PHYSICAL REVIEW B VOLUME 38, NUMBER 1 1 JULY 1988

Impurity states in liquid-helium films

E. Krotscheck, M. Saarela,* and J. L. Epstein
Center for Theoretical Physics, Department of Physics, Texas A&M University, College Station, Texas 77843

(Received 14 December 1987)

We study the properties of impurity atoms of ^{3}He and spin-polarized hydrogen isotopes in films of liquid ^{4}He adsorbed to a substrate. We find that a ^{3}He impurity has three or more bound states, two of which we associate with surface states. Hydrogen atoms are very weakly bound and have only one bound state. Depending on the thickness of the film, deuterium impurities may form two surface bound states, or may penetrate to the substrate. For all cases studied, tritium impurities also tend to penetrate to the substrate. We develop the theory of the formation of "self-bound" or "polaron" states, but we see no evidence that such states can become energetically favorable.

I. INTRODUCTION

The study of adsorbed films of quantum liquids is presently an active area of a variety of experimental studies.[1] These are, among others, the investigation of the excited states by neutron-scattering experiments,[2,3] measurements of the third-sound velocity,[4,5] and electron-mobility experiments[6,7] which provide information on the layer structure of such films.[8,9]

A further exciting experimental aspect of adsorbed helium films is the opportunity to generate two-dimensional Fermi systems of electrons and ^{3}He atoms. Electrons bound to helium surfaces have been investigated by Dahm and collaborators.[1]

Systems of ^{3}He atoms adsorbed on the free surface[10] or to films of liquid ^{4}He are of special interest.[11-13] This system is an (almost) ideal two-dimensional Fermi liquid; a layer of ^{3}He atoms with a density less than that of a monolayer may be treated as a dilute system of impurities. On the surface of bulk ^{4}He, the impurity particles have a binding energy[14,15] of about 2.2 K relative to the state where the ^{3}He atom exists in bulk ^{4}He. The physical picture becomes more complicated for the case of ^{3}He atoms on films of ^{4}He. A second "layered" state may exist within the medium,[16] which generates a mixture of a quasi-two-dimensional and a three-dimensional system. The ability to change the properties of the ^{3}He with the thickness of the ^{4}He background makes these systems experimentally very interesting,[17] and poses a challenging theoretical problem.

In the past there have been several studies of the interaction between a homogeneous or inhomogeneous helium liquid and an impurity atom.[18-28] Among those studies the work of Ref. 28, for the first time, included the layer structure of the adsorbed liquid. The major drawback of that work was that it has been based on an approximation introduced by Lekner[27] which assumes that the correlations between the impurity and the background atoms are the same as the correlations between the individual background atoms. This rules out the applicability of the theory to impurities of a different species, for example hydrogen atoms.

The possibility[14] of studying the low-temperature properties of systems of hydrogen isotopes as atoms, rather than molecules, has made necessary the extension of the impurity theories to systems, where the interaction between the impurity and the background is considerably different from the interaction between two background atoms. A theoretical description of such systems has been attempted by Mantz and Edwards[29] and Guyer and Miller.[22] Mantz and Edwards predict for a hydrogen a binding energy of a few tenths of a degree, whereas the prediction of Guyer and Miller was less than 0.01 K and therefore of no practical consequence. Both theories suffer from uncertainty in the impurity-background correlations and the lack of a layer structure of the underlying film, which leads, as we will see, to interesting effects. Therefore, we have generalized the theory of Ref. 28 to the inclusion of arbitrary impurities.

For didactic reasons, we briefly review the ground-state theory of Ref. 28. In the variational approach used there, the ground-state wave function of an A-particle system is approximated by a variational form

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

The Hamiltonian of the A-particle system is

$$H=\sum_{i=1}^{A}\left[\frac{\hbar^2}{2m}\nabla_i^2+U_{\text{sub}}(\mathbf{r}_i)\right]+\sum_{\substack{i,j\\ 1\le i<j\le A}} V(|\mathbf{r}_i-\mathbf{r}_j|)\,, \quad (1.2)$$

where $U_{\text{sub}}(\mathbf{r}_i)$ is an external potential, which we will identify with the substrate potential which the particles feel, and $V(|\mathbf{r}_i-\mathbf{r}_j|)$ is the two-body interaction.

The best choice of the one- and two-body functions $u_1(\mathbf{r})$ and $u_2(\mathbf{r}_i,\mathbf{r}_j)$ is the one that minimizes the energy expectation value

$$E_A = \frac{\int d^3r_1 \ldots d^3r_A \Psi_0(\mathbf{r}_1, \ldots, \mathbf{r}_A) H \Psi_0(\mathbf{r}_1, \ldots, \mathbf{r}_A)}{\int d^3r_1 \ldots d^3r_A \Psi_0^2(\mathbf{r}_1, \ldots, \mathbf{r}_A)} , \tag{1.3}$$

i.e., the solutions of the variational problems

$$\frac{\delta E_A}{\delta u_1(\mathbf{r})} = 0, \quad \frac{\delta E_A}{\delta u_2(\mathbf{r}_i, \mathbf{r}_j)} = 0 . \tag{1.4}$$

An exact evaluation of the energy expectation value (1.3) for *given* functions $u_1(\mathbf{r})$ and $u_2(\mathbf{r}_i,\mathbf{r}_j)$ is possible with Monte Carlo techniques, but restricted to simple choices of correlation functions. Alternatively, one may sacrifice some of the calculational accuracy for the sake of a more flexible choice of the correlations and the possibility to study more delicate effects like, for example, the propagation of collective excitations or the response of the system to external forces. Of course, using an approximation method to evaluate the energy expectation value has the consequence that the variational problem (1.4) is not necessarily meaningful. It was one of the most important results in the formal studies of variational wave functions that the hierarchy of the so-called hypernetted-chain (HNC) approximations is the only procedure that allows a meaningful optimization of the correlations at any level of sophistication,[30] giving a qualitatively correct description of the short- and the long-ranged correlations.

It is important, for the problem of describing the structure and excitations of an adsorbed film, to have an adequate treatment of both the short- and the long-ranged correlations: The short-ranged correlations are needed to keep the particles apart from each other in the region of the repulsive core of the two-body interaction. They are also responsible for the formation of the layer structure of the film. Long-ranged correlations describe the low-lying excitations which are needed to treat the response of the system to the perturbation due to the impurity potential. Finally, we note that the optimized HNC theory is simple and flexible enough to be used without further approximations in quantum liquids with simple symmetry breaking. Thus, we are able to study anisotropies of the correlations in the surface.

There are some well-known disadvantages to the HNC method: In its simplest version, the quantitative agreement between the predictions of the HNC theory and the experimental results is only fair. Being an entirely microscopic theory, there is no way to honestly introduce phenomenological parameters to improve the agreement with experiments. The cure for this problem has been known for some time:[31,32] It is the systematic improvement of the HNC approximation by the inclusion of "elementary diagrams," and the generalization of the wave function to incorporate three-body correlations. The most honest calculations done for bulk ^{4}He lead to an agreement of the binding energy and the velocity of sound within better than 10% and an excellent reproduction of the pair correlation function.[33] The most important point, for our purposes, is that more sophisticated wave functions lead to a better quantitative agreement, but lead to no further qualitative insight into the physics of the system. Therefore we restrict ourselves to the HNC approximation and expect it to give us the correct overall physical picture, whereas we allow for some quantitative uncertainties.

In Sec. II, we develop the HNC theory for the impurity in an inhomogeneous medium. We derive a variational energy expression and the Euler equations for the impurity-background correlations and develop an efficient algorithm for their numerical solution. Section III is devoted to the derivation of some formal properties of the relevant interaction matrix elements in the long-wavelength limit, and the illumination of the relation of our theory to perturbative approaches. The theory is applied in Sec. IV to the case of impurities of ^{3}He and atomic hydrogen isotopes in the surface of an adsorbed film of ^{4}He. We find that the ^{3}He impurity can have three or four bound states depending on the thickness of the film.

The H isotopes do not have a bound state within the bulk helium liquid. However, their attraction to the substrate is about twice as strong as that of ^{4}He background atoms. For the films under consideration, which display up to five layers, we find that the tritium atom is always so strongly attracted to the substrate that it penetrates the liquid layer. The same happens for the D atom for two- and three-layer substrates, whereas the D atom has two bound surface states for films with more than three layers. The H atom always stays on the surface of the liquid and has one bound state whose binding energy decreases with the film thickness. We argue that the asymptotic value of the binding energy should be about 0.3 K.

In Secs. II–IV, we have assumed that the state of the impurity is translationally invariant on the surface. In Sec. V we study the question whether it could be energetically favorable for the impurity particle to form a localized (or *self-trapped* or *polaron*) state. From the structure of our equations we conclude that the translationally invariant state is always a local minimum of the energy, i.e., it is locally stable. In order to break the symmetry of that state, macroscopic changes of the impurity wave function must be considered. We investigated such macroscopic changes of the impurity wave function but did not find indications for a polaron state.

Section VI summarizes our results and discusses further applications. An Appendix gives details of the numerical optimization procedure for the bulk and surface impurity problem.

II. WAVE FUNCTIONS, ENERGIES, AND EULER EQUATIONS

The simplest variational wave function for an inhomogeneous N-particle Bose system with an impurity has the form

$$\Psi_0(\mathbf{r}_0, \mathbf{r}_1, \ldots, \mathbf{r}_A) = \exp\tfrac{1}{2}\left[u_1^I(\mathbf{r}_0) + \sum_{i=1}^{A} u_1(\mathbf{r}_i) + \sum_{i=1}^{A} u_2^I(\mathbf{r}_0, \mathbf{r}_i) + \sum_{1 \le i < j \le A} u_2(\mathbf{r}_i, \mathbf{r}_j) \right] . \tag{2.1}$$

The superscript I stands for the impurity, i.e., $u_1^I(\mathbf{r}_0)$ is the one-body part of the impurity wave function, and $u_2^I(\mathbf{r}_0,\mathbf{r}_i)$ is the correlation function between the impurity and a background atom. To simplify the notation we adopt the following convention: We will *always* label the impurity coordinate as $\mathbf{r}_0$ and omit, whenever it can be done unambiguously, the superscript I. Thus, we will alternatively write the wave function as

$$\Psi_0(\mathbf{r}_0,\mathbf{r}_1,\ldots,\mathbf{r}_A) = \exp\tfrac{1}{2}\left[\sum_{i=0}^{A} u_1(\mathbf{r}_i) + \sum_{0\le i<j\le A} u_2(\mathbf{r}_i,\mathbf{r}_j)\right] \qquad (2.2)$$

with the understanding that $u_1(\mathbf{r}_0)$ and $u_2(\mathbf{r}_0,\mathbf{r}_i)$ are *different* from $u_1(\mathbf{r}_i)$ and $u_2(\mathbf{r}_i,\mathbf{r}_j)$ for $i\neq 0$, $j\neq 0$.

The Hamiltonian of the $(A+1)$-particle system consisting of A background particles (labeled $1 \ldots A$) and one impurity particle (labeled 0) is

$$H = -\frac{\hbar^2}{2m_0}\nabla_0^2 + U_{\rm sub}^I(\mathbf{r}_0) + \sum_{i=1}^{A}\left[-\frac{\hbar^2}{2m}\nabla_i^2 + U_{\rm sub}(\mathbf{r}_i)\right] + \sum_{i=1}^{A} V^I(|\mathbf{r}_0-\mathbf{r}_i|) + \sum_{\substack{i,j\\ 1\le i<j\le A}} V(|\mathbf{r}_i-\mathbf{r}_j|) \ . \qquad (2.3)$$

Here, the $U_{\rm sub}(\mathbf{r}_i)$ is the external substrate potential which the particles feel, and $V(|\mathbf{r}_i-\mathbf{r}_j|)$ is the interaction between two individual particles. Note that we will again drop the explicit reference to the impurity particle (otherwise indicated by the superscript I) whenever the meaning is unambiguous. Thus, the $U_{\rm sub}(\mathbf{r}_0)$ and $V(|\mathbf{r}_0-\mathbf{r}_j|)$ are *different* from the $U_{\rm sub}(\mathbf{r}_i)$ and $V(|\mathbf{r}_i-\mathbf{r}_j|)$ for $i\neq 0$.

The energy that is necessary (or that is gained) by adding one impurity atom to a system of A background atoms is

$$\Delta E \equiv E_{I,A} - E_A \ , \qquad (2.4)$$

where $E_{I,A}$ is the energy of the system containing one impurity and A background atoms, and E_A is the energy (1.3) of the unperturbed background system. In our approximation, $E_{I,A}$ is the energy expectation value of the Hamiltonian (2.3) with respect to the wave function (2.1). We deviate in our notation from the one of earlier works for the bulk system,[26] which considered the energy needed to *replace* a background particle by the impurity. Unlike in the bulk system, the energy difference (2.4) is the relevant physical quantity in our case of a liquid film adsorbed to a substrate. For the formal developments, that are necessary to extend the impurity theories for bulk systems to the inhomogeneous case, it also turns out that it is almost trivial to calculate (2.4) using the results from the density matrix theory of Ref. 28.

