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Theory of atomic scattering at the free surface of liquid ^{4}He

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The scattering of free helium atoms at the surface of liquid ^{4}He at zero temperature is discussed in terms of the Feynman variational method. If the scattered atom is distinguishable from those in the liquid target, as is true for the scattering of ^{3}He, the problem reduces to the motion of a single particle in an effective potential. Above the surface the effective potential is the same as the real van der Waals potential and, in the surface and below, it is related to the density and kinetic-energy distribution in the liquid ground state. If the theory is applied to the scattering of ^{4}He, neglecting the indistinguishability of the scattered atom, an excellent fit to the experimental reflection coefficient is obtained. When the trial wave function is fully symmetrized to calculate the reflection coefficient for ^{4}He more realistically, the theory describes the production of a single excitation (phonon) from each absorbed atom. The resulting reflection coefficient disagrees with experiment, predicting total reflection at certain critical angles. Even when multiple production of low-energy phonons is considered, total reflection will still occur. It follows that the multiple production of some other type of excitation, in particular low-energy ripplons, must be a dominant process in agreement with the calculations of Echenique and Pendry. It seems that the simple unsymmetrized theory fits the data because the reflection coefficient is mainly determined by the static van der Waals potential outside the liquid where the effects of symmetry, inelastic scattering, etc., are negligible. An atom which penetrates below this region is then effectively lost as far as the original beam is concerned because it begins to produce low-energy ripplons and is incoherently scattered. The problem of determining the density profile at the liquid surface from the experimental scattering data is briefly considered.

I. INTRODUCTION

In this paper we discuss the theory of the scattering of helium atoms (mainly ^{4}He) at the free surface of liquid ^{4}He. The liquid ^{4}He target is supposed to be at such a low temperature that it may be treated as being in its ground state. We are primarily interested in $R(k,\theta)$, the probability of elastic scattering, where the incident ^{4}He atom is specularly reflected without change in its kinetic energy $\hbar^2k^2/2m$ and with an angle of reflection equal to its angle of incidence θ (see Fig. 1). Other possibilities are: inelastic scattering, with probability $D(k,\theta)$, where the incident atom rebounds from the liquid surface but with loss of energy to the liquid; and absorption, with probability $f(k,\theta)$, where the kinetic energy of the atom $\hbar^2k^2/2m$ plus the binding energy is completely converted into excitations of the liquid. The excitations which may be produced include quantized capillary waves[1] (ripplons) as well as phonons. If the incident atom is ^{3}He, the energy of the resulting ^{3}He quasiparticle must be taken into account. Obviously

$$R(k,\theta)+D(k,\theta)+f(k,\theta)=1\quad .$$

A satisfactory theory of helium scattering should explain the results of recent experiments,[2-4] which measured $R(k,\theta)$ and $D(k,\theta)$ for ^{4}He and ^{3}He, and the distribution of excitations produced by the absorption of a ^{4}He beam. In addition, it would be very desirable to obtain, if possible, the density profile of the liquid from the experimental measurements. The density profile is the dependence of ρ, the number density in the ground state, on height z as one passes

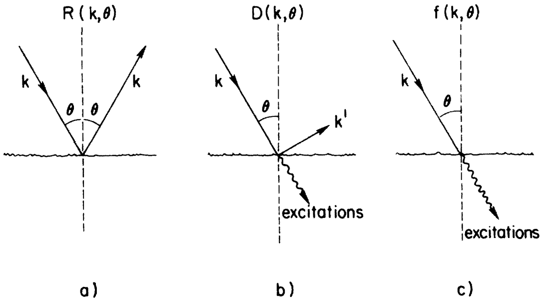

FIG. 1. Three possible fates for a free ^{4}He atom striking the surface of liquid ^{4}He and their probabilities: (a) elastic scattering (specular reflection), (b) inelastic scattering, converting some of the atom's kinetic energy into excitations (phonons, ripplons), (c) absorption, generating excitations whose total energy is equal to the kinetic energy plus the binding energy of the atom.

from the bulk liquid to the vacuum through the surface region. There have been numerous theoretical calculations[5-15] of $\rho(z)$ in the ground state of liquid ^{4}He but no measurements. One of the most interesting questions connected with $\rho(z)$ is whether there is a "peak" or maximum in the density at the surface[15] or even a series of oscillations.[8] Some calculations,[9-14] however, give a smooth monotonically decreasing $\rho(z)$, but with little agreement as to the precise width or shape of the profile, except that the width is of the order of a few angstroms. A review of these theories and of the scattering experiments will soon be published.[16]

