(\mathbf{r}_2)=0\;.$$

(2.1)

In the random-phase approximation (RPA), which is applicable if the wavelength of the excitations is long compared to the interparticle distance, the response function $X(\mathbf{r},\mathbf{r}';\omega)$ is related to the response function $X_0(\mathbf{r},\mathbf{r}';\omega)$ of the "noninteracting" system by

$$\chi^{-1}(\mathbf{r}_1,\mathbf{r}_2;\omega) = \chi_0^{-1}(\mathbf{r}_1,\mathbf{r}_2;\omega) - V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2) , \quad (2.2)$$

where

$$V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2) = \frac{\delta^2 E_c}{\delta\rho_1(\mathbf{r}_1)\delta\rho_1(\mathbf{r}_2)} \quad (2.3)$$

is the "particle-hole interaction." In Eq. (2.3) E_c is the correlation energy of the system, i.e., the ground-state energy *without* the kinetic energy. At this stage, no assumption is needed on the origin and the analytic form of the particle-hole interaction except that it is energy independent. To be specific, we have in mind to generate $V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)$ from the Skyrme model or from the variational energy functional discussed above. For the variational wave function, $V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)$ is just a function of the coordinates $\mathbf{r}_1$ and $\mathbf{r}_2$. For the Skyrme model, the $V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)$ obtained by the definition (2.3) has the form

$$V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2) = \left[b + c \left[1 + \frac{\gamma}{2} \right](1+\gamma)\rho_1^\gamma \left[\frac{\mathbf{r}_1+\mathbf{r}_2}{2} \right] \right]$$

$$\times \delta(\mathbf{r}_1-\mathbf{r}_2) - \tfrac{1}{2}d(\nabla_1-\nabla_2)^2\delta(\mathbf{r}_1-\mathbf{r}_2) . \quad (2.4)$$

The "noninteracting" response function $\chi_0(\mathbf{r}_1,\mathbf{r}_2;\omega)$ is the one corresponding to a system described by the local one-body Hamiltonain yielding the ground-state density $\rho_1(\mathbf{r})$ [see Eq. (1.6)]. The Hartree potential V_H can be eliminated in favor of the ground-state density. Then we can express H_1 more conveniently as

$$H_1 \equiv -\frac{\hbar^2}{2m}\frac{1}{\sqrt{\rho_1(\mathbf{r})}}\nabla\rho_1(\mathbf{r})\cdot\nabla\frac{1}{\sqrt{\rho_1(\mathbf{r})}} , \quad (2.5)$$

and $\chi_0(\mathbf{r}_1,\mathbf{r}_2,\omega)$ can be written symbolically in the form

$$\chi_0 = 2\sqrt{\rho_1}H_1[\hbar^2\omega^2 - H_1^2]^{-1}\sqrt{\rho_1} . \quad (2.6)$$

At this stage, the response function is usually formulated in terms of the eigenstates and excitation energies of H_1. One can avoid this intermediate step by considering the fourth-order eigenvalue problem[6,10]

$$[H_1 + 2\hat{V}_{p\text{-}h}]H_1\psi^{(l)} = \hbar^2\omega_l^2\psi^{(l)} , \quad (2.7)$$

where

$$\hat{V}_{p\text{-}h}(\mathbf{r},\mathbf{r}') = \sqrt{\rho_1(\mathbf{r})}V_{p\text{-}h}(\mathbf{r},\mathbf{r}')\sqrt{\rho_1(\mathbf{r}')} .$$

The eigenstates of $\psi^{(l)}$ of (2.7) are normalized such that

$$(\psi^{(l)} \mid H_1 \mid \psi^{(l')}) = \delta_{l,l'} . \quad (2.8)$$

From these states we can construct the response function

$$\chi(\mathbf{r},\mathbf{r}',\omega) = 2\sqrt{\rho_1(\mathbf{r})}$$

$$\times \sum_l \left[[H_1\psi^{(l)}](\mathbf{r})\frac{1}{\hbar^2\omega^2 - \hbar^2\omega_l^2}[H_1\psi^{(l)}](\mathbf{r}') \right]$$

$$\times \sqrt{\rho_1(\mathbf{r}')} \quad (2.9)$$

and its inverse

$$\chi^{-1}(\mathbf{r},\mathbf{r}',\omega) = \tfrac{1}{2}\sum_l \frac{\psi^{(l)}(\mathbf{r})}{\sqrt{\rho_1(\mathbf{r})}}(\hbar^2\omega^2 - \hbar^2\omega_l^2)\frac{\psi^{(l)}(\mathbf{r}')}{\sqrt{\rho_1(\mathbf{r}')}} .$$

$$(2.10)$$

From the orthogonality relation (2.8) we see immediately that the normal modes of the system are given by

$$\delta\rho_{(l)}^{1/2}(\mathbf{r},\omega) \equiv \frac{\delta\rho_{(l)}(\mathbf{r},\omega)}{2\sqrt{\rho_1(\mathbf{r})}} \sim [H_1\psi^{(l)}](\mathbf{r})\delta(\omega-\omega_l) . \quad (2.11)$$

For the estimate of the excitation energies it is sometimes useful to formulate a variational principle. Such a variational principle is easily derived from Eq. (2.7): for an arbitrary trial function $\psi_f(\mathbf{r})$, we obtain from (2.7)

$$\frac{(\psi_f \mid H_1(H_1 + 2\hat{V}_{p\text{-}h})H_1 \mid \psi_f)}{(\psi_f \mid H_1 \mid \psi_f)} \geq \hbar^2\omega^2 . \quad (2.12)$$