The total energy $E_{I,A}$ of the system with the impurity can be calculated just as it is done in the background system with, the usual Jackson-Feenberg (JF) integration by parts. We define a generalized wave function $\Psi_0(\mathbf{r}_0,\ldots,\mathbf{r}_A;\beta)$ by the following replacements in Eq. (2.2): (a) each single particle function $u_1(\mathbf{r}_i)$ replaced by

$$u_1(\mathbf{r}_i;\beta) = u_1(\mathbf{r}_i) + \beta\left[U_{\rm sub}(\mathbf{r}_i) - \frac{\hbar^2}{8m_i}\nabla^2 u_1(\mathbf{r}_i)\right] , \qquad (2.5)$$

for $i=0,\ldots A$ (note, again, that the mass and the substrate potential are different for $i=0$) and (b) each two-body function $u_2(\mathbf{r}_i,\mathbf{r}_j)$ replaced by

$$\begin{aligned} u_2(\mathbf{r}_i,\mathbf{r}_j;\beta) &= u_2(\mathbf{r}_i,\mathbf{r}_j) + \beta\left[V(|\mathbf{r}_i-\mathbf{r}_j|) - \left[\frac{\hbar^2}{8m_i}\nabla_i^2 + \frac{\hbar^2}{8m_j}\nabla_j^2\right] u_2(\mathbf{r}_i,\mathbf{r}_j)\right] \\ &\equiv u_2(\mathbf{r}_i,\mathbf{r}_j) + \beta \bar{V}_{\rm JF}(\mathbf{r}_i,\mathbf{r}_j) \ . \end{aligned} \qquad (2.6)$$

We now define the generalized wave function as

$$\Psi_0(\mathbf{r}_0,\mathbf{r}_1,\ldots,\mathbf{r}_A;\beta) = \exp\tfrac{1}{2}[U^*(\mathbf{r}_0,\ldots,\mathbf{r}_A) + \beta V^*(\mathbf{r}_0,\ldots,\mathbf{r}_A)] \ , \qquad (2.7)$$

with

$$U^*(\mathbf{r}_0,\mathbf{r}_1,\ldots,\mathbf{r}_A) = \sum_{i=0}^{A} u_1(\mathbf{r}_i) + \sum_{\substack{i,j\\ 0\le i<j\le A}} u_2(\mathbf{r}_i,\mathbf{r}_j) \ , \qquad (2.8)$$

and

$$V^*(\mathbf{r}_0,\mathbf{r}_1,\ldots,\mathbf{r}_A) = \sum_{i=0}^{A}\left[U_{\rm sub}(\mathbf{r}_i) - \frac{\hbar^2}{8m_i}\nabla^2 u_1(\mathbf{r}_i)\right] + \sum_{0\le i<j\le A} \bar{V}_{\rm JF}(\mathbf{r}_i,\mathbf{r}_j) \ . \qquad (2.9)$$

The energy, the one- and two-body densities, and the Euler-Lagrange (EL) equations (1.4) are now conveniently formulated in terms of the generalized wave function and its derivative with respect to β. We define generalized background and impurity one-body densities

$$\rho_1(\mathbf{r}_0;\beta) = \frac{\int d^3r_1 \ldots d^3r_A \Psi_0^2(\mathbf{r}_0,\mathbf{r}_1,\ldots,\mathbf{r}_A;\beta)}{\int d^3r_0 \ldots d^3r_A \Psi_0^2(\mathbf{r}_0,\ldots,\mathbf{r}_A;\beta)} \ , \qquad (2.10)$$

$$\rho_1(\mathbf{r}_1;\beta)=A\frac{\int d^3r_2\dots d^3r_A\Psi_0^2(r_1,\dots,\mathbf{r}_A;\beta)}{\int d^3r_1\dots d^3r_A\Psi_0^2(\mathbf{r}_1,\dots,\mathbf{r}_A;\beta)}\ , \tag{2.11}$$

$$\rho_2(\mathbf{r}_0,\mathbf{r}_1;\beta)=A\frac{\int d^3r_2\dots d^3r_A\Psi_0^2(\mathbf{r}_0,\mathbf{r}_1,\dots,\mathbf{r}_A;\beta)}{\int d^3r_0\dots d^3r_A\Psi_0^2(\mathbf{r}_0,\dots,\mathbf{r}_A;\beta)}\ , \tag{2.12}$$

$$\rho_2(\mathbf{r}_1,\mathbf{r}_2;\beta)=A(A-1)\frac{\int d^3r_3\dots d^3r_A\Psi_0^2(\mathbf{r}_1,\dots,\mathbf{r}_A;\beta)}{\int d^3r_1\dots d^3r_A\Psi_0^2(\mathbf{r}_1,\dots,\mathbf{r}_A;\beta)}\ , \tag{2.13}$$

$$\rho_2(\mathbf{r}_i,\mathbf{r}_j;\beta)\equiv\rho_1(\mathbf{r}_i;\beta)g_2(\mathbf{r}_i,\mathbf{r}_j;\beta)\rho_1(\mathbf{r}_j;\beta)\ . \tag{2.14}$$

For the background quantities, we can relate the β-derivatives of the generalized densities with the energy and the Euler-Lagrange equations (1.4)

$$E_A=\frac{\partial}{\partial\beta}\ln\left[\int d^3r_1\dots d^3r_A\Psi_0^2(\mathbf{r}_1,\dots,\mathbf{r}_A;\beta)\right]\Bigg|_{\beta=0}\ , \tag{2.15}$$

$$\begin{aligned}\frac{\delta E_A}{\delta u_1(\mathbf{r})}&=\frac{\partial}{\partial\beta}\rho_1(\mathbf{r}_1;\beta)\Bigg|_{\beta=0}-\frac{\hbar^2}{8m}\nabla^2\rho_1(\mathbf{r})\\ &\equiv\rho_1'(\mathbf{r})-\frac{\hbar^2}{8m}\nabla^2\rho_1(\mathbf{r})=0\ ,\end{aligned} \tag{2.16}$$

$$\begin{aligned}&\frac{\delta E_A}{\delta u_2(\mathbf{r}_1,\mathbf{r}_2)}\\ &\quad=\frac{\partial}{\partial\beta}\rho_2(\mathbf{r}_1,\mathbf{r}_2,\beta)\Bigg|_{\beta=0}-\frac{\hbar^2}{8m}(\nabla_1^2+\nabla_2^2)\rho_2(\mathbf{r}_1,\mathbf{r}_2)\\ &\quad\equiv\rho_2'(\mathbf{r}_1,\mathbf{r}_2)-\frac{\hbar^2}{8m}(\nabla_1^2+\nabla_2^2)\rho_2(\mathbf{r}_1,\mathbf{r}_2)=0\ ,\end{aligned} \tag{2.17}$$

and

$$\begin{aligned}\rho_2'(\mathbf{r}_i,\mathbf{r}_j)=&[\rho_1'(\mathbf{r}_i)\rho_1(\mathbf{r}_j)+\rho_1(\mathbf{r}_i)\rho_1'(\mathbf{r}_j)]g(\mathbf{r}_i,\mathbf{r}_j)\\ &+\rho_1(\mathbf{r}_i)\rho_1(\mathbf{r}_j)g_2'(\mathbf{r}_i,\mathbf{r}_j)\ .\end{aligned} \tag{2.18}$$

For the calculation of the impurity energy, we define

$$\exp[\pi(\mathbf{r}_0;\beta)]=\frac{\int d^3r_1\dots d^3r_A\Psi_0^2(\mathbf{r}_0,\mathbf{r}_1,\dots,\mathbf{r}_A;\beta)}{\int d^3r_1\dots d^3r_A\Psi_0^2(\mathbf{r}_1,\dots,\mathbf{r}_A;\beta)} \tag{2.19}$$

and find for the impurity density

$$\rho_1(\mathbf{r}_0)=\frac{\exp[\pi(\mathbf{r}_0;\beta)]}{\int d^3r_0\exp[\pi(\mathbf{r}_0;\beta)]} \tag{2.20}$$

and the binding energy

$$\Delta E\equiv E_{I,A}-E_A=\int d^3r_0\rho(\mathbf{r}_0)\frac{\partial}{\partial\beta}\pi(\mathbf{r}_0;\beta)\Bigg|_{\beta=0}\ . \tag{2.21}$$

The advantage of this procedure is that one can use the density-matrix results (5.18) and (5.19a) of Ref. 28 with a slight modification for the calculation of $\pi(\mathbf{r}_0;\beta)$. The detour through the mixture formalism used in Ref. 26 can therefore be avoided.

The derivations of this section have been exact so far. In order to derive computationally manageable expressions, we must now introduce the HNC procedure for inhomogeneous systems. This leads to a systematic sequence of approximate relations between the one- and two-body components of the wave function, the densities, and distribution functions. We restrict ourselves here to the simplest approximation, known as the HNC *approximation.* For any two-point quantity [i.e, any function of two variables $F(\mathbf{r}_i,\mathbf{r}_j)$], it is convenient to define

$$\tilde{F}(\mathbf{r}_i,\mathbf{r}_j)=\sqrt{\rho_1(\mathbf{r}_i)}F(\mathbf{r}_i,\mathbf{r}_j)\sqrt{\rho_1(\mathbf{r}_j)} \tag{2.22}$$

and the convolution product of two functions $A(\mathbf{r}_i,\mathbf{r}_j)$ and $B(\mathbf{r}_i,\mathbf{r}_j)$

$$[\tilde{A}*\tilde{B}](\mathbf{r}_i,\mathbf{r}_j)=\int d^3r_k\,\tilde{A}(\mathbf{r}_i,\mathbf{r}_k)\tilde{B}(\mathbf{r}_k,\mathbf{r}_j)\ . \tag{2.23}$$

The HNC approximations relating the two-body correlations $u_2(\mathbf{r}_i,\mathbf{r}_j)$ to the pair-distribution functions $g(\mathbf{r}_i,\mathbf{r}_j)$ are, for $i=0,1$

$$\begin{aligned}g(\mathbf{r}_i,\mathbf{r}_2)&=\exp[u_2(r_i,\mathbf{r}_2)+N(\mathbf{r}_i,\mathbf{r}_2)]\ ,\\ X(\mathbf{r}_i,\mathbf{r}_2)&=g(\mathbf{r}_i,\mathbf{r}_2)-1-N(\mathbf{r}_i,\mathbf{r}_2)\ ,\\ h(\mathbf{r}_i,\mathbf{r}_2)&=g(\mathbf{r}_i,\mathbf{r}_2)-1\ ,\\ \tilde{N}(\mathbf{r}_i,\mathbf{r}_2)&=[\tilde{X}*\tilde{h}](\mathbf{r}_i,\mathbf{r}_2)\ .\end{aligned} \tag{2.24}$$

Note that the two-body functions $g(\mathbf{r}_0,\mathbf{r}_1)$ etc. involving the impurity atom are *not* symmetric in the coordinates. To determine the one-body densities in terms of the one-body functions $u_1(\mathbf{r}_i)$ and the two-body correlations, we also need the Born-Green-Yvon (BGY) equation

$$\begin{aligned}\nabla\rho_1(\mathbf{r}_i)=&\rho_1(\mathbf{r}_i)\nabla u_1(\mathbf{r}_i)\\ &+\rho_1(\mathbf{r}_i)\int d^3r_2 g(\mathbf{r}_i,\mathbf{r}_2)\nabla_i u_2(\mathbf{r}_i,\mathbf{r}_2)\ .\end{aligned} \tag{2.25}$$

[In Eqs. (2.24), we also allow for an implicit β dependence of all functions.] We can now use the density matrix results of Ref. 28 to calculate $\pi(\mathbf{r}_0;\beta)$ [Eq. (2.19)] in terms of the diagrammatic quantities introduced in the HNC theory. We must only replace the $u_2(\mathbf{r}_1,\mathbf{r}_2)/2$ in (5.18a) of Ref. 28 by $u_2(\mathbf{r}_0,\mathbf{r}_1)$ and imply a dependence of the correlations on the parameter β. Using the results of Ref. 28, we can write[34]

$$\pi(\mathbf{r}_0;\beta)=u_1(\mathbf{r}_0)+\int d^3r_1[X(\mathbf{r}_0,\mathbf{r}_1)-\tfrac{1}{2}h(\mathbf{r}_0,\mathbf{r}_1)N(\mathbf{r}_0,\mathbf{r}_1)]\rho_1(r_1)\ , \tag{2.26}$$

where $X(\mathbf{r}_0,\mathbf{r}_1)$, $N(\mathbf{r}_0,\mathbf{r}_1)$ are calculated from Eqs. (2.24). The calculation of ΔE [Eq. (2.21)] is now an algebraic exercise. The calculations are considerably simplified by the assumption that the background correlations have been optimized, i.e., we can use Eqs. (2.16) and (2.17). After a number of straightforward manipulations, including the elimination of the one- and two-body functions $u_1(\mathbf{r}_0)$ and $u_2(\mathbf{r}_0,\mathbf{r}_i)$, we can find a variety of equivalent, but algebraically different formulations, among which we found the following form most convenient:

$$\begin{aligned}\Delta E=&\int d^3r_0\left[\frac{\hbar^2}{2m_0}\,|\nabla\sqrt{\rho_1(\mathbf{r}_0)}\,|^2+\rho_1(\mathbf{r}_0)U_{\text{sub}}(\mathbf{r}_0)\right]+\int d^3r_0d^3r_1\rho_1(\mathbf{r}_0)\rho_1(\mathbf{r}_1)g(\mathbf{r}_0,\mathbf{r}_1)V(\,|\mathbf{r}_0-\mathbf{r}_1|\,)\\&+\int d^3r_0d^3r_1\rho_1(\mathbf{r}_0)\rho_1(\mathbf{r}_1)\left[\frac{\hbar^2}{2m_0}\,|\nabla_0\sqrt{g(\mathbf{r}_0,\mathbf{r}_1)}\,|^2+\frac{\hbar^2}{2m}\,|\nabla_1\sqrt{g(\mathbf{r}_0,\mathbf{r}_1)}\,|^2\right]\\&+\tfrac{1}{2}\int d^3r_0d^3r_1\rho_1(\mathbf{r}_0)\rho_1(\mathbf{r}_1)h(\mathbf{r}_0,\mathbf{r}_1)w_I(\mathbf{r}_0,\mathbf{r}_1)\ ,\end{aligned} \tag{2.27}$$

where we have defined in view of later use

$$\tilde{w}_I(\mathbf{r}_0,\mathbf{r}_1)=\tfrac{1}{2}[H_1(0)+H_1(1)]\tilde{N}(\mathbf{r}_0,\mathbf{r}_1)+\tfrac{1}{2}[\tilde{X}*H_1\tilde{X}](\mathbf{r}_0,\mathbf{r}_1) \tag{2.28}$$

and the one-body Hamiltonian

$$H_1(i)\equiv-\frac{\hbar^2}{2m_i}\frac{1}{\sqrt{\rho_1(\mathbf{r}_i)}}\nabla\rho_1(\mathbf{r}_i)\cdot\nabla\frac{1}{\sqrt{\rho_1(\mathbf{r}_i)}}\ . \tag{2.29}$$