It has been suggested[10,15] that the density profile might be measured using other types of scattering experiments—neutron, low-energy electron, or optical. It is easy to show that the elastic scattering probability $R(k,\theta)$ for neutrons is directly related to the Fourier transform of $\rho(z)$. Since it is interesting to compare elastic scattering of neutrons with that of helium atoms we have discussed this case in the Appendix. However, the intensity of elastic neutron scattering at the surface is so weak that this is not a practicable method of obtaining $\rho(z)$ unless the scattering from about 10^7 parallel surfaces can be combined in some way. The possibility of obtaining the profile by electron or optical scattering remains an open question.

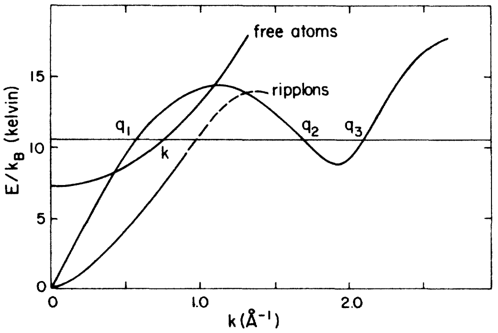

FIG. 2. Energy-momentum relation for free ^{4}He atoms compared to other excitations in liquid ^{4}He. A free atom with total energy greater than the roton energy, i.e., with momentum $\hbar k$ greater than ~0.5 Å^{-1}, can, in principle, generate a phonon of momentum $\hbar q_1$ or one of the rotons with momentum $\hbar q_2$ or $\hbar q_3$. The production of a single ripplon is impossible because of the conservation of transverse momentum, $\hbar k_x$. The ripplon spectrum is from Ref. 32.

This paper does not include a description of earlier theoretical and experimental work which preceded the measurements of $R(k,\theta)$; this may be found in Refs. 2 and 16, and in our earlier experimental paper.[17] However, we must mention an important theoretical suggestion which we shall call the "Anderson-Widom assumption,"[18,19] that the dominant process in the scattering or evaporation of ^{4}He atoms is a single-particle process—the conversion of an atom into a single high-energy phonon (or roton) or vice versa. This hypothesis would lead to an "edge" or "dip" in $R(k,\theta)$ at the roton threshold $k = 0.50$ Å^{-1} where an incident atom has enough energy $\hbar^2 k^2/2m + L_0 = \Delta$ to produce an excitation at the roton minimum (see Fig. 2). Here L_0 is the binding energy or latent heat of ^{4}He at 0 K ($L_0/k_B = 7.16$ K). The most recent calculations on the dip have been performed by Caroli *et al.*,[20] but without reaching a quantitative conclusion as to its size or shape. Since there is no dip or edge in the experimental data it was concluded in Ref. 2 that the production of multiple excitations is a dominant process. In Sec. IV of this paper we describe an explicit calculation of the consequences of the Anderson-Widom assumption that single-particle processes are the only ones of importance. The results are quite different from the experimental data. We also point out a qualitative feature of the assumption which was overlooked in previous work. This is the prediction of total external reflection [i.e., $R(k,\theta) = 1$] for certain values of k and θ. This happens when there is no single excitation which has the same energy and transverse momentum as the incident atom. It is not observed experimentally, again showing that multiple-particle processes must be considered.

The Anderson-Widom assumption was introduced in connection with the theory of evaporation from liquid helium. We have discussed the connection between evaporation and scattering in Refs. 2 and 21 in terms of detailed balance. Very briefly, the experimental measurements of $R(k,\theta)$ and $D(k,\theta)$ show that the momentum distribution of atoms evaporated from the liquid *in equilibrium* is extremely close to Maxwellian, and the accommodation coefficient is therefore almost indistinguishable from unity.[21] In some experimental situations, such as that described in Ref. 17 where the angular distribution of evaporated atoms from a thick film was not isotropic and therefore not Maxwellian, the liquid was not in equilibrium with the vapor, and the distribution of excitations in the liquid was not an equilibrium distribution. There has been little theoretical analysis of this type of experiment but at the present time it seem unlikely that evaporation measurements, as opposed to scattering experiments, can give much information about the surface of the liquid, unless the distribution of excitations incident on the surface does not correspond to equilibrium and is experimentally determined. An interesting experiment of this type has been performed by Balibar.[22] He has observed that rotons incident on the surface from below do, in fact, give rise to evaporated atoms.