The same variational principle follows independently from a sum-rule approach: For a given one-body excitation operator

$$F = \sum_i f(\mathbf{r}_i) \quad (2.13)$$

the RPA sum rules are defined as the nth energy-weighted moments of the imaginary part of the RPA response function (2.9) as

$$m_n = \int \frac{d(\hbar\omega)}{2\pi}(\hbar\omega)^n \int \int d^3r_1 d^3r_2 f^*(\mathbf{r}_1)f(\mathbf{r}_2)$$

$$\times \mathrm{Im}\chi(\mathbf{r}_1,\mathbf{r}_2;\omega) . \quad (2.14)$$

From Eqs. (2.7)—(2.9) one obtains the explicit expressions for the sum rules m_3, m_1, and m_{-1}:

$$m_3 = \langle \Psi_f \mid H_1[H_1 + 2\hat{V}_{p\text{-}h}]H_1 \mid \Psi_f \rangle , \quad (2.15)$$

$$m_1 = \langle \Psi_f \mid H_1 \mid \Psi_f \rangle , \quad (2.16)$$

$$m_0 = \langle \Psi_f \mid S(\mathbf{r},\mathbf{r}') \mid \Psi_f \rangle , \quad (2.17)$$

and

$$m_{-1} = \langle \Psi_f \mid [H_1 + 2\hat{V}_{p\text{-}h}]^{-1} \mid \Psi_f \rangle , \quad (2.18)$$

where

$$\Psi_f = f(\mathbf{r})\sqrt{\rho_1(\mathbf{r})} , \quad (2.19)$$

and $S(\mathbf{r},\mathbf{r}')$ is the static form factor.

The quantities m_3/m_1 and m_1/m_{-1} are average square excitation energies depending on the excitation operator F. These ratios are generally upper bounds for the square of the exact excitation energy following from the equation of motion (2.7). In our special case, m_3/m_1 is obviously identical to the functional (2.12) obtained directly from the RPA equation in the form (2.7). We see also that minimizing m_3/m_1 or m_1/m_{-1} with respect to the excitation operator.

$$\frac{\delta}{\delta\Psi_f}\frac{m_3}{m_1} = 0 \quad \text{or} \quad \frac{\delta}{\delta\Psi_f}\frac{m_1}{m_{-1}} = 0 \quad (2.20)$$

is identical to solving the RPA equation (2.7). Of course, evaluating m_3/m_1 or m_1/m_{-1} in the sense of a Ritz

principle for the same trial function Ψ_f will usually lead to different estimates of the excitation energy.

To conclude this section, let us briefly discuss the connection between our present RPA formulation and the work of Chang and Cohen.[14] These authors start with a wave function for the excited state of the form

$$\Psi(\mathbf{r}_1,\ldots,\mathbf{r}_N)=F\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_N) \; , \qquad (2.21)$$

where F is of the form (2.13), and $\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_N)$ is assumed to be the ground-state wave function. [In fact, it is sufficient to assume that $\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_N)$ is a Feenberg function (1.8) with optimized two-body correlations $u_2(\mathbf{r}_i,\mathbf{r}_j)$.] For a Feynman wave function of the form (2.21) for the excited states, Chang and Cohen derive an estimate for the excitation energy $\hbar\omega$ in terms of the static form factor $S(\mathbf{r},\mathbf{r}')$:

$$\hbar\omega \le \frac{\langle \Psi | H | \Psi \rangle}{\langle \Psi | \Psi \rangle}$$

$$= \frac{\hbar^2}{2m} \frac{\int d^3r \rho_1(\mathbf{r})\,|\,\nabla f(\mathbf{r})\,|^2}{\int d^3r\,d^3r'\,S(\mathbf{r},\mathbf{r}')f(\mathbf{r})\sqrt{\rho_1(\mathbf{r})}f^*(\mathbf{r}')\sqrt{\rho_1(\mathbf{r}')}}$$

$$= \frac{m_1}{m_0} \; , \qquad (2.22)$$

which is minimized by the solution of the Euler-Lagrange equation

$$H_1\sqrt{\rho_1(\mathbf{r})}f(\mathbf{r})=\hbar\omega \int d^3r'\,S(\mathbf{r},\mathbf{r}')\sqrt{\rho_1(\mathbf{r}')}f(\mathbf{r}') \; . \qquad (2.23)$$

(Note that our definition of the static form factor $S(\mathbf{r},\mathbf{r}')$ deviates from the one used by Chang and Cohen by a factor $\sqrt{\rho_1(\mathbf{r})}\sqrt{\rho_1(\mathbf{r}')}$. We have chosen this convention to make the static form factor dimensionless like in homogeneous systems.) If we use for $S(\mathbf{r},\mathbf{r}')$ the RPA expression

$$S(\mathbf{r},\mathbf{r}')=\sum_l \frac{1}{\hbar\omega_l}[H_1\psi^{(l)}(\mathbf{r})][H_1\psi^{(l)}(\mathbf{r}')] \; , \qquad (2.24)$$

where the ω_l and the Ψ_l are the solutions of Eq. (2.7), it follows from Eq. (2.24) that the solutions of (2.23) are

$$\sqrt{\rho_1(\mathbf{r})}f^{(l)}(\mathbf{r})=\psi^{(l)}(\mathbf{r}) \; , \qquad (2.25)$$

i.e., the minimization of m_3/m_1, m_1/m_{-1} or m_1/m_0 leads to identical solutions.

