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Theory of inhomogeneous quantum systems.
II. Linear response and collective excitations in Bose systems

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We generalize the variational theory of inhomogeneous Bose systems (presented in the preceding paper) to study collective excitations and the linear response to an external perturbing field. As a by-product of the optimized treatment of the ground state, we obtain dispersion relations and wave shapes for the spectrum of the ripplons and phonons. Results are presented for the surface and volume excitations of free films of ^{4}He. At wave numbers corresponding to the location of the roton minimum in bulk ^{4}He we find a level crossing between surface and volume excitations.

I. INTRODUCTION

In the preceding paper (Ref. 1, hereafter referred to as paper I) we have formulated a general theory of optimized variational wave functions for inhomogeneous Bose systems and applied the theory to calculate static properties of a film of ^{4}He atoms. The present work is devoted to the extension of that theory to the study of linear response to external perturbations and the normal modes of an inhomogeneous Bose liquid. One of the attractive features of optimized variational theories is that they provide simultaneously with a ground-state calculation, a straightforward way to study the low-lying excited states of the system. This is especially true in Bose systems where the structure of the theory is not obscured by the complications of exchange effects present in Fermi systems.[2]

The theory of collective excitations to be presented here leads immediately in the bulk case to the well-known Bijl-Feynman dispersion relation. It provides in the more complicated case of an inhomogeneous system, insight into the dispersion relations and wave shapes of surface and volume excitations. Such dispersion relations have mostly been derived in simplified models of sharp surfaces, due to the lack of detailed information on surface profiles and the anisotropy of the particle-hole interaction. Both of these uncertainties have been eliminated in paper I, and we will find that the dispersion relations of the collective modes are already implicitly contained in the numerical procedure developed there.

We have in paper I written the ground-state wave function in the Feenberg form[3]

$$\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_A)=\exp\left[\frac{1}{2}\sum_{i=1}^{A}u_1(\mathbf{r}_i)+\frac{1}{2}\sum_{\substack{i,j=1\\ i<j}}^{A}u_2(\mathbf{r}_i,\mathbf{r}_j)+\cdots\right] \tag{1.1}$$

and determined, within the hypernetted-chain (HNC) approximation for inhomogeneous systems,[4] the one- and two-body correlation factors by minimization of the ground-state energy

$$\frac{\delta}{\delta u_n(\mathbf{r}_1,\ldots,\mathbf{r}_n)}\left[\frac{\langle\Psi_0|H|\Psi_0\rangle}{\langle\Psi_0|\Psi_0\rangle}\right]=0,\quad n=1,2,\ldots\,. \tag{1.2}$$

Implicit in the HNC approximation is that we ignore all correlation functions except $u_1(\mathbf{r})$ and $u_2(\mathbf{r},\mathbf{r}')$ and the so-called "elementary" diagrams. In Sec. II we briefly review the formalism of paper I. Section III presents the formulation of the theory of linear response in a picture which is related to the random-phase approximation (RPA) in the sense that we assume that only the single-particle wave functions [i.e., $u_1(\mathbf{r})$] depend on the external perturbation. This *ansatz* allows contact to be made with conventional formulations of the RPA since all correlation effects introduced through the static multiparticle correlations can be absorbed in an effective particle-hole interaction. It is worth noting that the optimization of the ground-state correlations is an absolute necessity for a reliable treatment of excited states. Considering excitations on a background of nonoptimized ground states will lead to spurious instabilities reflecting simply the fact that a better variational wave function can be found.

Section IV presents the decomposition of density fluctuations into normal modes. We show that the numerical procedure developed in paper I is intimately related to this normal-mode decomposition, and allows the immediate extraction of dispersion relations. Finally, Sec. V gives results on the propagation of ripplons and phonons in the films of liquid ^{4}He studied in paper I.

II. HYPERNETTED-CHAIN AND EULER-LAGRANGE EQUATIONS

The variational theory of a Bose liquid is based on the Feenberg *ansatz* (1.1) for the many-body wave function. As noted above we shall restrict ourselves in the practical application of the theory to one- and two-body correlations. Most of the formal derivations of Secs. III and IV

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will, however, make no use of this simplification. The only essential assumption entering the present study will be that the one- and two-body correlations are optimized, i.e., that Eq. (1.2) has been solved for $n=1$ and 2.

The primary quantities of physical interest are the one- and two-body densities

$$\rho_1(\mathbf{r}_1)=A\frac{\int d^3r_2\cdots d^3r_A\,|\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_A)|^2}{\int d^3r_1\cdots d^3r_A\,|\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_A)|^2} \tag{2.1}$$

and

$$\rho_2(\mathbf{r}_1,\mathbf{r}_2)=A(A-1)\frac{\int d^3r_3\cdots d^3r_A\,|\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_A)|^2}{\int d^3r_1\cdots d^3r_A\,|\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_A)|^2}\ , \tag{2.2}$$

and the two-body distribution function

$$g(\mathbf{r}_1,\mathbf{r}_2)=\frac{\rho_2(\mathbf{r}_1,\mathbf{r}_2)}{\rho_1(\mathbf{r}_1)\rho_1(\mathbf{r}_2)}\ . \tag{2.3}$$

The unperturbed Hamiltonian is that of a system of identical bosons interacting via a local two-body interaction,

$$H=-\frac{\hbar^2}{2m}\sum_{i=1}^{A}\nabla_i^2+\sum_{\substack{i,j=1\\ i<j}}^{A}v(|\mathbf{r}_i-\mathbf{r}_j|)\ . \tag{2.4}$$

The formalism for the optimal determination of the first two components of the Feenberg function has been developed in paper I. For further reference we review here briefly the essential results of that work.