The density of the impurity is calculated by minimizing the energy (2.27) with respect to $\sqrt{\rho_1(\mathbf{r}_0)}$. We get the usual Hartree equation

$$-\frac{\hbar^2}{2m_0}\nabla^2\sqrt{\rho_1(\mathbf{r}_0)}+[U_{\text{sub}}(\mathbf{r}_0)+V_{\text{SC}}(\mathbf{r}_0)]\sqrt{\rho_1(\mathbf{r}_0)}=\mu\sqrt{\rho_1(\mathbf{r}_0)}\ , \tag{2.30}$$

with

$$\begin{aligned}V_{\text{SC}}(\mathbf{r}_0)=&\int d^3r_1\rho_1(\mathbf{r}_1)g(\mathbf{r}_0,\mathbf{r}_1)V(\,|\mathbf{r}_0-\mathbf{r}_1|\,)+\int d^3r_1\rho_1(\mathbf{r}_1)\left[\frac{\hbar^2}{2m_0}\,|\nabla_0\sqrt{g(\mathbf{r}_0,\mathbf{r}_1)}\,|^2+\frac{\hbar^2}{2m}\,|\nabla_1\sqrt{g(\mathbf{r}_0,\mathbf{r}_1)}\,|^2\right]\\&+\tfrac{1}{2}\int d^3r_1h(\mathbf{r}_0,\mathbf{r}_1)\rho_1(\mathbf{r}_1)w_I(\mathbf{r}_0,\mathbf{r}_1)-\frac{\hbar^2}{16m_0\rho_1(\mathbf{r}_0)}\nabla_0\rho_1(\mathbf{r}_0)\cdot\nabla_0\int d^3r_1d^3r_2h(\mathbf{r}_0,\mathbf{r}_1)h(\mathbf{r}_0,\mathbf{r}_2)\rho_1(\mathbf{r}_1)X(\mathbf{r}_1,\mathbf{r}_2)\rho_1(\mathbf{r}_2)\ .\end{aligned} \tag{2.31}$$

The two-body Euler equation is finally derived by variation with respect to $\sqrt{g(\mathbf{r}_0,\mathbf{r}_1)}$. After a number of algebraic manipulations that are virtually identical to those used in the derivation of the Euler equations for the background,[28] one arrives at

$$[H_1(0)+H_1(1)]\tilde{X}(\mathbf{r}_0,\mathbf{r}_1)-[\tilde{X}*H_1\tilde{X}](\mathbf{r}_0,\mathbf{r}_1)=-2\tilde{V}_{p\text{-}h}(\mathbf{r}_0,\mathbf{r}_1)\ . \tag{2.32}$$

The "particle-hole interaction" is, just as in the background system

$$V_{p\text{-}h}(\mathbf{r}_0,\mathbf{r}_1)=g(\mathbf{r}_0,\mathbf{r}_1)V(\,|\mathbf{r}_0-\mathbf{r}_1|\,)+\frac{\hbar^2}{2m_0}\,|\nabla_0\sqrt{g(\mathbf{r}_0,\mathbf{r}_1)}\,|^2+\frac{\hbar^2}{2m}\,|\nabla_1\sqrt{g(\mathbf{r}_0,\mathbf{r}_1)}\,|^2+h(\mathbf{r}_0,\mathbf{r}_1)w_I(\mathbf{r}_0,\mathbf{r}_1)\ . \tag{2.33}$$

Equations (2.30)–(2.33) form a complete set of equations determining the two-body correlations and the impurity density from the structure of the background and the one- and two-body potentials. The major numerical effort of the calculation is the solution of the two-body equation (2.32), which is complicated by the fact that the impurity distribution functions are not symmetric. A convenient way to solve the two-body equation (2.32) is to first solve the eigenvalue problems

$$H_1(\mathbf{r}_0)\psi_i(\mathbf{r}_0)=t_i\psi_i(\mathbf{r}_0) \tag{2.34}$$

and

$$\int d^3r_2H_1(\mathbf{r}_1)[\delta(\mathbf{r}_1-\mathbf{r}_2)-\tilde{X}(\mathbf{r}_1,\mathbf{r}_2)]\phi_i(\mathbf{r}_2)=\hbar\omega_i\phi_i(\mathbf{r}_1)\ . \tag{2.35}$$

Details on the procedure are described in the Appendix. It turns out that the solutions of the second eigenvalue problem (2.35) are already known from the density fluctuations and the collective excitations of the background system. We find (c.f., Appendix)

$$\phi_i(\mathbf{r}_1)=(\hbar\omega_i)^{-1/2}\delta[\rho_1^{(i)}(\mathbf{r}_1)]^{1/2}\ , \tag{2.36}$$

where the $\hbar\omega_i$ are the collective excitation energies of the background, and $\delta[\rho_1^{(i)}(\mathbf{r})]^{1/2}$ the spatial shape of the corresponding density fluctuations. We then obtain the static structure function of the impurity

$$\begin{aligned}\tilde{h}(\mathbf{r}_0,\mathbf{r}_1)&\equiv S(\mathbf{r}_0,\mathbf{r}_1)\\&=\int d^3r_2\tilde{X}(\mathbf{r}_0,\mathbf{r}_2)S(\mathbf{r}_2,\mathbf{r}_1)\\&=-2\sum_{i,j}\psi_i(\mathbf{r}_0)\frac{(\psi_i\,|\,V_{p\text{-}h}\,|\,\phi_j)}{t_i+\hbar\omega_j}\phi_j(\mathbf{r}_1)\ ,\end{aligned} \tag{2.37}$$

which is the generalization of Eq. (A3) for the inhomogeneous case. The other manipulations for the iterative procedure to obtain the optimal pair correlations are identical to the ones in the bulk system and need not be repeated here. Most of the work requiring extensive numerical calculations has already been done in the calculation of background system. The only new aspect is the diagonalization of the one-body Hamiltonian $H_1(0)$.

III. INTERACTION MATRIX ELEMENTS, STRUCTURE FUNCTION, AND UNIFORM LIMIT

Before we discuss the numerical application of our theory to ^{3}He and hydrogen atoms in the surface of a liquid ^{4}He film, we first investigate the coupling of the impurity to the low-lying excitations of the background. Information on this coupling is needed for the qualitative study of polaron formation, the calculation of the impurity effective mass, and the impurity-impurity interaction. As the lowest-lying collective mode "softens" with increasing film thickness, impurity properties that are determined by these soft modes can be a sensitive probe for the surface structure of the underlying liquid.[6,7] The physical interpretation of our theory is illuminated by comparison to conventional theories applicable in the weakly interacting limit.

The key elements of the theory are the matrix elements

$$(\psi_i\,|\,\tilde{X}^I\,|\,\phi_j)=-2\frac{(\psi_i\,|\,\tilde{V}^I_{p\text{-}h}\,|\,\phi_j)}{t_i+\hbar\omega_j} \tag{3.1}$$

between the excitations of the background system and the impurity atom. To study the singular behavior caused by the energy denominator in the long-wavelength limit it is sufficient to restrict the discussion to the lowest-lying states. From Eq. (2.34) we see that

$$\psi_0(\mathbf{r}_0)=\sqrt{\rho_1(z_0)}\frac{e^{i\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}}{L}\ , \tag{3.2}$$

where we have normalized the wave functions to an area L^2 parallel to the surface. The lowest-lying excitations of the background must be worked out separately for the adsorbed film and in the limit of an infinite half-space.

The lowest-lying collective excitation of an adsorbed film of finite thickness is the third-sound mode $\omega_0(q_\parallel)=c_3q_\parallel$, where c_3 is the third-sound velocity, and $q_\parallel$ the momentum parallel to the surface. The third-sound velocity c_3 is given in first approximation by the Van der Waals acceleration of the liquid in the substrate potential. The shape of the density fluctuation is determined by the solutions of the eigenvalue problem (A8); observing the normalization (A9), the lowest-energy eigenfunction of Eq. (2.35) is in the long-wavelength limit given by[35]

$$\begin{aligned}\phi_0(\mathbf{r})&=[\hbar\omega_0(q_\parallel)]^{-1/2}H_1\Psi^{(0)}(\mathbf{r})\\&=\left[\frac{\hbar^2q_\parallel^2}{2m\hbar\omega_0(q_\parallel)}\right]^{1/2}\frac{[H_1(0+)+2\tilde{V}_{p\text{-}h}(0+)]^{-1}\,|\sqrt{\rho_1})}{(\sqrt{\rho_1}\,|\,[H_1(0+)+2\tilde{V}_{p\text{-}h}(0+)^{-1}\,|\sqrt{\rho_1})}\frac{e^{i\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}}{L}\\&=\left[\frac{\hbar^2q_\parallel^2}{2m\hbar\omega_0(q_\parallel)}\right]^{1/2}\frac{\delta\rho_1(z)}{\sqrt{\rho_1(z)}}\frac{e^{i\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}}{L}\ .\end{aligned} \tag{3.3}$$

$\tilde{V}_{p\text{-}h}(0+)$ is the long-wavelength limit of the particle-hole interaction of the background

$$\tilde{V}_{p\text{-}h}(z_1,z_2,0+)\equiv\int d^2r_\parallel\sqrt{\rho_1(z_1)}V_{p\text{-}h}(z_1,z_2,r_\parallel)\sqrt{\rho_1(z_2)}\ , \tag{3.4}$$

and

$$H_1(0+)=-\frac{\hbar^2}{2m}\frac{1}{\sqrt{\rho_1(z_1)}}\frac{d}{dz_1}\rho_1(z_1)\frac{d}{dz_1}\frac{1}{\sqrt{\rho_1(z_1)}}\ . \tag{3.5}$$

We have not spelled out the z dependence of the operators in the second line of Eq. (3.3). The last line of Eq. (3.3) defines the $\delta\rho_1(z)$, which is the shape of the density fluctuation normalized such that $\int dz\,\delta\rho_1(z)=1$. Inserting the form (3.3) of the lowest-lying mode in (3.1) and keeping only the terms that dominate the long-wavelength behavior, we obtain

$$\begin{aligned}&(\psi_0(q_\parallel)\,|\,\tilde{X}\,|\,\phi_0(q_\parallel))\\&\quad=-\left[\frac{2}{\hbar m_0c_3^3q_\parallel}\right]^{1/2}(\rho_1^I\,|\,V^I_{p\text{-}h}(0+)\,|\,\delta\rho_1),\quad(q_\parallel\rightarrow0)\ ,\end{aligned} \tag{3.6}$$

where we have applied the definition (3.4) for $V^I_{p\text{-}h}(0+)$.

Using (2.37) we can finally work out the static structure function in the long-wavelength limit. We find

$$\begin{aligned}&S(z_0,z_1,q_\parallel)\\&\quad=-\sqrt{\rho_1(z_0)}\frac{(\rho_1^I\,|\,V^I_{p\text{-}h}(0+)\,|\,\delta\rho_1)}{2mc_3^2}\delta\sqrt{\rho_1(z_1)}\ .\end{aligned} \tag{3.7}$$

The derivations are a bit more delicate in a semi-infinite medium. Here, the linear third-sound spectrum is replaced by the ripplon dispersion relation

$$\omega_r^2(q_\parallel)=\frac{\sigma}{m\rho_\infty}q_\parallel^3 , \tag{3.8}$$

where ρ_∞ is the asymptotic density of the film, and σ the surface energy. The corresponding eigenfunction of the normal mode (A8), (A9) is[36]

$$\begin{aligned}\phi_0(\mathbf{r})&=[\hbar\omega_r(q_\parallel)]^{-1/2}H_1\Psi_r^0(z,q_\parallel)\\ &=\left[\frac{2\hbar q_\parallel}{m\rho_\infty\omega_r(q_\parallel)}\right]^{1/2}\frac{d}{dz}\sqrt{\rho_1(z)}\frac{e^{i\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}}{L} .\end{aligned} \tag{3.9}$$

Using the shape (3.9) of the collective modes and the ripplon dispersion relation Eq. (3.6) we find

$$\begin{aligned}&(\psi_0\,|\,\tilde{X}(\mathbf{r}_0,\mathbf{r}_1)\,|\,\phi_0)\\ &\quad=\left[\frac{\hbar q_\parallel}{2m\rho_\infty\omega_r(q_\parallel)}\right]^{1/2}\left[\rho_1^I(z_0)\,|\,X(q_\parallel)\,|\,\frac{d\rho_1(z_1)}{dz_1}\right] .\end{aligned} \tag{3.10}$$

In the case of a free surface, the density profile of the impurity particle is completely determined by the density profile of the background, i.e.,

$$\rho_1^I(z_0)=\rho_1^I[\rho_1(z)](z_0) . \tag{3.11}$$

In particular, the impurity density moves with the surface of the background if the background density profile is displaced rigidly by a distance ξ, i.e.,

$$\rho_1^I[\rho_1(z+\xi)](z_0)=\rho_1^I[\rho_1(z)](z_0+\xi) . \tag{3.12}$$

Expanding Eq. (3.12) to first order in the displacement ξ, we find the relation

$$\begin{aligned}\frac{d\rho_1^I(z_0)}{dz_0}&=\int dz_1\frac{\delta\rho_1^I(z_0)}{\delta\rho_1(z_1)}\frac{d\rho_1(z_1)}{dz_1}\\ &=\rho_1^I(z_0)\int d^3r_1X(\mathbf{r}_0,\mathbf{r}_1)\frac{d\rho_1(z_1)}{dz_1} ,\end{aligned} \tag{3.13}$$

by the definition of the "direct correlation function" $X(\mathbf{r}_0,\mathbf{r}_1)$. From the relation (3.13) we see that the matrix element of X appearing in Eq. (3.10) vanishes for $q_\parallel\to0$:

$$\left[\rho_1^I(z_0)\,|\,X^I(0+)\,|\,\frac{d\rho_1(z_1)}{dz_1}\right]=\int dz_0\frac{d\rho_1^I(z_0)}{dz_0}=0 , \tag{3.14}$$

if the impurity does not penetrate into the medium.