Of course, recovering the Chang-Cohen variational principle from the RPA is not surprising. In fact, it is straightforward to show that the ansatz (2.21) with a local excitation operator (2.13) leads always to a description of the excited states that is formally identical to the RPA with an energy independent particle-hole interaction.[6] But there is, compared with the Chang-Cohen variational principle (2.22), an important practical advantage in using either directly the RPA equation (2.7) or using a variational principle that is derived from the RPA (such as m_3/m_1) rather than m_1/m_0. Using the variational principle (2.22) requires in fact that one knows already the spectrum of low-lying excitations, and can at its best reproduce what one has put into the calculation of the

static form factor. In other words, precise knowledge on the static form factor in the long wavelength limit is needed. Little is known experimentally; the necessary information could, for example, be obtained from an RPA calculation as carried out here. The particle-hole interaction entering the RPA equations is a less critical input to the theory in the sense that it does not satisfy any identities at long wavelengths like, for example, the normalization of the static form factor. As a step beyond the Skyrme model used here one might introduce a phenomenological pseudopotential like in the theory of Aldrich and Pines.[15]

III. RIPPLON DISPERSION

Let us specialize now to the propagation of the collective excitations in an infinite half-space. To be specific, we assume a system that is translationally invariant in the x-y plane, and has a nonuniform density profile $\rho_1(z)$ describing a surface, i.e.,

$$\rho_1(z) \to \begin{cases} 0 & \text{if } z \to +\infty \; , \\ \rho_\infty & \text{if } z \to -\infty \; . \end{cases} \qquad (3.1)$$

An important identity follows from this geometry by considering a locally rigid displacement of the surface profile $\rho_1(z)\to\rho_1(z+\xi)$: The Hartree potential $V_H(z)$ must experience the same displacement, i.e.,

$$V_H[\rho_1(z+\xi)](z)=V_H[\rho(z)](z+\xi) \; , \qquad (3.2)$$

and the energy must change by an amount $-\xi\mu\rho_\infty$.

$$E[\rho(z+\xi)]=E[\rho(z)]-\xi\mu\rho_\infty \; . \qquad (3.3)$$

An identity between the Hartree potential and the particle-hole interaction is now easily derived from (3.2) by expanding the Hartree potential to first order in ξ. We obtain

$$V_H(z+\xi)=V_H(z)+\xi\frac{dV_H(z)}{dz}$$

$$= \frac{\delta E_c}{\delta\rho_1}(z+\xi)$$

$$= \frac{\delta E_z}{\delta\rho_1}(z)+\xi \int d^3r'\frac{\delta^2 E_c}{\delta\rho_1(z)\delta\rho_1(z')}\frac{d\rho_1(z')}{dz'} \; ,$$

$$\qquad (3.4)$$

i.e.,

$$\frac{dV_H(z)}{dz} = \int d^3r'\,V_{p\text{-}h}(\mathbf{r},\mathbf{r}')\frac{d\rho_1(z')}{dz'} \; . \qquad (3.5)$$

In our geometry, it is most convenient to work in configuration space in the z direction, and in momentum space in the coordinates parallel to the surface. The one-body operator H_1 and the particle-hole interaction $V_{p\text{-}h}$ become functions $H_1(q_{\parallel})$ and $V_{p\text{-}h}(z,z',q_{\parallel})$. From Eq. (3.5) and the Hartree equation (1.6) we conclude that $d\sqrt{\rho_1(\mathbf{r})}/dz$ is at $q_{\parallel}=0$ a zero-energy eigenfunction of $H_1+2V_{p\text{-}h}$, i.e.,

$$H_1(q_{||}=0)\left[\frac{d\sqrt{\rho_1(\mathbf{r})}}{dz}\right]+2\int dz'\,\hat{V}_{p\text{-}h}(z,z',q_{||}=0)\left[\frac{d\sqrt{\rho_1(z')}}{dz'}\right]=0 \tag{3.6}$$

From (3.6) and the structure (2.5) of H_1 we find that the RPA equation (2.7) has *two* zero-energy solutions in the limit $q_{||}\rightarrow 0$. The first solution is simply $[\rho_1(\mathbf{r})]^{1/2}$, which we readily identify with the phonon. Its dispersion relation is dominated by the bulk limit of the density and the particle-hole interaction. It is identical to the Feynman dispersion relation of the phonon $\omega_s=\hbar c_s q_{||}$ $(q_{||}\rightarrow 0)$, where the bulk velocity of sound c_s is related to the particle-hole interaction $V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)$ by

$$mc_s^2=\rho_\infty\int d^3r V_{p\text{-}h}(r)\quad(r=|\mathbf{r}_1-\mathbf{r}_2|)\ . \tag{3.7}$$

(Note that in the limit $z\rightarrow-\infty$, the particle-hole interaction is translationally invariant and isotropic.)