Using the Feenberg form (1.1) of the wave function, we can write the energy expectation value in the form

$$\begin{aligned}H_{00}=\frac{\langle\Psi_0|H|\Psi_0\rangle}{\langle\Psi_0|\Psi_0\rangle}&=\int d^3r\frac{\hbar^2}{2m}|\nabla\sqrt{\rho_1(\mathbf{r})}|^2\\&\quad+\tfrac{1}{2}\int d^3r_1\,d^3r_2\,\rho_2(\mathbf{r}_1,\mathbf{r}_2)\left[v(|\mathbf{r}_1-\mathbf{r}_2|)-\frac{\hbar^2}{8m}[D(1)+D(2)]u_2(\mathbf{r}_1,\mathbf{r}_2)\right]\ .\end{aligned} \tag{2.5}$$

In Eq. (2.5) we have used the abbreviated notation

$$D(i)=\rho_1^{-1}(\mathbf{r}_i)\nabla_i\rho_1(\mathbf{r}_i)\cdot\nabla_i\ . \tag{2.6}$$

Note that we have in the form (2.5) already eliminated the one-body component $u_1(\mathbf{r})$ of the Feenberg function using the Born-Green-Yvon equation

$$\nabla_1\rho_1(\mathbf{r}_1)=\rho_1(\mathbf{r}_1)\nabla_1u_1(\mathbf{r}_1)+\int d^3r_2\,\rho_2(\mathbf{r}_1,\mathbf{r}_2)\nabla_1u_2(\mathbf{r}_1,\mathbf{r}_2)\ . \tag{2.7}$$

Having used (2.7) to eliminate $u_1(\mathbf{r})$, the energy expectation value contains three unknown functions, i.e., the one- and two-body densities and the two-body part of the Feenberg function. The hypernetted-chain equations

$$g(\mathbf{r}_1,\mathbf{r}_2)=\exp[u_2(\mathbf{r}_1,\mathbf{r}_2)+N(\mathbf{r}_1,\mathbf{r}_2)+E(\mathbf{r}_1,\mathbf{r}_2)]\ , \tag{2.8a}$$

$$N(\mathbf{r}_1,\mathbf{r}_2)=\int d^3r_3\rho_1(\mathbf{r}_3)[g(\mathbf{r}_1,\mathbf{r}_3)-1]X(\mathbf{r}_3,\mathbf{r}_2)\ , \tag{2.8b}$$

$$X(\mathbf{r}_1,\mathbf{r}_2)=g(\mathbf{r}_1,\mathbf{r}_2)-1-N(\mathbf{r}_1,\mathbf{r}_2)\ , \tag{2.8c}$$

provide one relation and the optimization conditions (1.2) provide the other two equations necessary for the determination of these three functions. The set of elementary diagrams $E(\mathbf{r}_1,\mathbf{r}_2)$ is represented by a series of diagrams involving the one-body density $\rho_1(\mathbf{r})$ and the two-body distribution function $g(\mathbf{r}_1,\mathbf{r}_2)$. We will assume throughout this paper the hypernetted-chain approximation $E(\mathbf{r}_1,\mathbf{r}_2)=0$. This approximation is the minimum condition which guarantees that the solutions of the approximate Euler-Lagrange equations have qualitatively the same behavior as the solutions of the exact equations.

It was shown in paper I that the optimization conditions can be cast in the form of a Hartree equation (see also Ref. 5) for the one-body density and an RPA equation for the two-body quantities. The two-body equation was formulated for the set $X(\mathbf{r}_1,\mathbf{r}_2)$ of "non-nodal" diagrams:

$$V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)=\frac{\hbar^2}{4m}\left[[D(1)+D(2)]X(\mathbf{r}_1,\mathbf{r}_2)-\int d^3r_3\,\rho_1(\mathbf{r}_3)X(\mathbf{r}_1,\mathbf{r}_3)D(3)X(\mathbf{r}_3,\mathbf{r}_2)\right] \tag{2.9a}$$

with

$$\begin{aligned}V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)&=g(\mathbf{r}_1,\mathbf{r}_2)v(|\mathbf{r}_1-\mathbf{r}_2|)+\frac{\hbar^2}{2m}\{|\nabla_1[g(\mathbf{r}_1,\mathbf{r}_2)]^{1/2}|^2+|\nabla_2[g(\mathbf{r}_1,\mathbf{r}_2)]^{1/2}|^2\}\\&\quad+\frac{\hbar^2}{4m}[g(\mathbf{r}_1,\mathbf{r}_2)-1]\left[[D(1)+D(2)]N(\mathbf{r}_1,\mathbf{r}_2)+\int d^3r_3\,\rho_1(\mathbf{r}_3)X(\mathbf{r}_1,\mathbf{r}_3)D(3)X(\mathbf{r}_3,\mathbf{r}_1)\right]\ .\end{aligned} \tag{2.9b}$$

The quantity $V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)$ can be identified with the particle-hole (p-h) interaction. The essential new feature is that the variational theory provides an unambiguous prediction of the analytic form of $V_{p\text{-}h}$.

The one-body equation had been cast in the form of a Hartree equation for $\sqrt{\rho_1(\mathbf{r})}$:

$$0=-\frac{\hbar^2}{2m}\nabla^2\sqrt{\rho_1(\mathbf{r}_1)}+[U_{\text{ext}}(\mathbf{r})-\mu+V_{\text{SC}}(\mathbf{r})]\sqrt{\rho_1(\mathbf{r})}\ , \tag{2.10a}$$

$$V_{\text{SC}}(\mathbf{r}_1)=V_{\text{SC}}^{(1)}(\mathbf{r}_1)+V_{\text{SC}}^{(2)}(\mathbf{r}_1)\ , \tag{2.10b}$$

with

$$V_{\text{SC}}^{(1)}(\mathbf{r}_1)=\int d^3r_2\,\rho_1(\mathbf{r}_2)\left[V_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)-\frac{\hbar^2}{8m}\{[g(\mathbf{r}_1,\mathbf{r}_2)-1][D(1)+D(2)]N(\mathbf{r}_1,\mathbf{r}_2)+N(\mathbf{r}_1,\mathbf{r}_2)D(2)X(\mathbf{r}_1,\mathbf{r}_2)\}\right] \tag{2.10c}$$

and

$$V_{\text{SC}}^{(2)}(\mathbf{r}_1)=-\frac{\hbar^2}{16m}D(1)\int d^3r_2\,\rho_1(\mathbf{r}_2)[g(\mathbf{r}_1,\mathbf{r}_2)-1]N(\mathbf{r}_1,\mathbf{r}_2)\ . \tag{2.10d}$$

The HNC/EL (HNC/Euler-Lagrange) equations (2.8)–(2.10) form a closed set of equations that has been solved numerically[1] for various particle numbers for a film of ^{4}He interacting via the Aziz potential.[6]