At the present time it seems that the problem of atomic scattering is fairly well understood, at least in a semiquantitative way, on the basis of the theory described by Echenique and Pendry[23] or using the approach presented here. Both methods lead to the same general conclusion: that $R(k,\theta)$ is determined by the van der Waals potential in the vacuum outside the liquid region (i.e., several Å from the surface and above). Nearer the surface, the atoms are completely absorbed as a result of the production of multiple low-energy excitations, particulary ripplons. The problem of determining $\rho(z)$ from $R(k,\theta)$ is reduced to (a) fitting the van der Waals potential $V(z)$ in the "vacuum region" to the data and then (b) finding the distribution of matter $\rho(z)$ in the "liquid region" which gives rise to $V(z)$, using the known interatomic potential. With a rather rough fit to the data, Echenique and Pendry estimated the width of the density profile to be about 5 Å. In the present work we have an excellent fit to the data but the "effective" potential $U(z)$ which we have determined is the same as the real van der Waals potential $V(z)$ only above a certain ill-defined distance from the surface, and it has not been possible to obtain a unique density profile from the result. The principal objective of this paper is to show, explicitly, the consequences of the Anderson-Widom assumption and to demonstrate, by a method different from that of Echenique and Pendry, that ripplon production is a dominant effect. A brief account of some of the theory in Sec. III has already appeared.[21] In Sec. II we summarize the results of the scattering experiments before describing the calculations in Secs. III and IV.

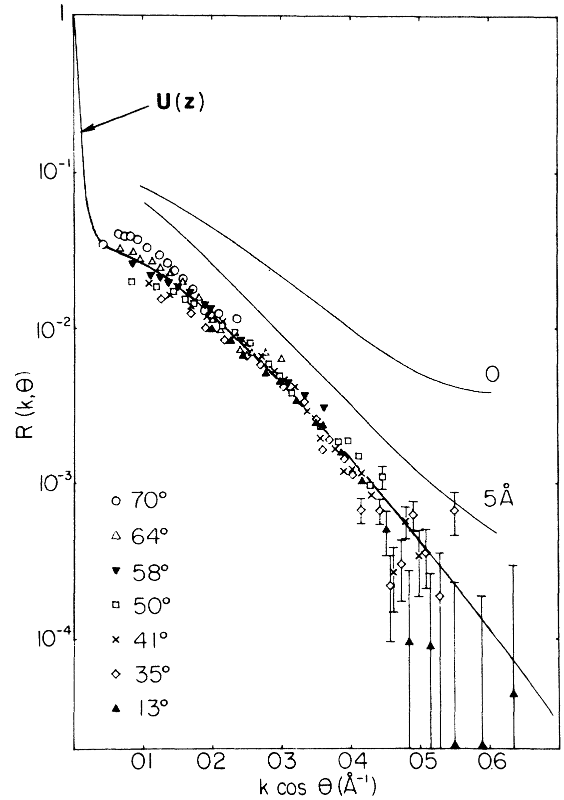

FIG. 3. Probability of elastic scattering $R(k,\theta)$ for a ^{4}He atom striking the free surface, as a function of the perpendicular component of its momentum, $\hbar k\cos\theta$. The points are experimental data from Ref. 2, for the angles of incidence shown. The theoretical cuves labeled 0 and 5 Å are from Echenique and Pendry (Ref. 23), for a step density profile at the surface ("0") and for a linear profile of width 5 Å. The curve labeled $U(z)$ has been fitted to the data using the model described in Sec. III.

II. SUMMARY OF EXPERIMENTAL RESULTS

The results of the atomic scattering experiments[2,4,17] for ^{4}He incident on liquid ^{4}He can be summarized as follows:

(a) Within experimental error, the probability of elastic scattering $R(k,\theta)$ does not depend on the transverse component of the momentum of the incident atoms but only on the vertical component, i.e., $R(k,\theta)=R(k\cos\theta)$, as shown in Fig. 3.

(b) The probability of inelastic scattering is zero within experimental error, $D(k,\theta)<\sim 2\times 10^{-3}$.

(c) There is no discernible dip or edge in $R(k,\theta)$ at the roton threshold, although rotons are produced at higher energies.[4]

(d) Within the experimental range of $k\cos\theta$ (from 0.05 to 0.5 Å^{-1}) $R(k,\theta)$ is very small so that the probability of absorption $f(k,\theta)\simeq 1$. Further, $R(k,\theta)$ does not seem to tend to unity as $k\cos\theta\rightarrow 0$, but to some limit near 0.05.