The second zero-energy solution is the one connected with the identity (3.6), i.e., it has the property [c.f. Eq. (2.11)]

$$\delta\sqrt{\rho_1(z)}\sim H_1(q_{||}=0)\psi_r(z)=\varphi d\sqrt{\rho_1(z)}/dz\ ,$$

where φ is a normalization constant to satisfy the orthonormalization (2.8). This solution corresponds, as discussed above, to a rigid displacement of the surface; we identify it with the long-wavelength limit of the ripplon. To obtain its dispersion relation, we consider the $q_{||}$ dependence of $H_1+2\hat{V}_{p\text{-}h}$ as a perturbation, the unperturbed state $\psi_r^0(z,q_{||})$ being determined by the differential equation

$$\delta[\rho_1(z,q_{||})]^{1/2}\sim\varphi\frac{d\sqrt{\rho_1(z)}}{dz}\exp(i\mathbf{r}_{||}\cdot\mathbf{q}_{||})$$

$$=H_1(q_{||})\psi_r^0(\mathbf{r})\ . \tag{3.8}$$

Equation (3.8) describes a density fluctuation given by a rigid displacement of the surface

$$\delta\rho_1(\mathbf{r})=\rho_1[z+\epsilon\exp(i\mathbf{r}_{||}\cdot\mathbf{q}_{||})]-\rho_1(z) \tag{3.8'}$$

which leaves the system in a situation of local equilibrium typical for the hydrodynamic regime. The solution of Eq. (3.8) is *for all* $q_{||}$ a zero-energy eigenfunction of Eq. (2.7) if the term $H_1+2\hat{V}_{p\text{-}h}$ is replaced by its $q_{||}\rightarrow 0$ limit. For small $q_{||}$, Eq. (3.8) has the solution

$$\psi_r^0(z)=-\left[\frac{2m}{\hbar^2 q_{||}}\frac{\rho_1(z)}{\rho_\infty}\right]^{1/2}\exp(q_{||}z+i\mathbf{r}_{||}\cdot\mathbf{q}_{||})$$

$$(q_{||}\rightarrow 0)\ . \tag{3.9}$$

Note that there is a subtle difference between using the solution of the differential equation (3.8) as the unperturbed state, and using (3.9) as a trial function for estimating the excitation energy. Equation (3.9) is an *approximate* solution of the differential equation (3.8), which is valid for small $q_{||}$. In particular, we have to use in the following derivations the property (3.8) whenever the combination $H_1(q_{||})\psi_r^0$ occurs. Taking Eq. (3.9) without this additional precaution generates a density fluctuation

that does not correspond to a rigid displacement of the ground-state density. Among others, an estimate of the excitation energy using the m_3/m_1 sum rule with this excitation operator would generate additional incorrect contributions to the ripplon dispersion.

Having determined the unperturbed state ψ_r^0 we are ready to calculate the first correction to the (zero) unperturbed ripplon energy:

$$\hbar^2\omega_r^2=q_{||}^2\frac{\left\langle\psi_r^0\left|H_1\left[\frac{\hbar^2}{2m}+2\hat{V}_1\right]H_1\right|\psi_r^0\right\rangle}{(\psi_r^0\mid H_1\mid\psi_r^0)}$$

$$=-\frac{\hbar^2 q_{||}^3}{m}\frac{T+\frac{1}{2}(\rho_1'\mid V_1(z,z')\mid\rho_1')}{(\sqrt{\rho_1(z)}\exp(q_{||}z)\mid d\sqrt{\rho_1(z)}/dz)}$$

$$=\frac{\hbar^2 q_{||}^3}{m\rho_\infty}\{2T+(\rho_1'\mid V_1(z,z')\mid\rho_1')\}\ . \tag{3.10}$$

Here, we have expanded the particle-hole interaction for small $q_{||}$

$$\hat{V}_{p\text{-}h}(z,z',q_{||})=\hat{V}_{p\text{-}h}(z,z',0)+q_{||}^2\hat{V}_1(z,z')+O(q_{||}^4) \tag{3.11}$$

and used, as usual, the definition

$$\hat{V}_1(z,z')\equiv\sqrt{\rho_1(z)}V_1(z,z')\sqrt{\rho_1(z')}\ .$$

Note that the expansion (3.11) does not contain a term linear in $q_{||}$ since $\hat{V}_{p\text{-}h}(z,z',r_{||})$ falls off as $r_{||}^{-6}$ for $r_{||}\rightarrow\infty$. Hence, its two-dimensional Fourier transform $\hat{V}_{p\text{-}h}(z,z',q_{||})$ is differentiable at the origin. Recall also that

$$T=\frac{\hbar^2}{2m}\int dz\mid d\sqrt{\rho_1(z)}/dz\mid^2$$

is the kinetic energy per surface area.

With Eq. (3.10) we have arrived at our final formal result for the long-wavelength behavior of the ripplon dispersion relation, which can be cast in the form

$$\omega_r^2=\frac{\sigma_*}{m\rho_\infty}q_{||}^3 \tag{3.12}$$

with

$$\sigma_*=2T+(\rho_1'\mid V_1\mid\rho_1')\ . \tag{3.13}$$

For completeness, we display also the form of the static structure function in the long-wavelength limit. In that case, we can restrict the mode sum (2.24) to the contribution from the ripplon, and insert our solutions (3.9) and (3.12). Thus we find

$$S(z,z',q_{||})=2\left[\frac{\hbar^2}{m\sigma_*\rho_\infty q_{||}}\right]^{1/2}\frac{d\sqrt{\rho_1(z)}}{dz}\frac{d\sqrt{\rho_1(z')}}{dz'}\ .$$

$$\tag{3.14}$$

The dispersion relation (3.12) is so far formally identical with the hydrodynamic prediction. In addition, the hydrodynamic theory identifies σ_* with the surface energy σ. This macroscopic surface energy is defined as

$$\sigma = \lim_{N \to \infty} [E - \mu N] , \qquad (3.15)$$

where E and N are energy and particle number per surface area.