III. LINEAR RESPONSE TO EXTERNAL PERTURBATIONS

We turn now to the formulation of linear-response theory. Our derivation follows the general formulation of linear response within the theory of correlated basis functions.[2,7] We give here a more rigorous derivation of the identification of $V_{p\text{-}h}(\mathbf{r},\mathbf{r}')$ as the "particle-hole interaction" in the sense that we are able to write the density-density response function in the form

$$\chi^{-1}(\mathbf{r},\mathbf{r}';\omega)=\chi_0^{-1}(\mathbf{r},\mathbf{r}';\omega)-V_{p\text{-}h}(\mathbf{r},\mathbf{r}')\ , \tag{3.1}$$

where $\chi_0(\mathbf{r},\mathbf{r}';\omega)$ is the response function of a suitably defined noninteracting system. The earlier studies of excited states with correlated wave functions have mainly concentrated on Fermi systems. Of course, the same formalism can be developed for Bose systems, and the absence of exchange effects leads to substantial simplifications. The inhomogeneous Bose system is a nontrivial and physically interesting case where the manipulations of Refs. 7 and 2 can be carried out analytically.

The only additional assumption beyond the Feenberg form (1.1) of the wave function is that only the one-body component of the Feenberg function, $u_1(\mathbf{r})$, is affected by the external perturbation. This assumption is based on common wisdom gained from studies of homogeneous quantum liquids for which the RPA leads in fact to the correct long-wavelength limit. To what extent the same assumption is justified for an inhomogeneous system is not clear.[8] The assumption of *static* two-body correlations $u_2(\mathbf{r},\mathbf{r}')$ limits the regime of validity of our theory to wavelengths which are long compared with the average distance between two particles, i.e., to the regime of validity of the Bijl-Feynman dispersion relation.

We assume that the system is subjected to a time-dependent external potential $U_{\text{ext}}(\mathbf{r},t)$. The system responds by a time-dependent change of the one-body part of the wave function $u_1(\mathbf{r})$:

$$u_1(\mathbf{r},t)=u_1(\mathbf{r})+\delta u_1(\mathbf{r},t)\ . \tag{3.2}$$

Following the derivation of the RPA via a least-action principle (see, e.g., Ref. 9), we write a correlated wave function of the form

$$|\Psi(t)\rangle=\exp\left[-iH_{00}\frac{t}{\hbar}\right]|\Psi_0(t)\rangle\ , \tag{3.3}$$

$$|\Psi_0(t)\rangle=\exp\left[\tfrac{1}{2}\sum_i^A u_1(\mathbf{r}_i,t)+\tfrac{1}{2}\sum_{\substack{i<j\\ i,j=1}}^A u_2(\mathbf{r}_i,\mathbf{r}_j)\right]\Big/\mathcal{N}\ , \tag{3.4}$$

where $\mathcal{N}$ is the norm. The wave function of the perturbed system is determined by the action principle

$$\delta\mathcal{L}=\delta\int_{t_0}^{t_1}dt\left\langle\Psi(t)\middle|\left[H+U_{\text{ext}}(\mathbf{r},t)-i\hbar\frac{\partial}{\partial t}\right]\middle|\Psi(t)\right\rangle=0\ . \tag{3.5}$$

Using our assumption that only the one-body component $u_1(\mathbf{r},t)$ is time dependent, we find

$$\mathcal{L}=\int_{t_0}^{t_1}dt\left[\langle\Psi_0(t)|(H-H_{00})|\Psi_0(t)\rangle+\int d^3r\,\rho_1(\mathbf{r},t)\left[U_{\text{ext}}(\mathbf{r},t)-\frac{i\hbar}{4}[\dot{u}_1(\mathbf{r},t)-\dot{u}_1^*(\mathbf{r},t)]\right]\right]\ . \tag{3.6}$$

Since the perturbing field is assumed to be weak, we can linearize the equations of motion for $\delta u_1(\mathbf{r},t)$. To this end, we have to expand the action integral (3.6) to second order in $\delta u_1(\mathbf{r},t)$. Considerable simplification is gained by the assumption that both static correlation factors, $u_1(\mathbf{r})$ and $u_2(\mathbf{r},\mathbf{r}')$, have been obtained by optimization, i.e., by the solution of

Eqs. (1.2) for $n=1$ and 2. The first condition, i.e., the optimization of the one-body correlations, has the consequence that no linear terms in $u_1(\mathbf{r},t)$ occur in (3.6). The second condition ($n=2$) has the consequence

$$\frac{\int d^3r_1\cdots d^3r_A\,|\Psi_0|^2(H-H_{00})\sum_{i<j}\delta u_1(\mathbf{r}_i,t)\delta u_1(\mathbf{r}_j,t)}{\int d^3r_1\cdots d^3r_A\,|\Psi_0(\mathbf{r}_1,\ldots,\mathbf{r}_A)|^2}=\int d^3r_1\,d^3r_2\frac{\delta H_{00}}{\delta u_2(\mathbf{r},\mathbf{r}')}\delta u_1(\mathbf{r},t)\delta u(\mathbf{r}',t)=0\,. \tag{3.7}$$

Using (3.7) we find

$$\langle\Psi_0(t)\,|\,(H-H_{00})\,|\,\Psi_0(t)\rangle=\frac{\hbar^2}{8m}\int d^3r\,\rho_1(r)\,|\nabla\delta u_1(\mathbf{r},t)|^2+O(|\delta u_1(\mathbf{r},t)|^3)\,. \tag{3.8}$$

Another useful relation is the connection between a change in the physical density and a change in the one-body wave function:

$$\delta\rho_1(\mathbf{r},t)=\rho_1(\mathbf{r})\mathrm{Re}\delta u_1(\mathbf{r},t)+\int d^3r'[\rho_2(\mathbf{r},\mathbf{r}')-\rho_1(\mathbf{r})\rho_1(\mathbf{r}')]\mathrm{Re}\delta u_1(\mathbf{r}',t)\,. \tag{3.9}$$

Using the (exact) chain equation (2.8b), we find also

$$\mathrm{Re}\delta u_1(\mathbf{r},t)=\delta\rho_1(\mathbf{r},t)/\rho_1(\mathbf{r})-\int d^3r'\,X(\mathbf{r},\mathbf{r}')\delta\rho_1(\mathbf{r}',t)\,. \tag{3.10}$$