(e) The results in (a) to (d) apply to the scattering by the ground state of pure liquid ^{4}He. This was shown by the absence of any temperature dependence between 0.03 and 0.12 K, demonstrating that, in this range, thermally-excited ripplons or phonons have no effect. (The range of temperature has now been extended up to 0.25 K.[24]) Adsorbed ^{3}He on the ^{4}He surface produces measurable inelastic scattering but, at least for low concentrations of ^{3}He, it has little effect on the values of $R(k,\theta)$.[3,17,24]

The observation in (d), that $R(k,\theta)$ does not seem to tend towards unity as k becomes small, was satisfactorily explained[2] by the effect of the attractive van der Waals potential outside the liquid, which, to a good approximation, varies as $1/z^3$. The Schrödinger equation for an atom in this region can be reduced to

$$\frac{\partial^2\psi}{\partial z^2}+\left[k_z^2+\frac{\lambda}{z^3}\right]\psi=0\ , \tag{1}$$

where $k_z=k\cos\theta$ and, for the van der Waals potential between a ^{4}He atom and liquid ^{4}He, $\lambda=20$ Å. Rewriting this in reduced form with $\xi=k_z z$ gives

$$\frac{\partial^2\psi}{\partial\xi^2}+\left[1+\frac{k_z\lambda}{\xi^3}\right]\psi=0\ . \tag{2}$$

The long-wavelength limit will not be reached until $k_z\lambda << 1$. Then, when $k_z\lambda \rightarrow 0$, the potential will act like a step function, so that $R(k,\theta)$ should approach unity when k is small compared to $1/\lambda$, that is when $k_z << 0.05$ Å^{-1}. This explanation is confirmed by the more detailed calculations described below.

The observations[3] of the scattering of ^{3}He atoms at the surface of ^{4}He, although limited in extent and in accuracy, confirm that $R(k,\theta)$ is mainly determined by the product $k_z\lambda$, at least for $k_z \sim 0.1$ Å^{-1}. For ^{3}He, due to the lower mass, $\lambda = 15$ Å. Assuming, once again, that the motion of the scattered particle in the static $1/z^3$ van der Waals field outside the liquid is governed by (1) and (2), we deduce that the reflection coefficient for ^{3}He is related to that for ^{4}He by[3]

$$R_3(k_z) = R_4(\tfrac{4}{3}k_z) \quad . \tag{3}$$

Equation (3) is confirmed, within experimental error, by the data.

III. SIMPLE MODEL

In this section we describe a simple model for scattering based on the Feynman variational method[25] in which we treat the scattered atom as distinguishable from those in the liquid-^{4}He target, i.e., the variational wave function is not symmetrized with respect to the scattered atom. If the scattered atom is not reflected elastically, it becomes an impurity "quasiparticle" in the liquid. The possibility of ripplon or phonon production is not included. The model gives a very satisfactory fit to the data for ^{4}He and it serves as an introduction and basis for the symmetrized theory in Sec. IV. The model was first described in Ref. 21 but no details of the fit to the experimental data were given there.

The Feynman trial wave function of a state in which one of N ^{4}He atoms has been replaced by an "impurity" He atom is $\Psi = f(\vec{r}_1)\Phi(\vec{r}_1, \ldots, \vec{r}_N)$ where Φ is the ground state for N ^{4}He atoms; Φ is real and positive and $H\Phi = 0$, where H is the Hamiltonian for pure ^{4}He, and we are now measuring energies from the ^{4}He ground state energy. (The ground-state energy for N ^{4}He atoms is $-NL_0$ plus the surface energy compared to the vacuum.) If the impurity has the same interatomic potential but a mass different from ^{4}He, $m_1 \neq m$, the Hamiltonian becomes

$$H' = (-\hbar^2/2m_1 + \hbar^2/2m)\nabla_1^2 + H \quad .$$

When the expectation value of the energy

$$E = \int \Psi^* H' \Psi \, d\vec{r}_1 \cdots d\vec{r}_N \times \left(\int \Psi^* \Psi \, d\vec{r}_1 \cdots d\vec{r}_N \right)^{-1} \tag{4}$$

is minimized, Feynman showed that, for a state with momentum $\hbar\vec{K}$ in bulk helium, $f(\vec{r}_1) = e^{i\vec{K}\,\vec{r}_1}$ The energy is then