There is no a priori reason that σ and σ_* can be identified in any approximate theory like, for example, the variational theory using a truncated Feenberg function. The situation is similar to the case of the phonon dispersion relation, where the identity $mc_s^2 = \rho \int d^3 r V_{p-h}(r)$ is rigorously true only for the exact wave function,[12] and the diagrammatic proof of the relation between the long-wavelength limit of the particle-hole interaction and the velocity of sound requires rather subtle arguments. A similar analysis is not yet available for the surface problem.

The situation is by construction different for the Skyrme model, in fact it is quite easy to prove here that the σ_* of Eq. (3.13) is identical to the macroscopic surface energy σ. From Eq. (2.4) we find that $V_1^{\text{Skyrme}}(z,z') = 2d\delta(z-z')$ and, hence

$$\sigma_*^{\text{Skyrme}} = 2T + 2d \int dz \, |\rho_1'(z)|^2 . \qquad (3.16)$$

Alternatively,[7] we can multiply the Hartree equation (1.6) with $d[\rho(z)]^{1/2}/dz$, integrate twice using the explicit form of the Hartree potential (1.7), and obtain

$$T + \int dz [\epsilon_c(z) - 2d \, |\nabla \rho_1(z)|^2] = \mu N , \qquad (3.17)$$

where N is the particle number. Combining this with (1.2), (1.4), and the macroscopic definition (3.15) of the surface energy we find

$$\sigma_*^{\text{Skyrme}} = \sigma^{\text{Skyrme}} \qquad (3.18)$$

which is the desired result.

The above derivation of the $q_{||}^{3/2}$ law for the ripplon dispersion relation is to our knowledge the first microscopic derivation of that law from linear-response theory. The structure of our result is quite interesting: We find two contributions to the surface energy. The first one is a local term that is simply determined by the density gradient and proportional to the kinetic energy of the ground state. The second one is an interaction term that reflects *the finite range* of the particle-hole interaction. Terms of this structure do not contribute to the propagation of bulk phonons. Conversely, we find that the zero-range terms in the Skyrme interaction do not contribute to the propagation of ripplons.

To conclude this section, we comment briefly on the estimate of the excitation energies by means of the m_1/m_0 sum rule, which has been attempted by Chang and Cohen[14] and others.[13] For the estimate of the excitation energy by means of the ratio m_1/m_0 we need an additional assumption on the long-wavelength behavior of the static form factor. We assume that it is given by Eq. (2.24). We assume further that one (the lowest) of the collective modes entering the sum (2.24) dominates for

$q_{||} \to 0$ such that the modesum can be restricted to that lowest state. This assumption is correct for the surface structure function, in which case the lowest mode ω_0 is the ripplon. It is also fulfilled for a local density approximation for $S(\mathbf{r}, \mathbf{r}')$ in which case the lowest mode ω_0 is the phonon. Evaluating m_1/m_0 for some trial excitation operator ψ_f we find

$$\left[\frac{m_1}{m_0}\right]_f = \hbar \omega_0 \frac{(\psi_f | H_1 | \psi_f)(\psi^{(0)} | H_1 | \psi^{(0)})}{|(\psi_f | H_1 | \psi^{(0)})|^2} \geq \hbar \omega_0 .$$

$$(3.19)$$

(We have displayed here explicitly the normalization of the solution $\psi^{(0)}$ of the RPA equation.) One verifies readily that equality holds in Eq. (3.19) for the true surface excitation (3.8) and the Chang-Cohen trial function

$$\psi_f^{\text{CC}} = \sqrt{\rho_1(z)} \exp(q_{||} z + i \mathbf{q}_{||} \cdot \mathbf{r}_{||}) .$$

In other words, if one knows the $S(\mathbf{r}, \mathbf{r}')$, the m_1/m_0 ratio gives for the CC excitation operator the exact excitation energy. This statement is, of course, not practically useful since one must have calculated the static form factor first, in which case the excitation energy is known anyway, and no additional information can be gained by studying m_1/m_0. If one uses the m_1/m_0 together with the additional local density approximation for the $S(\mathbf{r}, \mathbf{r}')$, the lowest-lying excitation present in the static form factor is a phonon. Whatever trial function one takes, the estimate m_1/m_0 will always lead to a linear ripplon dispersion relation.

IV. APPLICATIONS

The numerical solution of the RPA equation (2.7) is straightforward, details may be found in Refs. 6 and 10. For technical reasons, we have solved the RPA equation (2.7) not in a half space, but in a slab geometry, assuming translational invariance in the x-y plane, and symmetry around $z = 0$. Hence, our formal results of the preceeding sections are applicable only if the penetration depth of the ripplon is small compared with the slab width. The symmetric slab geometry supports modes that are symmetric or antisymmetric with respect to $z = 0$. A simple criterion whether our approximation of the half space by a finite slab is sufficient is that the symmetric and the antisymmetric modes have the same dispersion relation. All results to be reported here for the HNC theory apply to the largest slab considered in Ref. 10, with a particle number $n = 0.26 \text{Å}^{-2}$ per surface area. Slight differences from the results presented there are due to an improved numerical treatment. The calculations using the phenomenological Skyrme interaction could be performed for economical reasons in a much larger box, hence the agreement between the symmetric and the antisymmetric modes is considerably better.