In passing we note that Eqs. (3.8)—(3.10) lead, for adiabatic changes in the physical one-body density, immediately to

$$V_{p\text{-}h}(\mathbf{r},\mathbf{r}')=\frac{\delta^2H_{00}}{\delta\rho_1(\mathbf{r})\delta\rho_1(\mathbf{r}')},\quad \mathbf{r}\neq\mathbf{r}'\,. \tag{3.11}$$

We are now ready to derive the linearized equation of motion. Inserting (3.8) and (3.9) into the action integral (3.6) we find in second order in the variation of the one-body wave function

$$\begin{aligned}\mathscr{L}=\int_{t_0}^{t_1}dt\Bigg\{&\int d^3r\,\rho_1(\mathbf{r})\left[\frac{\hbar^2}{8m}|\nabla\delta u_1(\mathbf{r},t)|^2+\left[U_{\mathrm{ext}}(\mathbf{r},t)+\frac{\hbar}{2}\mathrm{Im}\delta\dot u_1(\mathbf{r},t)\right]\mathrm{Re}\delta u_1(\mathbf{r},t)\right]\\&+\int d^3r\,d^3r'[\rho_2(\mathbf{r},\mathbf{r}')-\rho_1(\mathbf{r})\rho_1(\mathbf{r}')]\left[U_{\mathrm{ext}}(\mathbf{r},t)+\frac{\hbar}{2}\mathrm{Im}\delta\dot u_1(\mathbf{r},t)\right]\mathrm{Re}\delta u_1(\mathbf{r}',t)\Bigg\}\,.\end{aligned} \tag{3.12}$$

Evaluating the stationarity conditions leads to

$$\begin{aligned}0=\frac{\delta\mathscr{L}}{\delta u_1^*(\mathbf{r})}=&-\frac{\hbar^2}{8m}\nabla[\rho_1(\mathbf{r})\cdot\nabla\delta u_1(\mathbf{r},t)]+\tfrac{1}{2}\rho_1(\mathbf{r})\left[U_{\mathrm{ext}}(\mathbf{r},t)-\frac{i\hbar}{2}\delta\dot u_1(\mathbf{r},t)\right]\\&+\tfrac{1}{2}\int d^3r'[\rho_2(\mathbf{r},\mathbf{r}')-\rho_1(\mathbf{r})\rho_1(\mathbf{r}')]\left[U_{\mathrm{ext}}(\mathbf{r}',t)-\frac{i\hbar}{2}\delta\dot u(\mathbf{r}',t)\right]\,,\end{aligned} \tag{3.13}$$

$$\begin{aligned}0=\frac{\delta\mathscr{L}}{\delta u_1(\mathbf{r})}=&-\frac{\hbar^2}{8m}\nabla[\rho_1(\mathbf{r})\cdot\nabla\delta u_1^*(\mathbf{r},t)]+\tfrac{1}{2}\rho_1(\mathbf{r})\left[U_{\mathrm{ext}}(\mathbf{r},t)+\frac{i\hbar}{2}\delta\dot u_1^*(\mathbf{r},t)\right]\\&+\tfrac{1}{2}\int d^3r'[\rho_2(\mathbf{r},\mathbf{r}')-\rho_1(\mathbf{r})\rho_1(\mathbf{r}')]\left[U_{\mathrm{ext}}(\mathbf{r}',t)+\frac{i\hbar}{2}\delta\dot u_1^*(\mathbf{r}',t)\right]\,.\end{aligned} \tag{3.14}$$

The further manipulations follow a standard pattern: We Fourier decompose the time dependence of the external field

$$U_{\mathrm{ext}}(\mathbf{r},t)=U_{\mathrm{ext}}(\mathbf{r})(e^{i\omega t}+e^{-i\omega t})\,, \tag{3.15}$$

and the one-body function

$$\delta u(\mathbf{r},t)=x(\mathbf{r})e^{-i\omega t}+y(\mathbf{r})e^{i\omega t}\,. \tag{3.16}$$

Some elementary manipulations, in which the chain equation (2.8b) and the Euler-Lagrange equation (2.9a) are used, lead to the coupled system of equations

$$\begin{aligned}\frac{\hbar^2}{4m}\nabla\left[\rho_1(\mathbf{r})\nabla\frac{\delta\rho_1(\mathbf{r})}{\rho_1(\mathbf{r})}\right]&-\rho_1(\mathbf{r})\int d^3r'\,V_{p\text{-}h}(\mathbf{r},\mathbf{r}')\delta\rho_1(\mathbf{r}')\\&=\rho_1(\mathbf{r})\left[U_{\mathrm{ext}}(\mathbf{r})-\frac{\hbar\omega}{4}[x(\mathbf{r})-y(\mathbf{r})]\right]\,,\end{aligned} \tag{3.17}$$

$$\frac{\hbar^2}{4m}\nabla\{\rho_1(\mathbf{r})\cdot\nabla[x(\mathbf{r})-y(\mathbf{r})]\}=-\hbar\omega\delta\rho_1(\mathbf{r})\,,$$

determining the response $\delta\rho(\mathbf{r})$ to the external field (3.15). In order to cast our results in a more familiar form and to clarify the connection to the RPA, we introduce the one-body operator[1]

$$H(1)=-\frac{\hbar^2}{2m}\frac{1}{\sqrt{\rho_1(\mathbf{r})}}\nabla\rho_1(\mathbf{r})\cdot\nabla\frac{1}{\sqrt{\rho_1(\mathbf{r})}}\ . \tag{3.18}$$

[Note that $H(1)$ is positive semidefinite.] We obtain by eliminating $x(\mathbf{r})-y(\mathbf{r})$ from Eqs. (3.17) the density response $\delta\rho_1(\mathbf{r})$ to an external perturbing field $U_{\text{ext}}(\mathbf{r})$. In symbolic notation we obtain

$$\left[\frac{1}{2}\frac{1}{\sqrt{\rho_1}}H^{-1}(1)[-H^2(1)+\hbar^2\omega^2]\frac{1}{\sqrt{\rho_1}}-V_{p\text{-}h}\right]\times\delta\rho=U_{\text{ext}}\ , \tag{3.19}$$

where we can identify the expression in the square brackets with the inverse of the density-density response function. Equation (3.19) has the structure (3.1) with

$$\chi_0=2\sqrt{\rho_1}H(1)[\hbar^2\omega^2-H^2(1)]^{-1}\sqrt{\rho_1}\ . \tag{3.20}$$