$$E = \hbar^2 K^2/2m_1 + (m/m_1 - 1)\nu_0/\rho_0 \quad ,$$

where ν_0/ρ_0 is the kinetic energy per atom in the bulk ^{4}He ground state. When the theory is applied to helium with a free surface, it is convenient to write

$$f(\vec{r}_1) = \psi(\vec{r}_1)/[\rho(\vec{r}_1)/\rho_0]^{1/2} \quad ,$$

where $\rho(\vec{r}_1)$ is the number density in the ground state Φ,

$$\rho(\vec{r}_1) = N \int \Phi^2 \, d\vec{r}_2 \cdots d\vec{r}_N \quad , \tag{5}$$

and ρ_0 is the number density in the bulk liquid. The probability density for atom 1 in the state Ψ is then $|\psi(\vec{r}_1)|^2$ apart from a normalization constant. Minimization of the energy in this case gives

$$\nabla_1^2\psi(\vec{r}_1) + [2m_1E/\hbar^2 - U(\vec{r}_1)]\psi(\vec{r}_1) = 0 \quad , \tag{6}$$

i.e., a single-particle Schrödinger equation with an effective potential $(\hbar^2/2m_1)U(\vec{r})$ which is related to the particle density $\rho(\vec{r})$ and kinetic energy density $\nu(\vec{r})$ in the ground state Φ. This result, which was derived by Lekner,[26] has been applied with considerable success[9,11,13] to the calculation of the binding energy of ^{3}He to the surface of ^{4}He; the potential function $U(\vec{r})$ for ^{3}He has a minimum at the surface which is strong enough to give a bound state.

If we assume that the "impurity" is a scattered ^{4}He atom so that $m_1 = m$, the effective potential is given by

$$\begin{aligned} U(\vec{r}) &= a''/a \quad , \\ a(\vec{r}) &\equiv [\rho(\vec{r})/\rho_0]^{1/2} \quad , \\ a''(\vec{r}) &= \nabla^2 a(\vec{r}) = \frac{d^2a}{dz^2} \quad . \end{aligned} \tag{7}$$

Since $\psi^*\psi$ is the probability density for the scattered atom and ψ obeys a single-particle Schrödinger equation, the current of scattered atoms is conserved and the problem is reduced to finding the single-particle reflection coefficient for the one-dimensional potential function $U(\vec{r}) = U(z) = a''/a$. The probability for elastic scattering is thus directly related to the density profile of the liquid $\rho(z) = [a(z)]^2\rho_0$.

The model has two features which agree very well with the experimental observations: (a) The predicted reflection coefficient only depends on the perpendicular component of the incident momentum $\hbar k \cos\theta$. (b) There is no inelastic scattering, only specular reflection or absorption. It is also clear that the theory can be applied directly to ^{3}He scattering from ^{4}He; in this case the effective potential would involve the distribution of kinetic energy $\nu(\vec{r})$ in the ground state as well as the density profile $\rho(\vec{r})$. The reflection coefficient for ^{3}He has not yet been calculated by this method.

In calculating the reflection coefficient, the proper asymptotic behavior of $a(z)$ outside the liquid must be taken into account. Far above the surface, where the liquid density decreases exponentially with z, the effective potential must be identical to the real, van der Waals potential. So for the ^{4}He potential we find

$$U \to \beta^2 - \lambda/z^3 \ , \tag{8}$$

which leads to

$$a \to \exp[-(\beta z + \text{const} + \lambda/4\beta z^2 + \cdots)] \ , \tag{9}$$

where $\hbar^2\beta^2/2m = L_0$, and $\beta = 1.087$ Å^{-1}. The fact that the density $\rho(z) = a^2\rho_0$ decreases as $\exp(-2\beta z)$ for large z was pointed out by Regge[8] and by Saam.[9] In writing Eq. (8) we have assumed that the leading term in the expansion in $1/z$ of the van der Waals potential outside the liquid is $-\hbar^2\lambda/2mz^3$. This follows if the leading term in the interatomic potential is proportional to $1/r^6$. In fact, of course, at distances r, larger than a few Å, retardation effects very gradually change this to $1/r^7$, which is the asymptotic form at distances which are large compared to several hundred Å.[27] Although an accurate interpolation formula between $1/r^6$ and $1/r^7$ is available, we have assumed that retardation plays a small role since the potential is so weak at large distances. Another problem is the precise value of λ. The value we have assumed ($\lambda = 20$ Å) is smaller than the $1/r^6$ term in the Lennard-Jones empirical potential but it is slightly larger than the theoretical value. See Sec. V for further discussion of both these points.