Now, using the Euler equation for long wavelength, we find, for $q_\parallel\to0$

$$\begin{aligned}&\left[\rho_1^I(z_0)\,|\,V_{p\text{-}h}^I(q_\parallel)\,|\,\frac{d\rho_1(z_1)}{dz_1}\right]\\ &\quad=\hbar\omega_r(q_\parallel)\left[\rho_1^I(z_0)\,|\,X(0+)\,|\,\frac{d\rho_1(z_1)}{dz_1}\right]\to0 .\end{aligned} \tag{3.15}$$

Since $V_{p\text{-}h}^I(\mathbf{r}_0,\mathbf{r}_1)$ is a short-ranged function, its Fourier transform is analytic at the origin. Therefore, $V_{p\text{-}h}^I(z_0,z_1,q_\parallel)$ has the expansion

$$V_{p\text{-}h}^I(z_0,z_1,q_\parallel)=V_{p\text{-}h}^I(z_0,z_1,0+)+q_\parallel^2V_1^I(z_0,z_1) , \tag{3.16}$$

with [c.f., Eq. (3.15)]

$$\left[\rho_1^I(z_0)\,|\,V_{p\text{-}h}^I(0+)\,|\,\frac{d\rho_1(z_1)}{dz_1}\right]=0 \tag{3.17}$$

in an infinite half-space. We can finally calculate the static structure function for $q_\parallel\to0$, using (3.16), (3.17), and (2.37):

$$\begin{aligned}S(z_0,z_1,q_\parallel)=&-\frac{2}{\sigma q_\parallel}\left[\rho_1(z_0)\,|\,V_1^I\,|\,\frac{d\rho_1(z_1)}{dz_1}\right]\\ &\times\sqrt{\rho_1(z_0)}\frac{d\sqrt{\rho_1(z_1)}}{dz_1} .\end{aligned} \tag{3.18}$$

Our results on the structure of the interaction matrix elements will be used in Sec. V for a qualitative study of the formation of "self-trapped" or "polaron" state.

To conclude this section, we investigate the relationship between the present variational theory and alternative approaches based on perturbation theory and effective interactions. A direct application of these simplifications is the interaction between electrons and liquid helium surfaces[1,37–40] where the assumption of a weak interaction is indeed justified.

Our formal derivation of the weakly interacting limit closely follows the derivation of the "uniform limit"[30] in the bulk system in the sense that we keep all impurity-background correlations in *coordinate space* to second order, but treat all convolution products between correlation functions exactly. This approximation corresponds to assuming *weak, but long ranged* correlations.

To be specific, we identify in this approximation the particle-hole interaction the bare impurity-background interaction

$$V_{p\text{-}h}(\mathbf{r}_0,\mathbf{r}_1)\approx V(\,|\mathbf{r}_0-\mathbf{r}_1|\,) , \tag{3.19}$$

and expand in the second line of Eq. (2.27)

$$|\nabla_i\sqrt{g(\mathbf{r}_0,\mathbf{r}_1)}\,|^2\approx\tfrac{1}{4}\,|\nabla_ig(\mathbf{r}_0,\mathbf{r}_1)|^2 . \tag{3.20}$$

Note that no approximations are made for the background quantities. Using (3.19) in the Euler equation (2.32) and (3.20) in the energy expression (2.27), we find

$$\begin{aligned}\Delta E=&\int d^3r_0\left[\frac{\hbar^2}{2m_0}\,|\nabla\sqrt{\rho_1(\mathbf{r}_0)}\,|^2+\rho_1(\mathbf{r}_0)U_{\text{sub}}(\mathbf{r}_0)\right]+\int d^3r_0d^3r_1\rho_1(\mathbf{r}_0)\rho_1(\mathbf{r}_1)V(\,|\mathbf{r}_0-\mathbf{r}_1|\,)\\ &+\tfrac{1}{2}\int d^3r_0d^3r_1\rho_1(\mathbf{r}_0)\rho_1(\mathbf{r}_1)h(\mathbf{r}_0,\mathbf{r}_1)V(\,|\mathbf{r}_0-\mathbf{r}_1|\,) ,\end{aligned} \tag{3.21}$$

and using the formal solution of the Euler equation (2.37) we obtain

$$\Delta E=\int d^3r_0\left[\frac{\hbar^2}{2m_0}\,|\nabla\sqrt{\rho_1(\mathbf{r}_0)}\,|^2+\rho_1(\mathbf{r}_0)U_{\text{sub}}(\mathbf{r}_0)\right]+\int d^3r_0d^3r_1\rho_1(\mathbf{r}_0)\rho_1(\mathbf{r}_1)V(\,|\mathbf{r}_0-\mathbf{r}_1|\,)-\sum_{i,j}\frac{|(\psi_i\,|\,\tilde{V}\,|\,\phi_j)|^2}{t_i+\hbar\omega_j}\ . \tag{3.22}$$

The first two terms in Eq. (3.22) describe the kinetic energy of the impurity particle and its energy in the substrate potential, and the third term the interaction between the impurity atom and the background. The last term in Eq. (3.22) is the energy that is stored in the elastic deformation of the background due to the presence of the impurity, and the energy that is gained by the fact that it is energetically favorable for the impurity atom to get closer to the substrate.

A somewhat heuristic derivation of a very similar expression draws the connection with the work of Guyer *et al.*[19] If the impurity-background interaction is weak, we may assume that the total energy of the system consists of the following.

(a) The energy of the impurity in the field of the substrate

$$E_I=\int d^3r_0\left[\frac{\hbar^2}{2m}\,|\nabla\sqrt{\rho_1(\mathbf{r}_0)}\,|^2+\rho_1(\mathbf{r}_0)U_{\text{sub}}(\mathbf{r}_0)\right]\ . \tag{3.23}$$

(b) An impurity-background interaction

$$E_{I\text{-}B}=\int d^3r_0d^3r_1\rho_1(\mathbf{r}_0)\rho_1(\mathbf{r}_1)V(\,|\mathbf{r}_0-\mathbf{r}_1|\,)\ . \tag{3.24}$$

(c) The elastic energy of the background due to a deformation $\delta\rho_1(\mathbf{r})$ of the background density due to the presence of an impurity:

$$E_{\text{el}}=\tfrac{1}{2}\int d^3r_1d^3r_2\frac{\delta^2E_A}{\delta\rho_1(\mathbf{r}_1)\delta\rho_1(\mathbf{r}_2)}\delta\rho_1(\mathbf{r}_1)\delta\rho_1(\mathbf{r}_2)\ , \tag{3.25}$$

where E_A the energy of the background atoms. The integral kernel in Eq. (3.25) is obviously related to the inverse of the zero-energy density-density response function. In our formulation of the background theory, we can express the elastic energy as

$$E_{\text{el}}=\int d^3r_1d^3r_2\delta\sqrt{\rho_1(\mathbf{r}_1)}\left[H_1(\mathbf{r}_1)\delta(\mathbf{r}_1-\mathbf{r}_2)+2\tilde{V}_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)\right]\delta\sqrt{\rho_1(\mathbf{r}_1)}\ . \tag{3.26}$$

Note that the density $\rho_1(\mathbf{r}_1)$ in Eq. (3.24) is the perturbed background density, $\rho_1(\mathbf{r}_1)=\rho_1^{(0)}(\mathbf{r}_1)+\delta\rho_1(\mathbf{r}_1)$. We may now minimize the total energy with respect to the change of the background density. The elimination of $\delta\rho_1(\mathbf{r})$ can be carried out explicitly using the spectral decomposition derived from (A8) for the integral kernel in Eq. (3.24). Thus, we find a final expression for the impurity energy

$$\begin{aligned}\Delta E&=E_I+E_{I\text{-}B}+E_{\text{el}}\\&=\int d^3r_0\left[\frac{\hbar^2}{2m_0}\,|\nabla\sqrt{\rho_1(\mathbf{r}_0)}\,|^2+\rho_1(\mathbf{r}_0)U_{\text{sub}}(\mathbf{r}_0)\right]+\int d^3r_0d^3r_1\rho_1(\mathbf{r}_0)\rho_1(\mathbf{r}_1)V(\,|\mathbf{r}_0-\mathbf{r}_1|\,)-\sum_{j}\frac{|(\psi_0\,|\,\tilde{V}\,|\,\phi_j)|^2}{\hbar\omega_j}\ .\end{aligned} \tag{3.27}$$

i.e., the same expression as Eq. (3.22) if one assumes, for the calculation of the background response, an infinite-mass impurity.

To derive an expression identical to (3.22) from a weakly interacting theory, one has to calculate the impurity self-energy and take the zero-energy limit. The important point for our purposes is to demonstrate that the deformation of the background is contained in the variational theory, even though one has assumed that the correlations between the background atoms are the same as in the unperturbed system.

IV. GROUND-STATE STRUCTURE

The physical model underlying our calculations of the ^{4}He background is described in detail in Refs. 28 and 35. There we have considered a number of helium atoms interacting via the Aziz potential.[37] The atoms are adsorbed to a substrate which is described by an external field $U_{\text{sub}}(z)$. A simple form for $U_{\text{sub}}(z)$ is the potential obtained by averaging Lennard-Jones interactions between helium and substrate atoms over a half-space.[38,39] One obtains

$$U_{\text{sub}}(z)=e\left[\frac{1}{15}\left[\frac{s}{z}\right]^9-\left[\frac{s}{z}\right]^3\right]\ . \tag{4.1}$$

Given the substrate potential, the two-body interaction, and the surface coverage,

$$n=\int dz\rho_1(z)\ , \tag{4.2}$$

the physical model of the background is completely defined.

We have described, in Ref. 35, the difficulty in using a realistic potential close to the substrate. This difficulty arises from the formation of a *solid* layer close to the substrate, which causes the HNC-EL equations to develop an instability at finite wave number. Such a property of the theory is in principle desirable, but the simultaneous treatment of a liquid and a solid is presently beyond our computational capabilities. To avoid a divergence, we have weakened the short-ranged attraction somewhat, and adjusted the attractive tail of the potential to a value about midway between the experimental and the theoretical prediction of Ref. 39. Thus, the substrate potential for the background particles is determined by the parameters $e = 48$ K and $s = 3.6$ Å.

For the ^{3}He impurities, the same substrate potential and the same two-body interaction were taken. The helium-hydrogen interaction is somewhat less attractive and has a considerably larger repulsive core. Here we used a Lennard-Jones 6-12 potential

$$V_{\text{H-He}}(r) = 4\varepsilon\left[\left[\frac{\sigma}{r}\right]^{12} - \left[\frac{\sigma}{r}\right]^{6}\right], \tag{4.3}$$

with the parameters $\varepsilon = 6.57$ K and $\sigma = 3.19$ Å as determined by Toenis *et al.*[40] The hydrogen isotopes are, on the other hand, more strongly attracted to the substrate. Following Ref. 41, we took the substrate potential (4.1) with the strength $e = 90$ K. The long-ranged attraction agrees with the value given by Bruch.[41] The short-ranged portion of the potential is less well known.[42] Since we are mainly interested in states where the impurity atoms are on the surface of the helium film, the detailed form of this part of the potential should be immaterial.

The numerical calculation of the impurity states is a somewhat delicate task if the impurities are located in the low-density region of the helium film. It is rather difficult and expensive to extend the calculations of the background system into a sufficiently large low-density region of the ^{4}He liquid. In that regime, the pair correlation function between two ^{4}He atoms develops a large and long-ranged overshoot, and a very large mesh is needed to avoid numerical instabilities. On the other hand, our method of solving the Euler equations for the impurity requires a complete set of normal modes. In order to keep the computational effort reasonable, we have therefore calculated the normal modes of the background [i.e., the eigenfunctions of Eq. (A8)] on an enlarged mesh by extrapolating the integral kernel of Eq. (A8) to its known asymptotic form $\tilde{V}_{p\text{-}h}(z,z',q_{\parallel}) \rightarrow 0$ as $z,z' \rightarrow \infty$ and $H_1(z) \rightarrow -(\hbar^2/2m)(d^2/dz^2) - \mu$ as $z \rightarrow \infty$. We have then generated the normal modes, but have not iterated the background equations any further on the extended mesh.

The helium and the hydrogen impurities in the bulk behave qualitatively differently. A ^{3}He atom is *bound* in bulk ^{4}He, but is energetically favorable by about 2.2 K to stay at the surface. The hydrogen isotopes, on the other hand, will be expelled from the bulk. The potential barrier can be obtained by calculating the impurity energy in the bulk as described in the appendix. At the calculated equilibrium density of bulk ^{4}He the ^{3}He atom feels an attractive potential of -2.6 K in the bulk ^{4}He, whereas the T, D, and H atoms are repelled from the bulk by a potential barrier of 4.3, 11.5, and 30 K, respectively. Whether a *surface bound state* like the one of ^{3}He develops is a question of a delicate balance between the attraction to the surface and the kinetic energy of the particle. In our case, the situation is even more complicated due to the attraction to the substrate.

We have attempted calculations of the binding energy of ^{3}He, H, D, and T atoms to a family of ^{4}He films with a surface coverage of $n = 0.15$, 0.20, 0.25, and 0.30, Å^{-2}. In all cases considered it turned out that the tritium atom penetrates the film and forms a very tightly bound state at the substrate. This can be understood considering the relatively weak repulsive barrier seen by the T atom that tries to penetrate into the bulk, and the stronger attraction of the hydrogen isotopes to the substrate. The deuterium atom is also seen to penetrate to the substrate for the films with the smaller surface coverage $n = 0.15$ and 0.20 Å^{-2}. We believe that our present model of the structure of the adsorbed films and the substrate potentials close to the substrate is too crude to make more definite statements on the structure and the binding energies of these tightly bound states. We therefore, restrict our discussion to those cases where the impurity atom forms a bound state at the free surface of the film. One should expect that a tritium surface-bound state develops when the film becomes thicker due to the weakening of the substrate attraction.

The binding energies of the ^{3}He, H, and D impurity atoms are collected in Table I, the shape of the ground state of the various impurities on the different substrates is shown in Figs. 1(a)–1(d). In Table I we also give the energies of the bound excited states of the impurities; we will discuss these states further below. The binding energies for the ^{3}He impurity are in reasonable agreement with the calculation of Ref. 28, where we assumed that the two-body correlations between the ^{3}He impurity and the ^{4}He atoms are the same as the ones between two ^{4}He atoms (Lekner approximation[27]). The particles appear to be bound somewhat weaker in our improved calculation, we expect that this is to some extent due to the extrapolation used there for the one-body potential.

The most interesting H isotope is atomic hydrogen, a case which has been under dispute as to whether the atom forms a surface-bound state. We found such a bound state in all cases considered. Our binding energy of a few tenths of a K may indeed be an underestimate due to mesh-size restrictions. We also feel that we have extended our calculation to sufficiently large films such that one expects that there should also be a bound state in the limit of infinite film thickness. To substantiate our argument, we show in Figs. 2(a)–2(c) the one-body potentials acting on the particles for the different surface coverages. The attractive parts of the potential seen by the H atom [Fig. 2(b)] for the two thickest films appear to be essentially parallel shifted by an amount comparable to the increase of the film thickness. In particular, a sizable part of the attraction comes from correlations. Taking these arguments and error estimates for the binding ener-

TABLE I. Binding energies and excited states of ^{3}He, H, and D impurities for the four examples of ^{4}He films discussed in the text. The surface coverage n is given in Å^{-2} and all energies in K. The deuterium impurity penetrates to the substrate for the two thin films with $n = 0.15$ Å^{-2} and $n = 0.20$ Å^{-2}.