An explicit coordinate-space representation of $\chi_0(\mathbf{r},\mathbf{r}';\omega)$ may be obtained using the spectral decomposition

$$H(1)(\mathbf{r},\mathbf{r}')=\sum_i e_i\varphi_i(\mathbf{r})\varphi_i(\mathbf{r}') \tag{3.21}$$

from which we find

$$\chi_0(\mathbf{r},\mathbf{r}';\omega)=\sum_i\frac{2\sqrt{\rho_1(\mathbf{r})}\varphi_i(\mathbf{r})e_i\varphi_i(\mathbf{r}')\sqrt{\rho_1(\mathbf{r}')}}{\hbar^2\omega^2-e_i^2}\ . \tag{3.22}$$

To summarize the analysis of this section, we have shown how the linear response of the system to external perturbations is treated within the variational theory. Due to our assumption that only the one-body component of the correlation operator is affected by the external field, we have obtained a result which is structurally identical to the RPA. While the formal structure of our result is quite plausible, it is worth pointing out the important difference with an ordinary RPA treatment: The one-body Hamiltonian and the particle-hole interaction are usually considered as a phenomenological input to the RPA. The variational theory provides unambiguous microscopic expressions for these quantities. The particle-hole interaction and the normal modes of the system are natural ingredients in an optimized treatment of the ground state. Hence we encounter again a situation for which the ground-state theory provides all the raw material necessary for the study of the low-lying excitations.

IV. NORMAL-MODE DECOMPOSITION

The key to a study of the elementary excitations is a normal-mode decomposition of the density fluctuations. We explain the procedure in some detail since it provides at the same time the formal background of the method used in paper I for the numerical optimization. In fact, the algorithm for the two-body functions $X(\mathbf{r},\mathbf{r}')$ and $g(\mathbf{r},\mathbf{r}')$ turns out to be equivalent to the solution of the RPA equation for a given $V_{p\text{-}h}(\mathbf{r},\mathbf{r}')$, the construction of the response function, and the computation of the static form factor via the fluctuation-dissipation theorem.

The derivations of this section are exact in the sense that they do not rely on a specific discretization of the problem. The essential difference between the formulation given here and its numerical implementation is that the discretization on a finite mesh leads to cutoffs in all spectra and eigenfunction expansions.

We have arrived in the preceding section (see also paper I) at the conclusion that the operator

$$H(1)=-\frac{\hbar^2}{2m}\frac{1}{\sqrt{\rho_1(\mathbf{r})}}\nabla\rho_1(\mathbf{r})\cdot\nabla\frac{1}{\sqrt{\rho_1(\mathbf{r})}} \tag{4.1}$$

is the proper one-body Hamiltonian of the system. It is positive-semidefinite; the lowest eigenstate of $H(1)$ is just the square root of the physical one-body density.

We found [cf. Eq. (3.19)] that the density-density response function, for example, can be written in the form

$$\chi(\mathbf{r},\mathbf{r}';\omega)=2\sqrt{\rho_1(\mathbf{r})}H^{1/2}(1)[\hbar^2\omega^2-H^2(1)-2H^{1/2}(1)\hat{V}_{p\text{-}h}H^{1/2}(1)]^{-1}H^{1/2}(1)\sqrt{\rho_1(\mathbf{r}')}\ , \tag{4.2}$$

where

$$\hat{V}_{p\text{-}h}(\mathbf{r},\mathbf{r}')=\sqrt{\rho_1(\mathbf{r})}V_{p\text{-}h}(\mathbf{r},\mathbf{r}')\sqrt{\rho_1(\mathbf{r}')}\ . \tag{4.3}$$

Usually all quantities are expanded in terms of the eigenfunctions of $H(1)$; we will find, however, that the expansion actually never needs to be carried out explicitly. The operator $H^{1/2}(1)$ is defined by

$$H^{1/2}(1)H^{1/2}(1)=H(1)\ , \tag{4.4}$$

and the existence of $H^{1/2}(1)$ is guaranteed by the symmetry and positive semidefiniteness of $H(1)$.

Consider now the eigenvalue problem

$$[H(1)+2\hat{V}_{p\text{-}h}]H(1)\psi^{(l)}=\hbar^2\omega_l^2\psi^{(l)}\ . \tag{4.5}$$

To be definite we may think of Eq. (4.5) as an eigenvalue problem in coordinate space representation. (The reason that we write the eigenvalues as squares will become obvious shortly.) The eigenvalues of Eq. (4.5) are real. To prove this, we use $H^{1/2}(1)$ to map the eigenvalue problem (4.5) on the symmetric problem

$$H^{1/2}(1)[H(1)+2\hat{V}_{p\text{-}h}]H^{1/2}(1)\xi^{(l)}=\hbar^2\omega_l^2\xi^{(l)}\ . \tag{4.6}$$

Each eigenvector $\xi^{(l)}$ of the problem (4.6) is related to an eigenvector $\psi^{(l)}$ of (4.5) through

$$\xi^{(l)}=H^{1/2}(1)\psi^{(l)}\ . \tag{4.7}$$

From the orthonormality of the $\xi^{(l)}$, we conclude that the $\psi^{(l)}$ are orthonormal in the metric defined by $H(1)$, i.e.,

$$(\psi^{(l)}\,|\,H(1)\,|\,\psi^{(l')})=\delta_{ll'}\ . \tag{4.8}$$

The remaining manipulations are quite straightforward. Inserting the spectral representation

$$[H^{1/2}(1)[2\hat{V}_{p\text{-}h}+H(1)]H^{1/2}(1)](\mathbf{r},\mathbf{r}') = \sum_l \xi^{(l)}(\mathbf{r})\hbar^2\omega_l^2\xi^{(l)}(\mathbf{r}') \tag{4.9}$$

in the response function (4.2), we find

$$\chi(\mathbf{r},\mathbf{r}';\omega)=2\sqrt{\rho_1(\mathbf{r})}\sum_l \left[[H(1)\psi^{(l)}(\mathbf{r})]\frac{1}{\hbar^2\omega^2-\hbar^2\omega_l^2}[H(1)\psi^{(l)}(\mathbf{r}')] \right]\sqrt{\rho_1(\mathbf{r}')}\ . \tag{4.10}$$

The most important feature of the representation (4.10) is that neither the eigenstates of the one-body Hamiltonian $H(1)$ nor the states $\xi^{(l)}$ need to be calculated. The solution of the RPA equation is, therefore, reduced to the diagonalization of a real, nonsymmetric matrix with real eigenvalues.