A simple formula which interpolates between the asymptotic behavior far from the liquid (8) and (9), and that deep inside, where $a \to 1$ and $U \to 0$ is

$$\begin{aligned} a(z) &= [\exp p(z) + 1]^{-1} \\ p(z) &= \beta z - g_1 + \lambda/4\beta(z^2 + g_2) \ , \end{aligned} \tag{10}$$

where g_1 and g_2 are adjustable constants, and g_2 must be positive to avoid singular behavior in $p(z)$. Then

$$U = a''/a = (1-a)[p'^2(1-2a) - p''] \ , \tag{11}$$

where p' and p'' are the first and second derivatives of p with respect to z. We have solved the Schrödinger Eq. (6) numerically using the Numerov[28] method with the potential specified by Eqs. (10) and (11). To calculate the reflection coefficient, we write $m_1 = m$ and $\psi = e^{ik_x x}\zeta(z)$ with $k_x = k\sin\theta$, $k_z = k\cos\theta$ in Eq. (6), yielding

$$\frac{d^2\zeta}{dz^2} + [k_z^2 + \beta^2 - U(z)]\zeta(z) = 0 \ . \tag{12}$$

Starting with z several angstroms below the surface where $U = 0$ and $\zeta(z) = \exp[-i(k_z^2 + \beta^2)^{1/2} z]$ we integrate out to $z \sim 200$ Å where $U = \beta^2$ and $\zeta(z) = Ae^{-ik_z z} + Be^{ik_z z}$. The reflection coefficient is then $R(k_z) = |B/A|^2$. The reflection coefficient fits the experimental data very well for

$$g_1 = 2.5, \quad g_2 = 8.5 \ \text{Å}^2 \ . \tag{13}$$

Neither parameter must be adjusted very carefully to get an excellent fit. With these values substituted in Eqs. (10) and (11) we shall refer to $U(z)$ and $[a(z)]^2$ as the "model potential" and "model profile." The fit to the experimental data is shown in Fig. 3. It is certainly within the experimental data. However, the model profile (shown in Fig. 4) cannot be taken seriously as a determination of the real density profile of liquid helium. Apart from the lack of symmetry in the variational wave function there is another reason: The reflection coefficient is not sensitive to the potential $U(z)$ in the region where $\rho(z)$ is substantially larger than zero. This is shown in Fig. 5 where we have plotted the reflection coefficient calculated from the model potential $U(z)$ and from a modification $U(z) + \frac{1}{2}\beta^2\rho/\rho_0$. The modification differs from $U(z)$ only where $a^2 = \rho/\rho_0$ is substantially larger than zero. The figure demonstrates that the reflection coefficient is not appreciably changed by the modification. This means that the fit to the data is not sensitive to $U(z)$ in the region where $\rho(z)$ is different from zero, i.e., in the "liquid region" where we would like to determine the profile.

The reason for this insensitivity can be better understood by studying the effect of another modification to the potential; we add an imaginary term to U, proportional to the density of the liquid, $U' = U - ia^2\gamma^2$. The result of using $\gamma = 1.5$ Å^{-1} is shown in Fig. 5. The effect of the imaginary part of the potential is to produce a strong absorption inside the liquid (the current of the scattered atom is not conserved). Deep inside the liquid where $a^2 = 1$ and

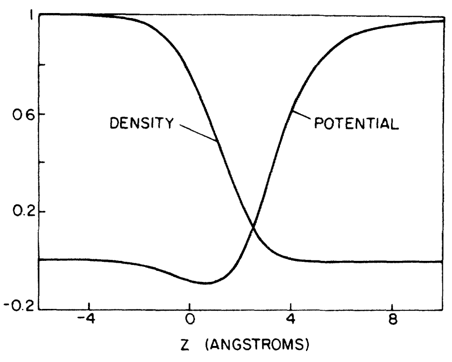

FIG. 4. "Model" potential and density profile (see Sec. III of the text) in dimensionless units plotted against the height z in angstrom units. The quantities plotted are $\rho(z)/\rho_0$ and $U(z)/\beta^2$. As discussed in the text, the density profile and the potential probably have no direct physical interpretation for z less than a few Å.