Our two Skyrme models employ the following parameter sets. For the parameters fitted to the *experimental* data (saturation density $\rho^{\text{expt}} = 0.002\,185 \text{ Å}^{-3}$, saturation

energy $\epsilon_0^{\text{expt}} = -7.15$ K, surface tension $\sigma^{\text{expt}} = 0.274$ K $\mathring{A}^{-2}$), Stringari and Treiner[7] find for the energy functional (1.4) the parameters $b = -887.87$ K $\mathring{A}^3$, $c = 1.041\,489 \times 10^7$ K $\mathring{A}^{3(1+\gamma)}$, $\gamma = 2.8$, and $d = 2376$ K $\mathring{A}^2$. For the parameters fitted to the HNC data (saturation density $\rho_\infty^{\text{HNC}} = 0.0017$ $\mathring{A}^{-3}$, saturation energy $\epsilon_0^{\text{HNC}} = -5.48$ K, surface tension $\sigma^{\text{HNC}} = 0.14$ K $\mathring{A}^{-2}$), the energy functional has the parameters $b = -884$ K $\mathring{A}^3$, $c = 0.8813686 \times 10^7$ K $\mathring{A}^{3(1+\gamma)}$, $\gamma = 2.58$, and $d = 1552$ K $\mathring{A}^2$.

The surface profiles obtained from these two parametrizations of the energy functional and from the HNC calculation of Ref. 10 are shown in Fig. 1. We find that the Skyrme fit to the HNC data generates a slightly more diffuse surface, the agreement between the Skyrme parametrization of the experimental data and the Monte Carlo results of Ref. 8 is somewhat better.[7] Test calculations of the dispersion relation with different surface profiles show that the surface diffuseness has little influence on the ripplon dispersion relation and the wave shape.

For the sake of comparison, we have also calculated the HNC approximation σ_* for the surface energy. We found $2T = 0.03$ K $\mathring{A}^{-2}$ and $(\rho_1' | V_1(z,z') | \rho_1') = 0.13$ K $\mathring{A}^{-2}$, thus giving $\sigma_*^{\text{HNC}} = 0.16$ K $\mathring{A}^{-2}$. This number agrees within the expected accuracy with the surface energy $\sigma^{\text{HNC}} = 0.14$ K $\mathring{A}^{-2}$ obtained from a mass formula (3.15). Note especially that the contribution from the finite range of the particle-hole interaction is overwhelming; a similar result has been found in the nonlocal Skyrme model[7] (1.4). This result makes any attempt to account for surface properties of self-bound finite systems with local mean-field theories highly questionable.

The ripplon dispersion relation corresponding to the two parametrizations of the effective interaction and the HNC result of Ref. 6 are shown in Fig. 2. Also shown

there are the hydrodynamic prediction (1.1) with the experimental and the HNC surface tension, and the Feynman dispersion relation for the bulk phonon. The following observations are made.

(i) The two dispersion relations derived from the Skyrme forces agree rather well with the hydrodynamic dispersion law (1.1) with the corresponding surface energy. The close agreement shows among others that the long-wavelength solution (3.9) of the differential equation (3.8) is also a very good approximation for in the regime $0 \le q_{||} \le 1$ $\mathring{A}^{-1}$. If the solution (3.9) were exact, the hydrodynamic $q_{||}^{3/2}$ law would be rigorous.

(ii) The HNC dispersion relation agrees, for long wavelengths, well with the hydrodynamic and the Skyrme dispersion relation. We attribute the small deviation at least partly to the smaller size of the system, which causes Eq. (3.5) to be satisfied only approximately. Further deviations are caused by the fact that σ and σ_* are, in the HNC approximation, not identical. The close agreement between the HNC and the corresponding "hydrodynamic" law gives confidence that neither the restriction to finite box size is severe, and that σ and σ_* are not very different.

(iii) At a wave number $q_{||} \approx 1$ $\mathring{A}^{-1}$ the Skyrme and the
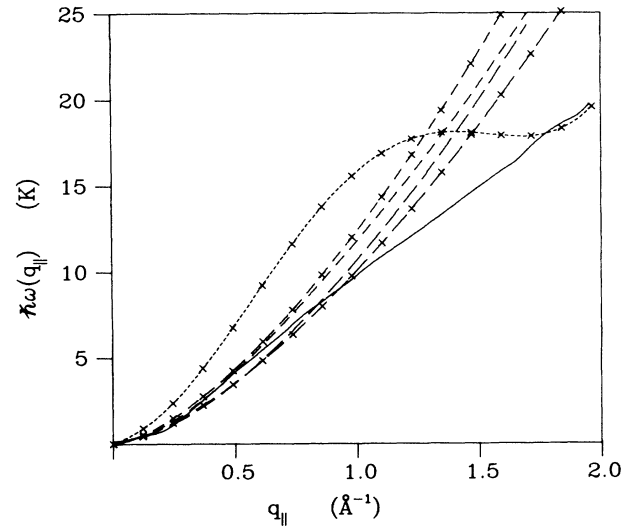
FIG. 2. The dispersion relations of the lowest collective modes are compared with the hydrodynamic predictions and the Feynman dispersion relation. Solid line: ripplon dispersion relation obtained from the HNC response function. Long-dashed line: ripplon dispersion relation obtained from the Skyrme parametrization fitted to the HNC data. Long-dashed line with marks: hydrodynamic ripplon dispersion relation corresponding to the HNC asymptotic density and surface energy. Medium-dashed line: ripplon dispersion relation obtained from the Skyrme parametrization fitted to the experimental data. Medium-dashed line with marks: hydrodynamic ripplon dispersion relation corresponding to the experimental asymptotic density and surface energy. Short-dashed line with marks: phonon dispersion relation obtained in the HNC approximation *at the HNC equilibrium density.*
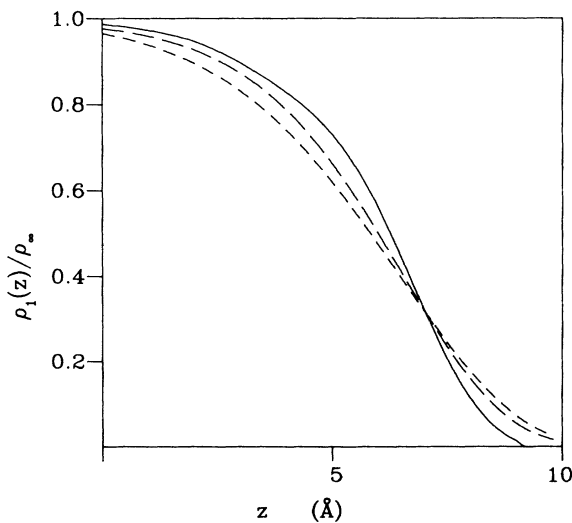