The static form factor follows from (4.10) via the fluctuation-dissipation theorem

$$\begin{aligned} S(\mathbf{r},\mathbf{r}') &= \frac{1}{\sqrt{\rho_1(\mathbf{r})\rho_1(\mathbf{r}')}}\int \frac{d(\hbar\omega)}{2\pi}\mathrm{Im}\chi(\mathbf{r},\mathbf{r}';\omega) \\ &= \sum_l \frac{1}{\hbar\omega_l}[H(1)\psi^{(l)}(\mathbf{r})][H(1)\psi^{(l)}(\mathbf{r}')]\ . \end{aligned} \tag{4.11}$$

From Eq. (4.11) it is also clear why we wrote the eigenvalues of (4.5) and (4.6) as squares. The positivity of the ω_l^2 is a necessary condition for the existence of solutions of the RPA equations for a given particle-hole interaction. It is the generalization of the positivity condition of the compressibility in the bulk system.

We may now also construct the second two-body quantity of interest, the set $X(\mathbf{r},\mathbf{r}')$ of non-nodal diagrams. Either using the above line of arguments for Eqs. (2.10), or by direct verification of the chain equation, we find

$$\delta(\mathbf{r}-\mathbf{r}')-\sqrt{\rho_1(\mathbf{r})}X(\mathbf{r},\mathbf{r}')\sqrt{\rho_1(\mathbf{r}')} = \sum_l \hbar\omega_l\psi^{(l)}(\mathbf{r})\psi^{(l)}(\mathbf{r}')\ . \tag{4.12}$$

Equations (4.11) and (4.12) were used in paper I for the numerical construction of the solutions of the Euler-Lagrange equation.

Let us finally derive the identification of the states $H(1)\psi^{(l)}(\mathbf{r})$ with the normal modes. The normal modes are the solutions of the homogeneous equation

$$\int d^3r'\chi^{-1}(\mathbf{r},\mathbf{r}';\omega)\delta\rho(\mathbf{r}';\omega)=0\ . \tag{4.13}$$

In the above spectral decomposition we find

$$\chi^{-1}(\mathbf{r},\mathbf{r}';\omega)=\frac{1}{2}\sum_l \frac{\psi^{(l)}(\mathbf{r})}{\sqrt{\rho_1(\mathbf{r})}}(\hbar^2\omega^2-\hbar^2\omega_l^2)\frac{\psi^{(l)}(\mathbf{r}')}{\sqrt{\rho_1(\mathbf{r}')}} \tag{4.14}$$

and the normal modes are, due to (4.8),

$$\begin{aligned} \delta\rho_{(l)}^{1/2}(\mathbf{r},\omega) &= \frac{\delta\rho_{(l)}(\mathbf{r},\omega)}{2\sqrt{\rho_1(\mathbf{r})}} \\ &= [H(1)\psi^{(l)}(\mathbf{r})]\delta(\omega-\omega_l)\ . \end{aligned} \tag{4.15}$$

V. RESULTS

We have seen in the preceding derivations that, within the "correlated RPA" picture, the shape and the dispersion relations of the normal modes are a natural byproduct of the optimized treatment of the ground state. We reiterate that the validity of the results to be discussed here is limited by the validity of the Bijl-Feynman dispersion relation in bulk ^{4}He. To the extent that time-dependent two-body correlations influence the dispersion relation of the normal modes significantly at long wavelengths, the regime of validity can be even smaller. An additional drawback of our strictly microscopic approach is that there is no clean way to improve the agreement with experimental data by inclusion of phenomenological modifications. The most immediate consequence is that the results are limited by the accuracy of the calculated velocity of sound and surface energy within the optimized HNC approximation. Our theory provides on the other hand a straightforward way of abandoning oversimplifications such as a sharp profile, or a zero-range particle-hole interaction. We note also that our results should be better for very thin films which have a low central density since the HNC approximation becomes increasingly better with decreasing density.

The collective excitations in films are classified according to their driving mechanism and their wavelength compared with the film thickness. Surface modes can be driven by the interaction of the fluid with a supporting substrate (third sound),[10] or by the surface tension (ripplons).[11] We have considered in this work a free surface, in other words there is no third sound. The ripplons are called "shallow" if their wavelength is long compared with the film thickness, otherwise they are called "deep." The normal modes discussed in this work are ripplons to the extent that they can be identified as surface excitations. Volume modes are to be identified with zero sound. Of course, the notion of shallow and deep ripplons can be introduced only *a posteriori.* For an excellent discussion of the physical mechanisms of the different modes in a phenomenological picture, see Ref. 12.

As a representative example we discuss here results for the elementary excitations of a system of ^{4}He atoms which is translationally invariant in the x and y directions, and symmetric about $z=0$. The specific example to be studied here is the one with

$$n=\int_{-\infty}^{\infty}dz\,\rho_1(z)=0.22\ \text{Å}^{-2} \tag{5.1}$$

particles per unit area. The central density of this film is $\rho_c = 0.0165$ Å^{-3}, its width is roughly 14 Å. The results are qualitatively very similar for the other calculations performed in paper I, we will give evidence for this point further below.

The elementary excitations in the film may be classified according to their symmetry with respect to the $z=0$ plane. Translational invariance in the direction parallel to the surface allows one to study the collective modes as functions of their momentum $q_{||}$ in that plane. Figure 1 shows the dispersion relation for the four lowest-lying modes. The lowest one and the third one are symmetric with respect to $z=0$, the second and the fourth one are antisymmetric. Attention is now directed to the following interesting features.

(i) The two lowest-lying modes are identical except for the very long-wavelength region. The branch point of the dispersion relation of these modes is at a wavelength of roughly 15 Å, which is comparable with the film thickness. This branch point may serve to distinguish between deep and shallow ripplons, if such a distinction is desired. A similar branch between the dispersion relation of symmetric and antisymmetric modes can be observed for the two higher-lying states at about twice the wave number.