	^{3}He			H	D	
n	ε_0	ε_1	ε_2	ε_0	ε_0	ε_1
0.15	−5.82	−2.73	−1.91	−2.86		
0.20	−5.10	−3.01	−1.51	−0.89		
0.25	−4.40	−2.68	−1.30	−0.50	−2.75	−0.78
0.30	−3.99	−2.47	−1.26	−0.27	−2.42	−0.57

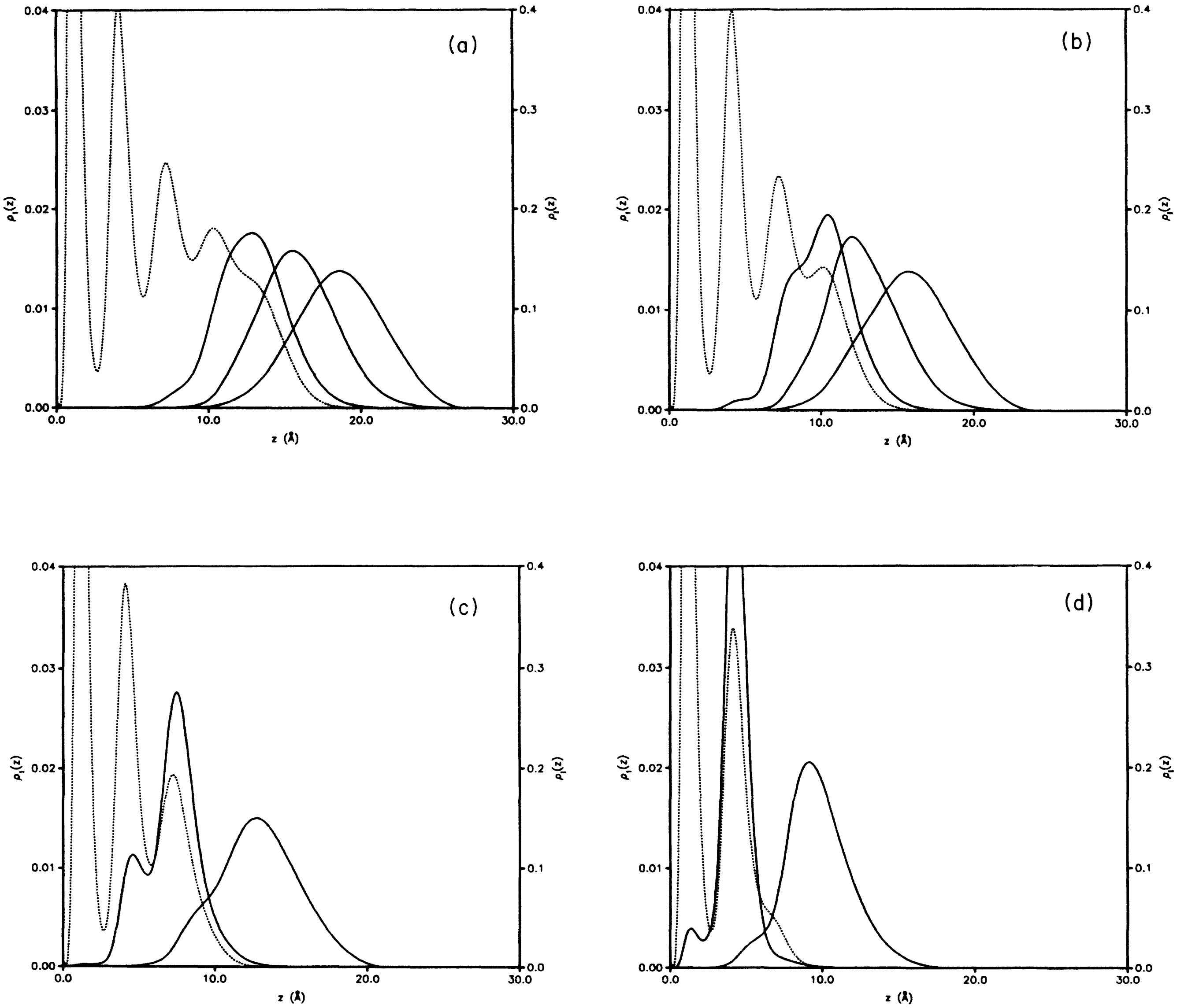

FIG. 1. (a) The densities for the ^{3}He, H, and D impurities are shown on the background of a ^{4}He film with a surface coverage of $n = 0.30$ Å^{-2} (dotted line). The ^{3}He impurity is closest to the surface, followed by D and H. (b) Same as Fig. 1(a) for the background of a ^{4}He film with a surface coverage of $n = 0.25$ Å^{-2} (dotted line). (c) Same as Fig. 1(a) for the background of a ^{4}He film with a surface coverage of $n = 0.20$ Å^{-2} (dotted line). The deuterium impurity has no surface-bound state and penetrates to the substrate. (d) Same as Fig. 1(a) for the background of a ^{4}He film with a surface coverage of $n = 0.15$ Å^{-2} (dotted line). The deuterium impurity has no surface-bound state and penetrates to the substrate.

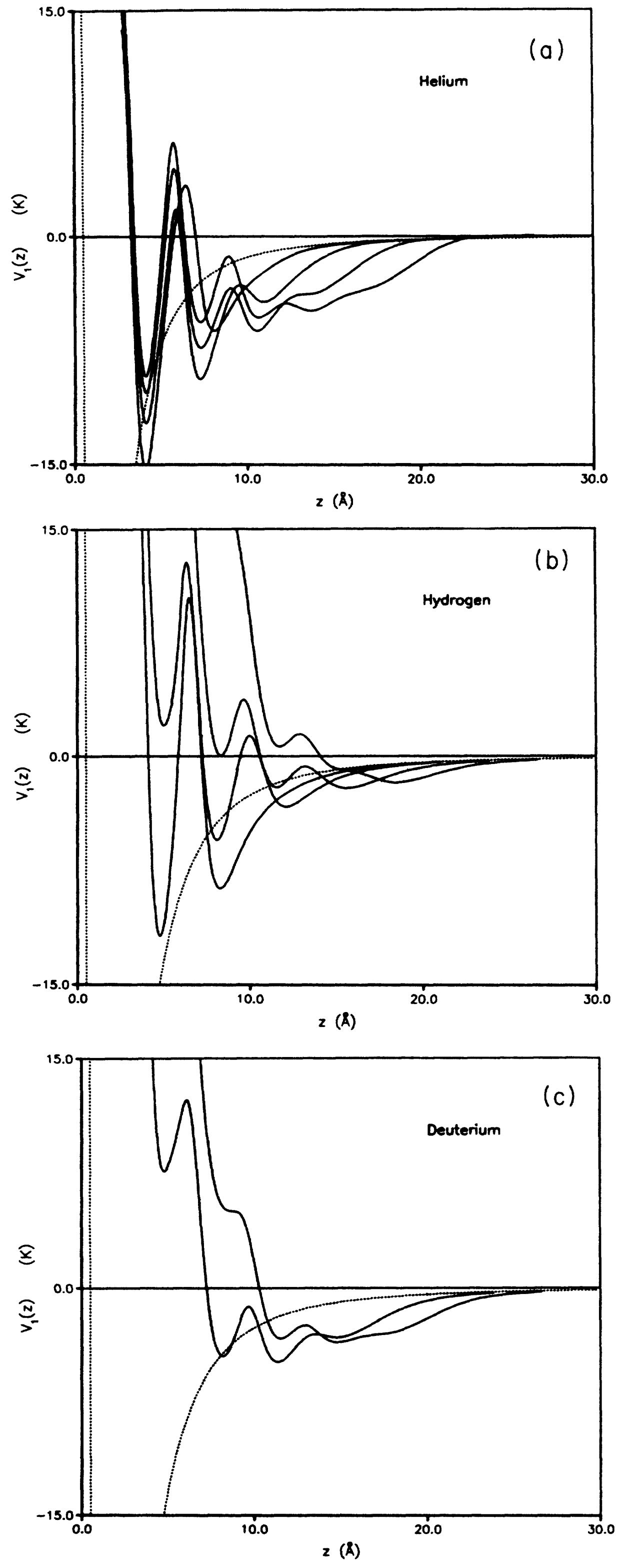

FIG. 2. (a) The self-consistent one-body potential $V_1(z)=U_{\rm sub}(z)+V_{\rm SC}(z)$ for a ^{3}He impurity is shown for the four surface coverages $n=0.15$, 0.20, 0.25, and 0.30 Å^{-2}. The potentials with the longer ranges correspond to the higher surface coverages. Also shown is the bare-substrate potential $U_{\rm sub}(z)$ (dotted line). (b) Same as Fig. 2(a) for the hydrogen impurity. (c) Same as Fig. 2(a) for the deuterium impurity for the surface coverages $n=0.25$ and 0.30 Å^{-2}.

gy for the H atom due to mesh restrictions into account, we estimate that a H atom would have a binding energy to the free surface of about 0.2 K.

The binding energy of a deuterium atom turned out to be about 2.5 K. The numerical uncertainties due to the finite mesh-size are smaller in this case since the D atoms are more tightly bound, c.f., Figs. 1(a) and 1(b). The widths of the bound states for the two surface coverages are very similar. A comparison of the one-body potential for the different surface coverages [c.f., Fig. 2(c)] suggests that the substrate potential contributes about twenty percent of the total binding energy, i.e., we expect that a D atom will have a binding energy of about 2 K to the free surface of liquid ^{4}He.

Our results are in reasonable agreement with those of Mantz and Edwards.[29] It is expected that our binding energy of the deuterium atom is somewhat larger since the authors of Ref. 29 assumed that the ^{4}He-D distribution function is identical to the ^{4}He-^{4}He distribution function. This should lead to a weaker binding due to the stronger repulsion between the He and the D atom. One expects that the effect is less severe for the case of the H impurity, since this atom is located farther away from the surface. The short-ranged structure of the correlations should therefore, be less important. A further source of uncertainty is the intrinsic inaccuracy of the HNC approximation, which predicts for bulk ^{4}He a binding energy of -5.4 K at a saturation density of 0.017 fm^{-3}, as opposed to the experimental value of -7.2 K at 0.021 85 fm^{-3}. One may therefore argue that the HNC approximation may also underestimate the binding energy of an impurity.

Whereas the hydrogen isotopes form essentially surface states, the ^{3}He impurity always has a significant overlap with the last filled layer. In the double-layer film [Fig. 1(d)], the ^{3}He impurity is seen to be a part of the second layer. As the film thickness increases [Figs. 1(c) and 1(b)], the overlap with the inner parts of the film becomes smaller and the particle gets more expelled to the surface. We expect that our calculation for the largest film [Fig. 1(a)] is representative for the limit of an infinite half-space. Our ^{3}He densities shown in Figs. 1(a)–1(d) are in essential agreement with the results of our earlier work.[28]

Figures 3(a) and 3(b) finally show the excited states of the helium and the deuterium particles in the film with the surface coverage $n=0.30$ Å^{-2}, i.e., the densities $\rho_1(z_0)=|\psi_1(z_0)|^2$ corresponding to the bound eigenstates of Eq. (2.10) above the ground state. We note that for the two films with surface coverage $n=0.25$ and 0.30 Å^{-2} we found a very weak fourth bound state of a ^{3}He impurity with an energy of about 0.1 K. This state is very widely spread into the vacuum region. We therefore felt that our energy estimate would not be very accurate due to numerical uncertainties. Our results for the ^{3}He excited states are in essential agreement with those of Ref. 28. We also found a first excited state of the deuterium impurity of a shape very similar to the first ^{3}He excited state. It is again worth noting here the qualitative difference between the two systems: If the thickness of the film is increased, one will obtain for the ^{3}He impurity a dense band of bound states corresponding to ^{3}He impurities living in the bulk. The energy difference between this continuum and the lowest state is about 2.2 K, and one is tempted to identify the *third* state of the ^{3}He impurity with the lower boundary of this continuum. A similar continuum will not exist for the hydrogen isotopes which will be expelled from the bulk liquid. The first excited state of both the ^{3}He and the D impurity appear to be surface-bound states. The existence of such a second state of the ^{3}He impurity, which is further spread out than the surface-bound state, seems to be necessary to interpret the experimental data[12] on the specific heat of dilute ^{3}He systems in the surface of ^{4}He.

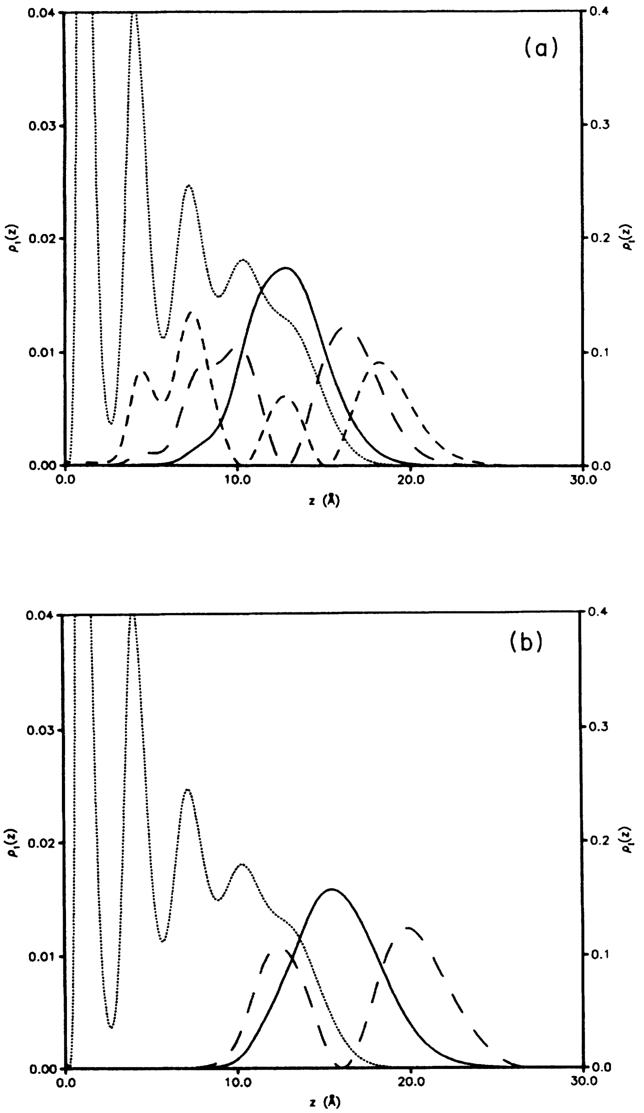

FIG. 3. (a) The shape of the excited states of the ^{3}He impurity is shown for the surface coverage $n = 0.30$ Å^{-2}. Solid line: ground state. Long-dashed line: first excited state. Short-dashed line: second excited state. Also shown is the background density (dotted line). (b) Same as Fig. 3(a) for the deuterium impurity.