FIG. 1. The density profile $\rho_1(z)/\rho_\infty$ are shown, in the surface region, for the HNC density (solid line), the Skyrme fit to the HNC result (long-dashed line), and the Skyrme fit to the experimental data (short-dashed line).

HNC dispersion relations start to deviate. The Skyrme dispersion relations follow essentially the ripplon dispersion law $\omega \sim q_{||}^{3/2}$; the energy of the lowest collective mode in the HNC theory becomes lower. At still higher momenta, i.e., in the regime of the roton minimum, we find that the energy of the lowest mode agrees very well with the energy of the Feynman *phonon*. In that regime, the lowest lying collective mode is indeed not a surface mode but rather a bulk phonon.[6] The effect is clearly related to the finite-range structure of the particle-hole interaction, which is included in the variational theory, but apparently not adequately treated in the special Skyrme force utilized here. We reiterate that the suppression of the lowest mode by the bulk roton minimum can in our theory be taken only qualitatively, since it is known that the roton minimum is only poorly described by the Feynman dispersion relation. An interesting point is the observation that no further suppression of the dispersion relation below the bulk roton minimum is observed. Such a suppression ("surface roton"[16-19]) has occasionally been suggested from the study of two-dimensional layers. Of course, the systems considered here are three dimensional, and care has been taken to make them large enough such that finite-size effects are suppressed.

Figures 3(a)—3(c) show the shape of the density fluctuations, $\delta \rho_1(z)$, in our three models for $0 \leq q_{||} \leq 1$ Å^{-1}. In the long-wavelength limit $q_{||} = 0$ [Fig. 3(a)], the $\delta \rho_1(z)$ calculated in our three different procedures agree rather well. We attribute the differences essentially to the numerical restrictions imposed by the HNC theory: The calculation of the collective modes in the HNC theory has been done for a symmetric film in a box of only 22 Å width, whereas the Skyrme calculation was done in a box of 40 Å width. The smaller box size for the HNC calculation has the effect that the lowest mode has, if the penetration depth is large, still a considerable overlap with the bulk phonon. The necessity of a rather narrow cutoff in the low-density regime also seems to have the effect that the ripplon does not extend far enough into the low-density tail. This also has the consequence that the calculated ripplon energy in the HNC approximation (Fig. 2) is somewhat too high. We consider for these technical reasons the shape of the density fluctuation predicted by the Skyrme force to be the more reliable one. We note also the $\delta \rho_1(z) \sim d\rho_1(z)/dz$ is quite well satisfied by the Skyrme result, the agreement is somewhat worse for the HNC result.

Profound differences between the $\delta \rho_1(z)$ predicted by the Skyrme and the HNC theory start to appear around $q_{||} = 0.5$ Å^{-1} [Figs. 3(b) and 3(c)]. It appears that the HNC theory predicts a stronger localization of the ripplon in the surface, whereas the Skyrme effective interaction rather predicts a migration of the density fluctuation further into the low-density tail. Of course, one has to keep in mind that the Skyrme force has been constructed to reproduce the *static*, or the long-wavelength properties of the system. The accuracy of the predictions for shorter wavelengths is naturally limited. But it appears that the Skyrme model is adequate essentially in the regime of quantitative validity of our RPA theory of the collective excitations.