(ii) Beyond the branch point and up to a wave number of about 1.6 Å^{-1}, the dispersion relation of the lowest symmetric and antisymmetric modes are indistinguishable. This is characteristic for a pure surface excitation in which the excitation of the two surfaces of the film are essentially decoupled.

(iii) All four modes become very close at a wave number of about 1.8 Å^{-1}. Beyond that point, the four modes split and are roughly equidistant in energy.

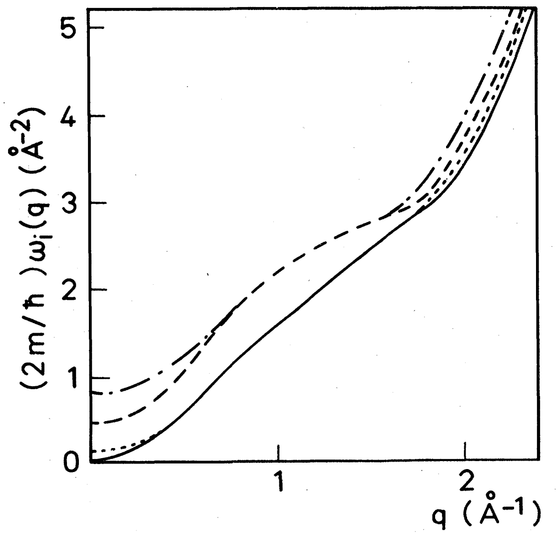

FIG. 1. Dispersion relation of the lowest-lying symmetric mode (solid line), the lowest-lying antisymmetric mode (short-dashed line), the second symmetric (long-dashed line), and the second antisymmetric mode (dashed-dotted line) as a function of wave number $q_{||}$ for a symmetric film of ^{4}He atoms as described in the text.

The physical interpretation of these effects becomes quite clear from considering the spatial shape $\delta\rho_{(l)}^{1/2}(z)$ of these modes. These are shown, for several momenta parallel to the surface, in Figs. 2(a)—2(e). Figure 2(a) shows the amplitudes of the collective modes at $q_{||}=0$. The two lowest-lying excitations are clear surface modes. The identification of two higher-lying modes as surface excitations is not so clear, but suggestive. The distinction between the lowest symmetric and the lowest antisymmetric shallow modes is due to the fact that the penetration depth is not small compared with the film thickness.

The picture becomes clearer at the large momenta. As a typical example, we show in Fig. 2(b) the same amplitudes for a momentum $q_{||}=0.632$ Å^{-1}. All four modes are clearly surface excitations. The picture is supported by the fact that the symmetric and the antisymmetric modes are virtually indistinguishable in energy and shape. Note that we are already in a region where the wavelength is much smaller than the film thickness, i.e., the shallow excitations shown in Fig. 2(b) represent deep ripplons.

Shortly before the closest encounter of the four dispersion relations [see Fig. 2(c) for $q_{||}=1.625$ Å^{-1}], the two lowest-lying modes are still very close, and suggest their identification at surface excitations. The same identification cannot be made any more for the higher-lying excitations. A dramatic change in the shape of the elementary excitations occurs between $q_{||}=1.625$ Å^{-1} and $q_{||}=1.800$ Å^{-1} [Fig. 2(d)]. The lowest-lying mode is now a clear volume excitation, and none of the higher ones is particularly concentrated in the surface. We conclude that the elementary excitations with wave numbers larger than 1.8 Å^{-1} are zero sound modes. This conclusion is supported by the shape of the normal modes with larger wave numbers [cf. Fig. 2(e)].

To summarize, we find that the low-lying collective modes with wave numbers up to roughly 1.6 Å^{-1} are surface excitations. In a very small transition region, the lowest-lying modes turn into compression modes. The picture is rather independent of the film thickness. To support this we show in Fig. 3 the dispersion relations of the lowest-lying collective modes for the particle numbers $n=0.18$ Å^{-2} and $n=0.26$ Å^{-2}.

A qualitative understanding of the rapid change of the character of the lowest-lying excitation may be gained from recalling that the Bijl-Feynman dispersion relation has a broad saddle in the regime of our transition point. It is therefore tempting to identify the rapid change in the shape of the lowest-lying mode as a level-crossing phenomenon between ripplons and zero sound.

Guyer *et al.*[12] suggest for the dispersion relation of the surface-tension driven modes an interpolation formula of the form

$$\omega^2(k) = \frac{\sigma k^3}{m\rho}\tanh(kd) , \tag{5.2}$$

where ρ is a typical density within the film σ the surface tension, and d the thickness of the film. The limits of this formula for $kd \gg 1$ and $kd \ll 1$ were derived in a simplified model of a sharp surface; hence Eq. (5.2) is not