V. POLARON STATES

So far we have assumed that the state of the impurity atom is translationally invariant in the plane parallel to the surface. In this section, we investigate in some detail the possibility of a spontaneous breaking of that symmetry. Such a situation can occur when the impurity becomes more strongly bound with decreasing thickness of the background film. The impurity particle can then gain potential energy by moving closer to the substate. In that process, however, the background has to be deformed, which increases its energy. The localization of the impurity costs additional kinetic energy. The resulting state is a *localized* impurity state on a *deformed* background.

Most closely related to our work is that of Guyer *et al.*,[19] who studied the formation of self-trapped states of hydrogen atoms on the surface of ^{4}He. In that work a hydrodynamic approach was taken towards the calculation of the collective excitations, and the impurity-background interaction was assumed to be weak. Some delicate considerations are due in order to formulate the polaron problem in the framework of the optimized variational theory.

The original objective in writing the ground-state wave function in the form (2.1) has been to separate one- and two-body quantities. The pair correlations have been made unique by requiring that they vanish when two particles are moved far from each other. If the impurity particle is localized, such a distinction is no longer possible, and $u_1(\mathbf{r}_0)$ and $u_2(\mathbf{r}_0,\mathbf{r}_i)$ are no longer independent functions. We therefore, have a certain amount of freedom to describe the polaron state.

For the sake of the practical application of the theory, we adopt the following picture: We *assume* that the Euler equations have been solved for the translationally invariant case as described in Sec. II. Let us denote the corresponding one-body functions $u_1^{(0)}(\mathbf{r}_i)$ for $i=0,\ldots A$, the corresponding wave function with $\Psi_0(\mathbf{r}_0,\ldots,\mathbf{r}_A)$, and the energy expectation value with respect to this wave function with $E_{(0)}$. The impurity density is, in that case,

$$\rho_1^{(0)}(\mathbf{r}_0)=\frac{\phi^2(z)}{L^2} , \tag{5.1}$$

where L is the assumed box size parallel to the surface, and $\phi(z)$ is normalized such that

$$\int_0^\infty \phi^2(z)dz=1 . \tag{5.2}$$

We now try to find a lower-energy state by breaking the translational invariance of the one-body functions in the x-y plane, but keep the two-body functions fixed. We assume a *macroscopic* change of $u_1(\mathbf{r}_0)$ and an *infinitesimal* change of the one-body function of the background

$$u_1(\mathbf{r}_i)=u_1^{(0)}(\mathbf{r}_i)+\delta u_1(\mathbf{r}_i) \quad (i=1,\ldots,A) , \tag{5.3}$$

in order to accomodate the response of the background.

Assuming the center of the polaron state at the coordinate origin, we now have an impurity density

$$\rho_1(\mathbf{r}_0)\to 0 \quad (\,|\mathbf{r}_0|\to\infty\,)\ , \tag{5.4}$$

with the normalization

$$\int d^3r_0\rho_1(\mathbf{r}_0)=1\ . \tag{5.5}$$

Let us denote this new, "polaronic" ground-state wave function with $\Psi_P(\mathbf{r}_0,\ldots,\mathbf{r}_A)$. For this wave function, we must evaluate the energy to second order in $\delta u_1(\mathbf{r}_i)$, assuming that the two-body correlations have been optimized for the reference functions. In other words, we calculate for some $u_1(\mathbf{r}_0)$, which is not necessarily close to $u_1^{(0)}(\mathbf{r}_0)$, the expansion for the polaron energy

$$\begin{aligned}\Delta E_P &\equiv E[u_1(\mathbf{r}_0),u_1(\mathbf{r}_i)]-E_{(0)}\\ &=E[u_1(\mathbf{r}_0),u_1^{(0)}(\mathbf{r}_i)]-E_{(0)}+\int d^3r_1\delta u_1(\mathbf{r}_1)\left.\frac{\delta E[u_1(\mathbf{r}_0),u_1(\mathbf{r}_i)]}{\delta u_1(\mathbf{r}_1)}\right|_{u_1(\mathbf{r}_i)=u_1^{(0)}(\mathbf{r}_i)}\\ &\quad+\tfrac{1}{2}\int d^3r_1d^3r_2\delta u_1(\mathbf{r}_1)\delta u_1(\mathbf{r}_2)\left.\frac{\delta^2E[u_1(\mathbf{r}_0),u_1(\mathbf{r}_i)]}{\delta u_1(\mathbf{r}_1)\delta u_1(\mathbf{r}_2)}\right|_{u_1(\mathbf{r}_i)=u_1^{(0)}(\mathbf{r}_i)}=\Delta E_0+\Delta E_1+\Delta E_2\ .\end{aligned} \tag{5.6}$$

The calculations are somewhat delicate and are therefore, displayed here in some detail. The relation between the one-body functions $u_1(\mathbf{r}_0)$, the densities, and the normalization integrals

$$I=\int d^3r_0\ldots d^3r_A\Psi_P^2(\mathbf{r}_0,\ldots,\mathbf{r}_A)\ . \tag{5.7}$$

[The normalization integral $I^{(0)}$ is defined correspondingly with $\Psi_0(\mathbf{r}_0,\ldots,\mathbf{r}_A)$] is

$$\frac{\rho_1(\mathbf{r}_0)}{\rho_1^{(0)}(\mathbf{r}_0)}=\frac{I^{(0)}}{I}\exp[u_1(\mathbf{r}_0)-u_1^{(0)}(\mathbf{r}_0)]\ , \tag{5.8}$$

and the Euler equations for the one- and two-body components of the reference wave function $\Psi_0(\mathbf{r}_0,\ldots,\mathbf{r}_A)$ are

$$\frac{1}{I^{(0)}}\int d^3r_1\ldots d^3r_A\Psi_0^2(\mathbf{r}_0,\ldots,\mathbf{r}_A)[V_{(0)}^*(\mathbf{r}_0,\ldots,\mathbf{r}_A)-E_{(0)}]=\frac{\hbar^2}{8m_0}\nabla^2\rho_1^{(0)}(\mathbf{r}_0)\ , \tag{5.9}$$

and

$$\frac{A}{I^{(0)}}\int d^3r_2\ldots d^3r_A\Psi_0^2(\mathbf{r}_0,\ldots,\mathbf{r}_A)[V_{(0)}^*(\mathbf{r}_0,\ldots,\mathbf{r}_A)-E_{(0)}]=\left[\frac{\hbar^2}{8m_0}\nabla_0^2+\frac{\hbar^2}{8m}\nabla_1^2\right]\rho_2^{(0)}(\mathbf{r}_0,\mathbf{r}_1)\ . \tag{5.10}$$

Here, $E_{(0)}$ is the total energy of the system evaluated for the reference functions $u_1^{(0)}(\mathbf{r}_i)$, and $V_{(0)}^*$ the quantity (2.9) for the same wave function.

The second-order change in the total energy of the system due to the change in the one-body function of the background can be calculated rather easily if one assumes that this term is dominated by the deformation energy of the background. We then can follow the linear response theory of Ref. 28 and obtain *for optimized two-body correlations* $u_2(\mathbf{r}_i,\mathbf{r}_j)$

$$\Delta E_2=\frac{\hbar^2}{8m}\int d^3r_1\rho_1(\mathbf{r}_1)\,|\,\nabla\delta u_1(\mathbf{r}_1)\,|^2\ . \tag{5.11}$$

For the first term in the expansion (5.6) we find

$$\begin{aligned}\Delta E_0&=\frac{1}{I}\int d^3r_0\ldots d^3r_A\Psi_P^2(\mathbf{r}_0,\ldots,\mathbf{r}_A)[V^*(\mathbf{r}_0,\ldots,\mathbf{r}_A)-E_{(0)}]\\ &=\frac{1}{I}\int d^3r_0\ldots d^3r_A e^{[u_1(\mathbf{r}_0)-u_1^{(0)}(\mathbf{r}_0)]}\Psi_0^2(\mathbf{r}_0,\ldots,\mathbf{r}_A)\left[V_{(0)}^*(\mathbf{r}_0,\ldots,\mathbf{r}_A)-E_{(0)}-\frac{\hbar^2}{8m_0}\nabla^2[u_1(\mathbf{r}_0)-u_1^{(0)}(\mathbf{r}_0)]\right]\\ &=\frac{I^{(0)}}{I}\frac{\hbar^2}{8m_0}\int d^3r_0e^{[u_1(\mathbf{r}_0)-u_1^{(0)}(\mathbf{r}_0)]}\left[\nabla^2\rho_1^{(0)}(\mathbf{r}_0)-\rho_1^{(0)}(\mathbf{r}_0)\nabla^2[u_1(\mathbf{r}_0)-u_1^{(0)}(\mathbf{r}_0)]\right]\\ &=\frac{\hbar^2}{2m_0}\int d^3r_0\rho_1^{(0)}(\mathbf{r}_0)\left|\nabla\left[\frac{\rho_1(\mathbf{r}_0)}{\rho_1^{(0)}(\mathbf{r}_0)}\right]^{1/2}\right|^2\equiv\int d^3r_0t(\mathbf{r}_0)\ .\end{aligned} \tag{5.12}$$

In the derivation of Eq. (5.12) we have used the Euler equation (5.9) and the relation (5.8). The derivation of the second term in the expansion (5.6) is similar. We write

$$\left.\frac{\delta E[u_1(\mathbf{r}_0),u_1(\mathbf{r}_i)]}{\delta u_1(\mathbf{r}_1)}\right|_{u_1(\mathbf{r}_i)=u_1^{(0)}(\mathbf{r}_i)}$$

$$=-\frac{\hbar^2}{8m}\nabla^2\rho_1(\mathbf{r}_1)+\frac{A}{I}\int d^3r_0d^3r_2\dots d^3r_A\Psi_P^2(\mathbf{r}_0,\dots,\mathbf{r}_A)[V^*(\mathbf{r}_0,\dots,\mathbf{r}_A)-E]\ , \tag{5.13}$$

where E is the energy of the system calculated with the *deformed* one-body function $u_1(\mathbf{r}_0)$, i.e., $E=E_{(0)}+\Delta E_0$. Using the two-body Euler equation (5.10) to eliminate $V^*(\mathbf{r}_0,\dots,\mathbf{r}_A)$, we find

$$\begin{aligned}\left.\frac{\delta E[u_1(\mathbf{r}_0),u_1(\mathbf{r}_i)]}{\delta u_1(\mathbf{r}_1)}\right|_{u_1(\mathbf{r}_i)=u_1^{(0)}(\mathbf{r}_i)}&=\frac{\hbar^2}{8m_0}\int d^3r_0\rho_2(\mathbf{r}_0,\mathbf{r}_1)\,|\,\nabla[u_1(\mathbf{r}_0)-u_1^{(0)}(\mathbf{r}_0)]\,|^2-\Delta E_0\rho_1(\mathbf{r}_1)\\&=\int d^3r_0\left[t(\mathbf{r}_0)-\Delta E_0\rho_1(\mathbf{r}_0)\right]\frac{\rho_2(\mathbf{r}_0,\mathbf{r}_1)}{\rho_1(\mathbf{r}_0)}\\&=\int d^3r_0\tau(\mathbf{r}_0)h(\mathbf{r}_0,\mathbf{r}_1)\rho_1(\mathbf{r}_1)\ .\end{aligned}\tag{5.14}$$

Here, we have defined in the last line

$$\tau(\mathbf{r}_0)=t(\mathbf{r}_0)-\Delta E_0\rho_1(\mathbf{r}_0)\ , \tag{5.15}$$

and $h(\mathbf{r}_0,\mathbf{r}_1)\equiv g(\mathbf{r}_0,\mathbf{r}_1)-g(\infty)$, where $g(\infty)$ is the value of the pair correlation function in the limit $|\mathbf{r}_0-\mathbf{r}_1|\to\infty$, which may deviate from unity by an amount of $1/L^2$. We can now take for $h(\mathbf{r}_0,\mathbf{r}_1)$ the correlation function for the reference system. Finally, we can eliminate the deformation of the background one-body function $\delta u_1(\mathbf{r})$ in favor of the deformation of the background density $\delta\rho_1(\mathbf{r})$ and write the total energy shift in terms of the fluctuation of the densities

$$\rho_1(\mathbf{r})\delta u_1(\mathbf{r})=\delta\rho_1(\mathbf{r})-\rho_1(\mathbf{r})\int d^3r'X(\mathbf{r},\mathbf{r}')\delta\rho_1(\mathbf{r}')\ , \tag{5.16}$$

and find

$$\Delta E_1=\int d^3r_0d^3r_1\tau(\mathbf{r}_0)X(\mathbf{r}_0,\mathbf{r}_1)\delta\rho_1(\mathbf{r}_1) \tag{5.17}$$

and

$$\Delta E_2=\tfrac{1}{4}\int d^3r_1d^3r_2\frac{\delta\rho_1(\mathbf{r}_1)}{\sqrt{\rho_1(\mathbf{r}_1)}}[H_1(\mathbf{r}_1)\delta(\mathbf{r}_1-\mathbf{r}_2)+2\tilde{V}_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)]\frac{\delta\rho_1(\mathbf{r}_2)}{\sqrt{\rho_1(\mathbf{r}_2)}}\ . \tag{5.18}$$