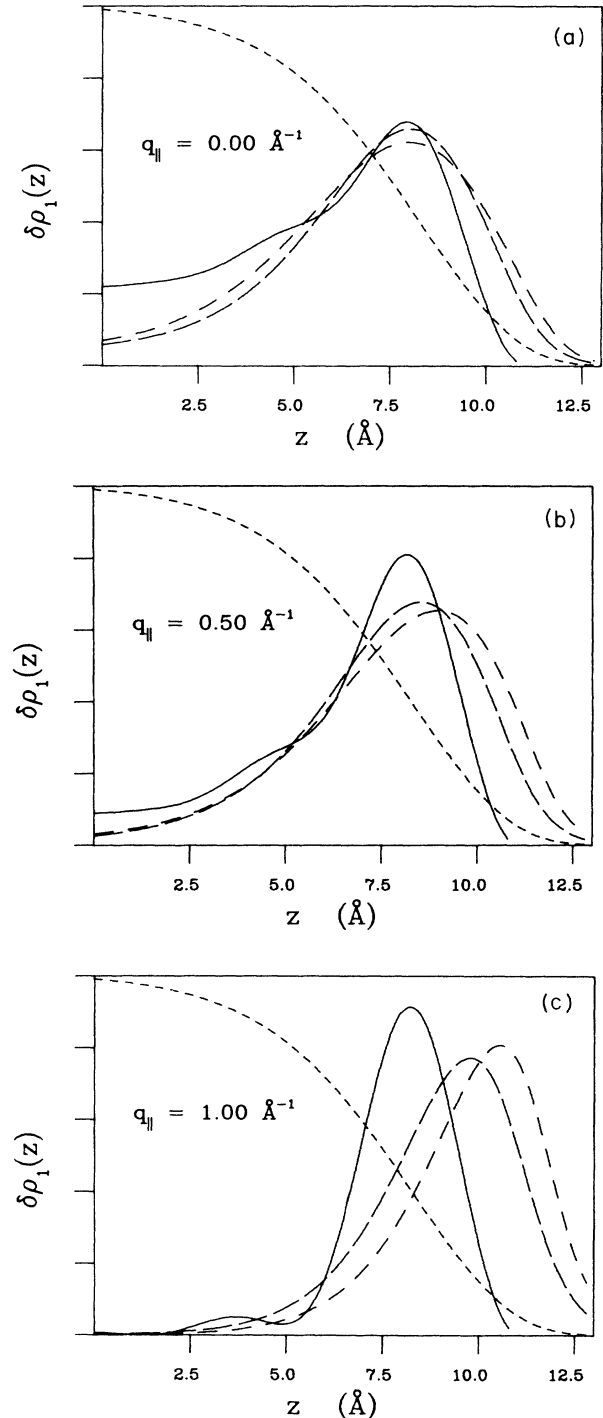Improvements of our description of collective surface

FIG. 3. (a) The shapes $\delta \rho_1(z)$ of the surface excitations obtained in our three calculations are shown in the long-wavelength limit $q_{||} \rightarrow 0$. Solid line: HNC prediction; long-dashed line: prediction from the Skyrme parametrization of the HNC results; Medium-dashed line: prediction from the Skyrme parametrization of the experimental data. For comparison we show also the density profile $\rho_1(z)/\rho_\infty$ obtained from the Skyrme fit of the experimental data. The normalization is arbitrary. (b) Same as Fig. 3(a) for $q_{||} = 0.5$ Å^{-1}. (c) Same as Fig. 3(a) for $q_{||} = 1.0$ Å^{-1}.
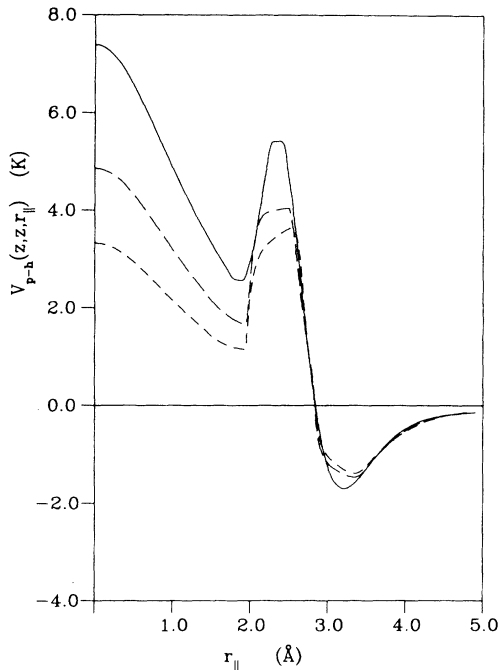
FIG. 4. The HNC particle-hole interaction $V_{p-h}(r_1,r_2)$ is shown for two particles at the same distance from the surface, as a function of their separation parallel to the surface. (a) Solid line: in the bulk limit, (b) long-dashed line: in the surface at 65% of the asymptotic density, and (c) medium-dashed line: in the surface at 34% of the asymptotic density. We found that the particle-hole interaction is almost isotropic.

phenomenological particle-hole interaction $V_{p-h}(r_1,r_2)$. We show in Fig. 4 the HNC result for $V_{p-h}(z,z',r_{||})$ for two particles having the same z coordinate, as a function of their distance $r_{||}$ parallel to the surface. We note also that this function, unlike the pair distribution function $g(r_1,r_2)$, is almost isotropic. Expectedly, the repulsive part of the particle hole interaction becomes weaker in the low-density regime, whereas the attractive part remains essentially the same. One may quite well replace the HNC form for $V_{p-h}(r_1,r_2)$ by an isotropic, phenomenological, $V_{p-h}(r_1,r_2)$ which is constructed according to the arguments of the pseudopotential theory,[15] the relation (3.13) between the pseudopotential and the surface energy, and the restriction (3.6). Note, however, that a "local density approximation" to the particle-hole interaction does not exist in any self-bound system below approximately 65% of the equilibrium density.

The same goal of constructing a better particle-hole interaction can be reached in the HNC theory by including[11,12] three-body correlations and "elementary diagrams." The theory is well understood, but the practical application for surface problems is not yet available.

Improved local pseudopotentials (or particle-hole interactions) can at most lead to the "best" Feynman theory for the ripplon. As a further step, one must improve upon the description of the collective excitation by (in a phenomenological approach) introducing "backflow"[19] corrections, or (in a microscopic theory) allowing for fluctuating two-body correlations.[20] These effects are expected to become significant when the wavelength of the excitation becomes comparable with the interparticle distance.

modes are, if desired, mainly necessary in the regime of momenta $q_{||} \geq 1$ Å^{-1}. One can seek such improvements in both phenomenological and microscopic theories. In a phenomenological theory, one would first incorporate finite-range effects in a more systematic way. An obvious starting point is the generalization of the pseudopotential theory of Aldrich and Pines,[15] i.e., one could introduce a

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