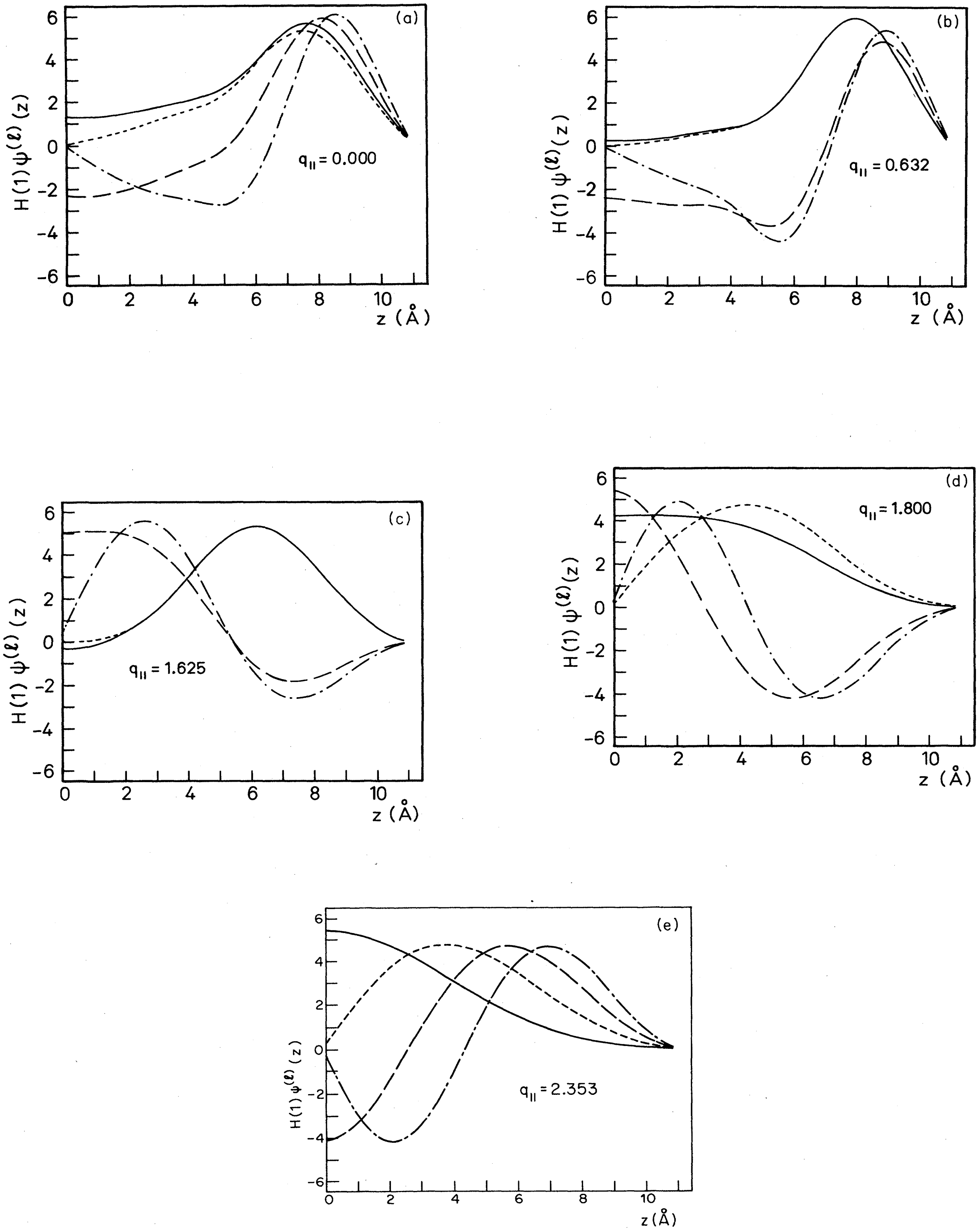

FIG. 2. (a) Spatial shape $\delta\rho_{(l)}^{1/2}(z)=H(1)\psi^{(l)}(z)$ of the lowst-lying symmetric mode (solid line), the lowest-lying antisymmetric mode (short-dashed line), the second symmetric (long-dashed line), and the second antisymmetric mode (dashed-dotted line) in the long-wavelength limit $q_{\parallel}=0$. (b) Same as (a) for $q_{\parallel}=0.632$ Å^{-1}. (c) Same as (a) for $q_{\parallel}=1.625$ Å^{-1}. (d) Same as (a) for $q_{\parallel}=1.800$ Å^{-1}. (e) Same as (a) for $q_{\parallel}=2.353$ Å^{-1}.

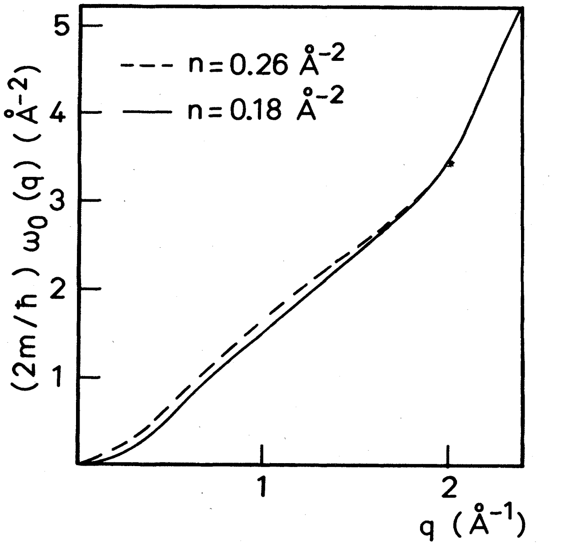

FIG. 3. Dispersion relation of the lowest symmetric mode for films of particle numbers $n=0.26$ Å^{-2} (dashed line) and $n=0.18$ Å^{-2} (solid line) per unit surface area.

immediately applicable in our present case of a diffuse film. Nevertheless, we find, inserting our results $\rho_c=0.0165$ Å^{-3}, $\sigma=0.14$ K Å^{-2}, and $d=14$ Å, that the formula (5.2) fits the dispersion relation of the lowest symmetric mode shown in Fig. 1 extremely well in a regime of wave numbers $0\le q\le 0.9$ Å^{-1}. Beyond that region, the ripplon dispersion relation is shifted to somewhat lower energies by the approaching phonon branch.

Let us conclude with a short remark on the experimental implications of the present work. Our studies are at the present level only semiquantitative. This is mainly due to two effects: (i) the calculated surface tension is too low, and (ii) the Bijl-Feynman dispersion relation does not well reproduce the roton minimum. More realistic dispersion relations for the phonon and the ripplon should both work in the direction that the level crossing occurs at lower wave numbers. It is hard to estimate the wave number where the level crossing should occur since three is also a substantial repulsion at work between the levels of the ripplon and the zero sound. Superimposing the interpolation formula (5.1) and the Bijl-Feynman dispersion relation would suggest that the level crossing should occur in our calculation at a wave number of about 1 Å^{-1}. In fact, this is roughly the wave number where the interpolation formula (5.1) becomes invalid. Superimposing in the same manner the ripplon dispersion relation obtained from Eq. (5.1) with the *experimental* surface tension $\sigma=0.273$ K Å^{-2} with the *experimental* zero-sound dispersion relation, and taking some level repulsion into account, suggests that the actual level crossing occurs at wave numbers between $q_{\parallel}$ between 0.7 and 1.0 Å^{-1}. The prospects of making more quantitative predictions on the excitation spectra of thinner films ($\rho_c<0.014$ Å^{-3}) are much better since the HNC approximation is more accurate in the low-density regime.

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