The next step is to minimize the total energy shift ΔE_P with respect to the deformation of the background density. The optimal deformation is given by the relation

$$\frac{\delta\rho_1(\mathbf{r}_1)}{\sqrt{\rho_1(\mathbf{r}_1)}}=-2\int d^3r_2[H_1(\mathbf{r}_1)\delta(\mathbf{r}_1-\mathbf{r}_2)+2\tilde{V}_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)]^{-1}\int d^3r_0\tilde{X}(\mathbf{r}_2,\mathbf{r}_0)\frac{\tau(\mathbf{r}_0)}{\sqrt{\rho_1(\mathbf{r}_0)}}\ , \tag{5.19}$$

where the inverse is understood as an operator inverse, which can easily be calculated from the normal-mode decomposition of the background excitations. Inserting the result (5.19) into ΔE_P we find

$$\Delta E_P=\Delta E_0-\int d^3r_0d^3r_0'\tau(\mathbf{r}_0)W(\mathbf{r}_0,\mathbf{r}_0')\tau(\mathbf{r}_0')\ , \tag{5.20}$$

with

$$\tilde{W}(\mathbf{r}_0,\mathbf{r}_0')=\int d^3r_1d^3r_2\tilde{X}(\mathbf{r}_0,\mathbf{r}_1)[H_1(\mathbf{r}_1)\delta(\mathbf{r}_1-\mathbf{r}_2)+2\tilde{V}_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)]^{-1}\tilde{X}(\mathbf{r}_2,\mathbf{r}_0')\ . \tag{5.21}$$

The expression for the interaction $W(\mathbf{r}_0,\mathbf{r}_0')$ becomes especially simple when we consider its matrix elements between the impurity states $\psi_i(\mathbf{r}_0)$ [Eq. (2.34)]. Using the spectral decomposition of the operator $[H_1(\mathbf{r}_1)\delta(\mathbf{r}_1-\mathbf{r}_2)+2\tilde{V}_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)]$ derived from the eigenvalue problem (A8), and the formal solution (A12) for the matrix elements of $X(\mathbf{r}_0,\mathbf{r}_1)$, we obtain

$$(\psi_i\,|\,\tilde{W}(\mathbf{r}_0,\mathbf{r}_0')\,|\,\psi_j)=4\sum_l\frac{(\psi_i\,|\,\tilde{V}^I_{p\text{-}h}\,|\,\phi_l)}{t_i+\hbar\omega_l}\frac{1}{\hbar\omega_l}\frac{(\psi_j\,|\,\tilde{V}^I_{p\text{-}h}\,|\,\phi_l)}{t_j+\hbar\omega_l}\ . \tag{5.22}$$

In plane surface geometry as studied here, the subscripts labeling the states contain both the discrete variable perpendicular to the surface and the continuous momentum parallel to the surface.

Before we proceed with a numerical evaluation of the polaron energy, it is worth considering the formal structure of our result. Attention is directed to the Euler equation for the impurity-background correlations (2.32) and its formal solution in terms of the excitation energies of the background system (2.37). There is no way that an instability of the equations could build up as it does, for example, for the bulk liquid for sufficiently low and high densities. This means that the translationally invariant impurity state is *always a local minimum* of the total energy of the system. The Euler equations give no indication for the instability of the system against the formation of a polaron. This result is in agreement with the structure of our energy functional (5.20), (5.15) for the polaron state. Note that the quantity $\tau(\mathbf{r}_0)$ is of *second order* in the deviation of the polaron wave function from the translationally invariant state. This means that the potential-energy term in Eq. (5.20) is of *fourth order* in that deviation, and small perturbations of the translationally invariant impurity state will only contribute to the kinetic energy contribution ΔE_0, which is positive. In other words, small perturbations of a translationally invariant state will never indicate the formation of a polaron.

From our derivations of Sec. III we conclude that the softening of the surface excitations for large films does not cause an instability against the formation of a polaron. Using the form (3.3) for the lowest-lying mode we obtain for the dominant contribution to polaron self-interaction matrix element in the limit $q_\parallel \to 0$

$$(\psi_0 \,|\, \tilde{W} \,|\, \psi_0) = \frac{2}{\hbar^2 m c_3^4 q_\parallel^2} \,|\, (\rho_1^i \,|\, V_{p\text{-}h}^I(0+) \,|\, \delta\rho_1) \,|^2 \,. \tag{5.23}$$

The singularity at $q_\parallel \to 0$ does not cause a problem since the volume integral of $\tau(\mathbf{r}_0)$ [c.f., Eq. (5.15)] vanishes.

From Eq. (5.23) one might be led to the impression that the polaron self-interaction becomes stronger with increasing film thickness. However, as the film approaches the infinite half-space, the shape of the collective excitation approaches the form (3.9) for the ripplon, and the interaction matrix element in the numerator vanishes. To obtain the correct limit of the infinite half-space, we have to use the form (3.9) for the shape of the density fluctuation, use the expansion (3.16) and observe (3.17). We then obtain

$$\begin{aligned}(\psi_0 \,|\, \tilde{W} \,|\, \psi_0) &= \frac{2 q_\parallel^5}{\hbar^2 m \rho_\infty \omega_r^4} \left| \left[\rho_1^I(z_0) \,|\, V_1^I \,|\, \frac{d\rho_1(z_1)}{dz_1} \right] \right|^2 \\ &= \frac{2 m \rho_\infty}{\hbar^2 \sigma^2 q_\parallel} \left| \left[\rho_1^I(z_0) \,|\, v_1^I \,|\, \frac{d\rho_1(z_1)}{dz_1} \right] \right|^2 .\end{aligned} \tag{5.24}$$

We see that the singularity of the polaron self-interaction becomes even weaker in the surface of an infinite half-space.

Let us now turn to our numerical investigation of the polaron states. The energy functional (5.20) would still lead to a two-dimensional equation describing the polaron density. In order to simplify the problem, we assume that the z dependence of the polaron state is identical to the one of the translationally invariant state, i.e., we assume a density

$$\rho_1(\mathbf{r}_0) \approx \phi^2(z) \Theta^2(r_\parallel) \,, \tag{5.25}$$

with the normalization

$$\int d^2 r_\parallel \Theta^2(r_\parallel) = 1 \,. \tag{5.26}$$

This specific form has the effect that only the ground-state matrix elements $[\psi_0 \,|\, W(\mathbf{r}_0, \mathbf{r}_0') \,|\, \psi_0)]$ contribute to the energy functional (5.20). The z dependence can be integrated out exactly, and the polaron energy can then be written in momentum space as

$$\Delta E_P = \Delta E_0 - \int \frac{d^2 q_\parallel}{(2\pi)^2} \tau^2(q_\parallel) W(q_\parallel) \,, \tag{5.27}$$

where ΔE_0 simplifies to

$$\Delta E_0 = \frac{\hbar^2}{2m_0} \int d^2 r_\parallel \,|\, \nabla \Theta(r_\parallel) \,|^2 \,, \tag{5.28}$$

and

$$\tau(r_\parallel) = \frac{\hbar^2}{2m_0} \,|\, \nabla \Theta(r_\parallel) \,|^2 - \Delta E_0 \Theta(r_\parallel)^2 \,, \tag{5.29}$$

and $\tau(q_\parallel)$ is the two-dimensional Fourier transform of $\tau(r_\parallel)$. For a bound state, $\tau(r_\parallel)$ will fall off exponentially for large $r_\parallel$, its Fourier transform is therefore, analytic at the origin. Since the volume integral of $\tau(r_\parallel)$ vanishes, we have

$$\tau(q_\parallel) \sim q_\parallel^2 (q_\parallel \to 0) \,. \tag{5.30}$$

The resulting energy functional can now be minimized with respect to the function $\Theta(\mathbf{r}_\parallel)$ which leads to an equation reminiscent of a Hartree equation in two dimensions. Instead of solving this equation, we have chosen to minimize the energy using a trial function

$$\Theta(\mathbf{r}_\parallel) = \frac{1}{a\sqrt{\pi}} e^{-r^2/2a^2} \,, \tag{5.31}$$

which leads to an energy functional of the form

$$\Delta E_P = \frac{\hbar^2}{2m_0 a^2} - \tfrac{1}{16} \int \frac{d^2 q_\parallel}{(2\pi)^2} \left[\frac{\hbar^2 q_\parallel^2}{2m_0} \right]^2 W(q_\parallel) e^{-1/2 q_\parallel^2 a^2} \,. \tag{5.32}$$

The $q_\parallel^4$ factor under the potential-energy integral of Eq. (5.31) is a consequence of Eq. (5.30).

The polaron self-interaction weighted with the $q_\parallel^4$ factor coming from Eq. (5.30) is shown in Fig. 4. The function is seen to have a double-peak structure and falls off rather rapidly for momentum transfers larger than 2 Å^{-1}. The potential is strongest for the very thin film, since the impurity particle is attracted in that case most strongly to the substrate, which favors the formation of a polaron. Considering only the long-wavelength structure (5.23) of the polaron self-interaction without taking into account the change of the interaction matrix element

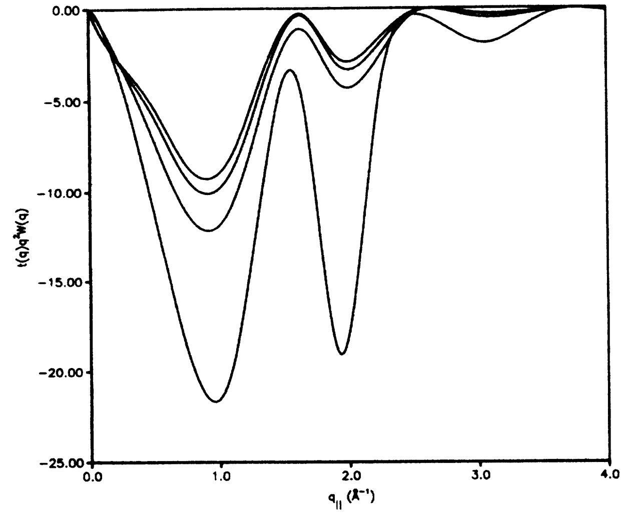

FIG. 4. The weighted polaron self-interaction $t(q)q^2W(q)$, where $t(q)=\hbar^2/2m\ q^2$, is shown for the ^{3}He impurity for the surface coverages $n=0.15$, 0.20, 0.25, and 0.30 Å^{-2}. The strongest potential corresponds to $n=0.15$ Å^{-2}; the interaction becomes weaker with increasing surface coverage.

with the film thickness would have led here to the exactly opposite conclusion. The corresponding quantities for the H isotopes are a factor of 3–10 smaller.

We have evaluated the polaron energy expression ΔE_P as a function of the polaron width a. We found that the potential-energy contribution never exceeds 15% of the kinetic energy. Since the attractive part of the energy is so small, we have decided that the rigorous minimization of Eq. (5.25) with respect to $\Theta(r_\parallel)$ is not necessary. We therefore conclude that the formation of a polaron state is energetically unfavorable in all cases studied here.

VI. SUMMARY

We have, in this paper, developed a systematic variational theory for the study of impurities in liquid surfaces. We have concentrated on the static properties of the impurities, i.e., the structure of the ground state and its discrete excited states. The interpretation of our results, an assessment of their accuracy, and the general route of improving the calculational accuracy has been given in the text and does not need to be repeated here. One of the most important results is our conclusion that neither of the impurities studied here form a "polaron" state.

The same theory could be applied equally well to the free surface, where we expect that one will find a tritium bound state. For our present purposes, we found that the adsorbed films studied here provide richer physics and were computationally more convenient. A study of ^{3}He bound states in the free surface of ^{4}He was recently performed by Dalfovo *et al.*,[43] who found, using a nonlocal density-functional theory,[44,36] also two surface-bound states. The theory of Ref. 44 give an impressively accurate description of the structure of the free surface of ^{4}He, and we expect therefore, that the results of Ref. 43 are quite reliable. Unfortunately, the nonlocal density-functional theory does not reproduce the layer structure of the adsorbed films, which is apparently due to the absence of short-ranged correlation.[45]

Two problems have remained open for further study: The impurity self-energy (and the related effective mass), and the impurity-impurity interaction. It is rather straightforward to calculate the impurity effective mass using either correlated-basis functions (CBF) theory for the self-energy,[46–48] or using "backflow" correlation functions.[49,50] In the "uniform limit," i.e., if one assumes that the "backflow" correlations are weak, but long ranged, the second-order CBF theory and the variational ("backflow") approach lead to exactly the same answer. Since the effective mass of the impurity particles comes out to be rather large, we felt that low-order methods must be evaluated very carefully before microscopic conclusions on the effective mass can be drawn. A second problem which remains to be studied is the impurity-impurity interaction,[51] in particular the question whether impurities can form bound states in the surface of a helium film. There is experimental evidence that this can happen for both ^{3}He (Ref. 52) and electronic impurities.[53]

We have commented at various places in the text that an obvious alternative field of application would be electrons adsorbed to the surface.[53–57] Since this system is sufficiently different from the problem of atomic impurities, we have decided to devote an independent study to the electronic surface states.

ACKNOWLEDGMENTS

This work was supported, in part, by the U.S. National Science Foundation under Contract No. PHY-8505979 and the Robert A. Welch foundation under Grant No. A111. We would like to thank G. Agnolet, W. Kirk, R. A. Smith, and W. Saslow for stimulating discussions. One of us (J.L.E.) would like to thank the College of Engineering, Texas A&M University, for providing a part of the computational resources.