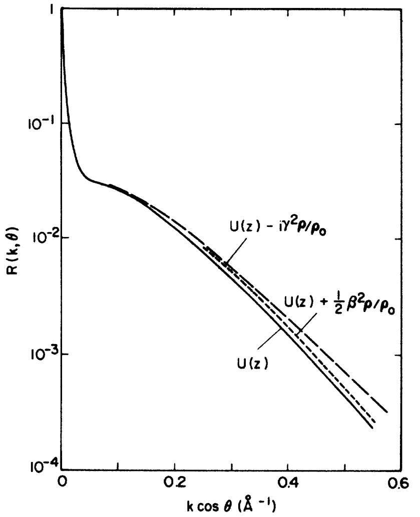

FIG. 5. Elastic-scattering probability $R(k,\theta)$ for ^{4}He as a function of the perpendicular component of the momentum, calculated from the model potential $U(z)$, and from the model potential with modifications inside the liquid region where ρ/ρ_0 is nonzero. For the potential with the imaginary term $U(z) - i\gamma\rho/\rho_0$ there is very strong absorption in the liquid, corresponding to a free path of less than 2 Å at the bulk density. For the potential with the real term, $U(z) + \frac{1}{2}\beta^2\rho/\rho_0$, the binding energy of the incident particle has been halved inside the liquid.

$U=0$, the wave function of the scattered atom is of the form $e^{\alpha z}e^{i\vec{q}\cdot\vec{r}}$, so that the probability density $|\psi|^2$ (and the current) decreases as z decreases with a free path $1/2\alpha$. With $\gamma = 1.5$ Å, the free path in the bulk $1/2\alpha$ is approximately 2 Å—less than the diameter of an atom—this corresponds to the curve shown in Fig. 5. Even with this very strong absorption the reflection coefficient is not seriously affected. With $\gamma = 1$ Å^{-1} and $1/2\alpha \sim 2.5$ Å the effect is so small that it would hardly be visible in Fig. 5. This shows that, for an atom moving in the model potential which has reached the liquid region, the probability of reflection is negligibly small, and that R is determined by the potential further away, in the vacuum above the liquid. We have also tried the effect of modifications to U proportional to the amplitude function $a(z)$ rather than a^2. These modifications do have an appreciable effect on R showing that the reflection coefficient is sensitive to the effective potential where $a(z)$ is not negligible but $a^2(z)$ is. This is roughly in the region $z > 3.5$ Å for the model profile.

Although R is not sensitive to U in the liquid region, the profile $\rho(z)$ in the liquid can still be determined, in principle, from the effective potential $U(z)$ in the vacuum region. Sufficiently far from the liquid $U(z)$ must be identical to the real Van der Waals potential which is related to the overall distribution of matter in the liquid through the interatomic potential. In practice, however, it has proved to be quite difficult to determine $\rho(z)$ unambiguously by this method.

IV. SYMMETRIZED THEORY

The theory in Sec. III describes the absorption of an atom into the liquid in terms of the creation of a single "quasiparticle" in the liquid. This might be an accurate description for the scattering of ^{3}He but it is obviously incorrect when the incident beam is ^{4}He. (Since it does not include the production of ripplons or phonons it probably does not apply to ^{3}He either.) When the trial wave function is symmetrized, as in the present section, the theory describes the conversion of an incident atom into a single high-energy excitation (phonon) with the same energy and transverse momentum $\hbar k_x$. The possibility of inelastic scattering or the production of several excitations is not allowed. The theory, therefore, corresponds to what we have called the Anderson-Widom assumption, that only single-particle processes occur or are important. As we shall see, the results of the theory are in strong disagreement with experiment, particularly in the prediction of complete or almost complete reflection, $R(k,\theta) \sim 1$, for certain large k and θ, and the principal usefulness of the theory is to demonstrate conclusively that multiple production of excitations, and particularly ripplons, must be important.

The symmetrized Feynman wave function for an excited state of the liquid containing N ^{4}He atoms is

$$\Psi = \sum_{i=1}^{N} f(\vec{r}_i)\Phi(\vec{r}_1,\ldots,\vec{r}_N) \quad , \tag{14}$$

where Φ is the ground state and $H\Phi = 0$, with

$$H = -\frac{\hbar^2}{2m}\sum_i \nabla_i^2 + \sum_{i<j} v(r_{ij}) - E_0 \quad . \tag{15}$$