APPENDIX

Numerical optimization

As an introductory exercise, and to display the general scheme for the numerical solution of the two-body Euler equation, we discuss first the paired-phonon-analysis (PPA) iterations for the impurity problem in the bulk. The solutions of the bulk problem are needed to generate reasonable initial solutions for the inhomogeneous problem. The binding energy (2.27) reduces in that case to

$$\begin{aligned}\Delta E(\text{bulk})=\rho\int d^3r\Bigg[&g_I(r)v_I(r)\\&+\left[\frac{\hbar^2}{2m_0}+\frac{\hbar^2}{2m}\right]|\nabla\sqrt{g_I(r)}\,|^2\Bigg]\\&-\frac{\hbar^2}{8m_0\rho}\int\frac{d^3k}{(2\pi)^3}k^2S_I^2(k)[1-1/S(k)]\\&-\frac{\hbar^2}{8m\rho}\int\frac{d^3k}{(2\pi)^3}k^2S_I^2(k)[1-1/S^2(k)]\ .\end{aligned}\tag{A1}$$

We have labeled here all impurity quantities with a subscript I. Note that $S_I(k)$ is the Fourier transform of $g_I(r)-1$. The induced potential in momentum space is (as usual, we use the dimensionless Fourier transform which contains a density factor)

$$\begin{aligned}\tilde{w}_I(k)= & -\frac{\hbar^2k^2}{4m_0}\frac{S_I(k)}{S(k)}[S(k)-1]\\ & -\frac{\hbar^2k^2}{4m}\frac{S_I(k)}{S^2(k)}[S^2(k)-1]\end{aligned} \tag{A2}$$

and the EL equation is

$$S_I(k)=-\frac{2\tilde{V}^I_{p\text{-}h}(k)S(k)}{\hbar\omega(k)+t(k)} , \tag{A3}$$

with

$$\begin{aligned}V^I_{p\text{-}h}(r)= & g_I(r)v_I(r)+\left[\frac{\hbar^2}{2m_0}+\frac{\hbar^2}{2m}\right]|\nabla\sqrt{g_I(r)}|^2\\ & +[g_I(r)-1]w_I(r) .\end{aligned} \tag{A4}$$

Equation (A3) has been written, in view of our algorithm for solving the inhomogeneous problem, in a somewhat suggestive way. $\hbar\omega(k)=\hbar^2k^2/2mS(k)$ is the zero-sound spectrum, and $t(k)=\hbar^2k^2/2m_0$ the kinetic energy of the impurity particle. The PPA procedure to solve the Euler equation is as follows.

(a) Start from a reasonable guess for $g_I(r)$. For the ^{3}He impurity, it is sufficient to start from a distribution function for ^{4}He, for the hydrogen impurities the same initial correlations can be taken after shifting them somewhat in the radial direction to accomodate the larger core size of the hydrogen-helium potential. Obtain from this $g_I(r)$ the corresponding $S_I(k)$, $\tilde{w}_I(k)$, and $w_I(r)$.

(b) Calculate the impurity particle-hole interaction (A4) in coordinate space.

(c) Fourier transform and get $S_I(k)$ from Eq. (A3). Use this $S_I(k)$ to get a new $\tilde{w}_I(k)$.

(d) Fourier transform $S_I(k)$ and $w_I(k)$. This leads to a new estimate $g_I^{\text{PPA}}(r)$. However, this pair-distribution function will not, in general, vanish inside the repulsive core of the potential unless convergence has been reached. A better approximation $g_I^{\text{new}}(r)$ is therefore generated by

$$g_I^{\text{new}}(r)=g_I^{\text{old}}(r)\exp[g_I^{\text{PPA}}(r)-g_I^{\text{old}}(r)] . \tag{A5}$$

The steps (b)–(d) are then repeated until convergence is reached. One sees that the PPA procedure requires three Fourier transforms per iteration and is therefore extremely fast.

Exactly the same procedure is used in the inhomogeneous case. What is left is to derive the relationship between the solutions of the eigenvalue problem (2.35) and the collective modes of the background. The eigenvalues of (2.35) are real since $H_1(\mathbf{r}_1)$ is a positive definite operator, but eigenfunctions $\phi_i(\mathbf{r})$ are not orthogonal on a Euclidean mesh. Rather, they satisfy the orthogonality relation

$$(\phi_i\,|\,\delta(\mathbf{r}-\mathbf{r}')-\tilde{X}(\mathbf{r},\mathbf{r}')\,|\,\phi_j)=\delta_{ij} . \tag{A6}$$

In the course of the development of the HNC theory for inhomogeneous systems,[28] it was shown that the $\tilde{X}(\mathbf{r}_1,\mathbf{r}_2)$ has the representation

$$\delta(\mathbf{r}_1-\mathbf{r}_2)-\tilde{X}(\mathbf{r}_1,\mathbf{r}_2)=\sum_l \hbar\omega_l\Psi^{(l)}(\mathbf{r}_1)\Psi^{(l)}(\mathbf{r}_2) , \tag{A7}$$

where the $\Psi_i(\mathbf{r})$ are the solutions of the eigenvalue problem

$$\int d^3r_2[H_1(\mathbf{r}_1)\delta(\mathbf{r}_1-\mathbf{r}_2)-2\tilde{V}_{p\text{-}h}(\mathbf{r}_1,\mathbf{r}_2)]H_1(\mathbf{r}_2)\Psi^{(l)}(\mathbf{r}_2)=\hbar^2\omega_l^2\Psi^{(l)}(\mathbf{r}_1) , \tag{A8}$$

with the orthogonality relation

$$(\Psi^{(l)}\,|\,H_1\,|\,\Psi^{(m)})=\delta_{l,m} . \tag{A9}$$

We have identified in Ref. 28 the energies $\hbar\omega_l$ with the collective excitations of the system, and $\delta[\rho_1^{(l)}(\mathbf{r})]^{1/2}=H_1(\mathbf{r})\Psi^{(l)}(\mathbf{r})$ with the spatial shape of the density fluctuations. From that we get

$$\begin{aligned}\int d^3r_2 & [\delta(\mathbf{r}_1-\mathbf{r}_2)-\tilde{X}(\mathbf{r}_1,\mathbf{r}_2)]H_1(\mathbf{r}_2)\Psi^{(m)}(\mathbf{r}_2)\\ & =\sum_l \hbar\omega_l\Psi^{(l)}(\mathbf{r}_1)(\Psi^{(l)}\,|\,H_1\,|\,\Psi^{(m)})\\ & =\hbar\omega_m\Psi^{(m)}(\mathbf{r}_1) .\end{aligned} \tag{A10}$$

Therefore, we have that the eigenvectors and eigenvalues of Eq. (2.35) are the density fluctuations and excitation energies of the background system. The exact relationship is finally determined by the normalization (A6), which leads to

$$|\,\phi_i)=(\hbar\omega_i)^{-1/2}H_1\,|\,\Psi^{(i)}) . \tag{A11}$$

To obtain the solution of Eq. (2.32), we sandwich the equation between $\psi_i(\mathbf{r}_0)$ from the left, and $\phi_i(\mathbf{r}_1)$ from the right. This gives us

$$(\psi_i\,|\,\tilde{X}^I\,|\,\phi_j)=-2\frac{(\psi_i\,|\,\tilde{V}^I_{p\text{-}h}\,|\,\phi_j)}{t_i+\hbar\omega_j} . \tag{A12}$$

Having the matrix elements of $X(\mathbf{r}_0,\mathbf{r}_1)$ we can now also get the $X(\mathbf{r}_0,\mathbf{r}_1)$:

$$X(\mathbf{r}_0,\mathbf{r}_1)=-2\sum_{i,j}\psi_i(\mathbf{r}_0)\frac{(\psi_i\,|\,V_{p\text{-}h}\,|\,\phi_j)}{\varepsilon_i+\hbar\omega_j}[(1-\tilde{X})\phi_j](\mathbf{r}_1) \tag{A13}$$

and

$$\begin{aligned}S(\mathbf{r}_0,\mathbf{r}_1) & =\int d^3r_2\tilde{X}(\mathbf{r}_0,\mathbf{r}_2)S(\mathbf{r}_2,\mathbf{r}_1)\\ & =-2\sum_{i,j}\psi_i(\mathbf{r}_0)\frac{(\psi_i\,|\,V_{p\text{-}h}\,|\,\phi_j)}{\varepsilon_i+\hbar\omega_j}\phi_j(\mathbf{r}_1) .\end{aligned} \tag{A14}$$

[*] On leave from the Department of Theoretical Physics, University of Oulu, Linnanmaa, SF-90570 Oulu 57, Finland.

[1] A. Dahm and L. Vinen, Phys. Today **40**(2), 43 (1987).

[2] H. J. Lauter, H. Godfrin, C. Tiby, H. Wiechert, and P. E. Obermayer, Surf. Sci. **125**, 265 (1983).

[3] H. J. Lauter, H. Godfrin, and H. Wiechert, in *Proceedings of the Second International Conference on Phonon Physics, Singapore, 1985* edited by J. Kollàr, N. Kroo, M. Meynhard, and T. Siklos (World-Scientific, Singapore, 1985), p. 842.

[4] J. D. Maynard and M. H. W. Chan, Physica B&C **109-110B**, 2090 (1982).

[5] J. A. Roth, G. J. Jelatis, and J. D. Maynard, Phys. Rev. Lett. **44**, 333 (1980).

[6] M. A. Paalanen and Y. Iye, Phys. Rev. Lett. **55**, 1761 (1985).

[7] D. Cieslikowski, A. J. Dahm, and P. Leiderer, Phys. Rev. Lett. **58**, 1751 (1987).

[8] J. G. Dash and M. Schick, in *Physics of Liquid and Solid Helium,* edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1976) Vol. 2.

[9] M. Bretz, in *Monolayer and Submonolayer Helium Films,* edited by J. G. Daunt and E. Lerner (Plenum, New York, 1973), p. 11.

[10] D. O. Edwards and W. F. Saam, in *Progress in Low Temperature Physics,* edited by D. F. Brewer (North-Holland, New York, 1978), Vol. 7A, p. 282.

[11] M. J. DiPirro and F. M. Gasparini, Phys. Rev. Lett. **44**, 269 (1980); B. Bhattacharyya and F. M. Gasparini, *ibid.* **49**, 919 (1982).

[12] F. M. Gasparini, B. Bhattacharyya and M. J. DiPirro, Phys. Rev. B **29**, 4921 (1984).

[13] D. S. Sherill and D. O. Edwards, Phys. Rev. B **31**, 1338 (1985).

[14] I. F. Silvera and J. T. M. Walraven, Phys. Rev. Lett. **44**, 164 (1980).

[15] H. M. Guo, D. O. Edwards, E. Sarwinski, and J. T. Tough, Phys. Rev. Lett. **27**, 1259 (1971).

[16] K. K. Mon and W. F. Saam, Phys. Rev. B **23**, 5824 (1981).

[17] B. Bhattacharyya, M. J. DiPirro, and F. M. Gasparini, Phys. Rev. B **31**, 2719 (1985).

[18] Y. M. Shih and C.-W. Woo, Phys. Rev. Lett. **30**, 478 (1973).

[19] R. A. Guyer, M. D. Miller, and J. Yaple, Phys. Rev. B **25**, 4570 (1982).

[20] W. F. Saam, Phys. Rev. A **4**, 1278 (1971).

[21] C. C. Chang and M. Cohen, Phys. Rev. B **8**, 3131 (1973).

[22] R. A. Guyer and M. D. Miller, Phys. Rev. Lett. **47**, 349 (1981).

[23] J. Owen, Phys. Rev. Lett. **47**, 586 (1981).

[24] A. Kallio, P. Pietiläinen, and L. J. Lantto, Phys. Scr. **25**, 943 (1982).

[25] P. Pietiläinen and A. Kallio, Phys. Rev. B **27**, 224 (1983).

[26] K. E. Kürten and M. L. Ristig, Phys. Rev. B **27**, 5479 (1983).

[27] J. Lekner, Philos. Mag. **22**, 669 (1970).

[28] E. Krotscheck, Phys. Rev. B **32**, 5713 (1985).

[29] I. B. Mantz and D. O. Edwards, Phys. Rev. B **20**, 4518 (1979).

[30] E. Feenberg, *Theory of Quantum Liquids,* (Academic, New York, 1969).

[31] C. E. Campbell, Phys. Lett. **44A**, 471 (1973).

[32] C. C. Chang and C. E. Campbell, Phys. Rev. B **15**, 4238 (1977).

[33] E. Krotscheck, Phys. Rev. B **33**, 3158 (1986).

[34] Eq. (5.18b) of Ref. 28 should contain a factor $\rho_1(\mathbf{r}_3)$, and Eqs. (5.19) should have a minus sign in front of the second integral. The correct form has been used for the numerical evaluation of the density matrix presented there, c.f., Eqs. (5.23).

[35] J. L. Epstein and E. Krotscheck, Phys. Rev. B **37**, 1666 (1988).

[36] E. Krotscheck, S. Stringari, and J. Treiner, Phys. Rev. B **35**, 4754 (1987).

[37] R. A. Aziz, V. P. S. Nain, J. C. Carley, W. L. Taylor, and G. T. McConville, J. Chem. Phys. **70**, 4330 (1979).

[38] F. Ricca, Nuovo Cimento Suppl. **5**, 339 (1967).

[39] M. W. Cole, D. R. Frankl, and D. L. Goodstein, Rev. Mod. Phys. **53**, 199 (1981).

[40] J. P. Toennies, W. Welz, and G. Wolf, Chem. Phys. Lett. **44**, 5 (1986).

[41] L. W. Bruch, Surf. Sci. **125**, 194 (1983).

[42] M. Cole (private communication).

[43] F. Dalfovo and S. Stringari, Phys. Scr. (to be published).

[44] S. Stringari and J. Treiner, J. Chem. Phys. (to be published).

[45] G. Ji and M. Wortis, Phys. Rev. B **34**, 7704 (1986).

[46] T. B. Davison and E. Feenberg, Phys. Rev. **178**, 306 (1969).

[47] B. L. Friman and E. Krotscheck, Phys. Rev. Lett. **49**, 1705 (1982).

[48] E. Krotscheck and R. A. Smith, Phys. Rev. B **27**, 4222 (1983).

[49] R. P. Feynman and M. Cohen, Phys. Rev. **102**, 1189 (1956).

[50] J. Owen, Phys. Rev. B **23**, 5815 (1981).

[51] J. Owen, Phys. Rev. Lett. **47**, 586 (1981).

[52] B. K. Bhattacharyya and F. M. Gasparini, in *Proceedings of the Seventeenth International Conference on Low Temperature Physics, LT-17,* edited by U. Eckern, A. Schmid, W. Weber, and H. Wühl (American Elsevier, New York, 1984), p. 1237.

[53] P. Leiderer, W. Ebner, and V. B. Shikin, Surf. Sci. **113**, 405 (1982).

[54] M. W. Cole, Phys. Rev. B **2**, 4239 (1970).

[55] L. M. Sander, Phys. Rev. B **11**, 4350 (1975).

[56] S. A. Jackson and P. M. Platzman, Phys. Rev. B **24**, 499 (1979); Surf. Sci. **113**, 401 (1982).

[57] O. Hipolito, G. A. Farias, and N. Stuart, Surf. Sci. **113**, 394 (1982).