E_0 is the ground-state energy relative to the vacuum and $v(r)$ the interatomic potential. Feynman showed that, when the expectation value of the energy is minimized for the homogeneous liquid, $f(\vec{r}) = e^{i\vec{q}\cdot\vec{r}}$ and the state Ψ has momentum $\hbar\vec{q}$, mass current $\vec{j} = \rho_0 m\vec{v} = \rho_0(\hbar\vec{q}/N)$, and energy $E = \hbar^2 q^2/2mS(q)$. The structure factor $S(q)$ in the homogeneous ground state is related to the two-particle density $\rho(\vec{r}_1,\vec{r}_2)$ and the two-particle correlation function $g(r)$. In general

$$\begin{aligned}\rho(\vec{r}_1,\vec{r}_2) &\equiv \rho(\vec{r}_1)\rho(\vec{r}_2)g(\vec{r}_1,\vec{r}_2)\\ &= N(N-1)\int \Phi^2\, d\vec{r}_3 \cdots d\vec{r}_N \quad .\end{aligned} \tag{16}$$

For the homogeneous ground state, $\rho(\vec{r}_1) = \rho(\vec{r}_2) = \rho_0$ and $g(\vec{r}_1,\vec{r}_2) = g(|\vec{r}_1 - \vec{r}_2|)$ so that

$$\rho(\vec{r}_1,\vec{r}_2) = \rho_0^2 g(|\vec{r}_1 - \vec{r}_2|) \tag{17}$$

and

$$S(q)=1-\rho_0\int d\vec{r}\,e^{-i\vec{q}\cdot\vec{r}}[1-g(r)]\ . \tag{18}$$

Since, for low q, $S(q)=\hbar q/2mc$ (where c is the velocity of sound), the theory gives an accurate description of the phonon states on the linear branch of the helium energy spectrum (Fig. 2) where $E=\hbar cq$. At high q the roton minimum is predicted to be at the correct wave vector $q \sim 2$ Å^{-1} but the energy at the minimum is too large by a factor of two. Although more refined trial wave functions give better agreement with experiment in the roton region, our conclusions concerning the rotons will not require accurate quantitative calculation and we will confine ourselves to the original Feynman wave function, Eq. (14).

When the energy E is minimized for the inhomogeneous liquid with a free surface, the Euler-Lagrange equation is

$$\nabla_1(\rho_1\nabla_1 f_1)+\frac{2mE}{\hbar^2}f_1\rho_1 +\int d\vec{r}_2\left[\nabla_2(\rho_{12}\nabla_2 f_2)+\frac{2mE}{\hbar^2}f_2\rho_{12}\right]=0\ . \tag{19}$$

Here f_1 means $f(\vec{r}_1)$, $f_2=f(\vec{r}_2)$, the density

$$\rho_1=\rho(\vec{r}_1)=N\int \Phi^2\,d\vec{r}_2\cdots d\vec{r}_N$$

and the two-particle density function $\rho_{12}=\rho(\vec{r}_1,\vec{r}_2)$, given by Eq. (16), will no longer just depend on $|\vec{r}_1-\vec{r}_2|$ in the region of the surface. Writing $f(\vec{r})=\psi(\vec{r})/[\rho(\vec{r})/\rho_0]^{1/2}$ and dropping terms which are zero, Eq. (19) becomes

$$\nabla_1^2\psi_1+\left[\frac{2mE}{\hbar^2}-U_1\right]\psi_1=\frac{2mE}{\hbar^2}\int d\vec{r}_2\,(\rho_1\rho_2)^{1/2}c_{12}\psi_2\ . \tag{20}$$

In this equation $U_1=U(\vec{r}_1)$ is the potential function derived from the density profile $U=a''/a$ and $c_{12}=c(\vec{r}_1,\vec{r}_2)\equiv 1-g(\vec{r}_1,\vec{r}_2)$. The function c_{12} is 1 for $\vec{r}_1=\vec{r}_2$ and zero when $\vec{r}_1$ and $\vec{r}_2$ are far apart; in the bulk of the liquid it is $1-g(|\vec{r}_1-\vec{r}_2|)$. In general, c_{12} can be written $c_{12}=c_{12}(z_1,z_2,h)$ where z_1 and z_2 are the vertical components of $\vec{r}_1$ and $\vec{r}_2$ and h is the magnitude of the horizontal component of $\vec{r}_1-\vec{r}_2$.

Writing $\psi(\vec{r})$ as $e^{ik_x x}\zeta(z)$, and $a(z)\equiv[\rho(z)/\rho_0]^{1/2}$, Eq. (20) becomes

$$\frac{d^2\zeta(z_1)}{dz_1^2}+\left[\frac{2mE}{\hbar^2}-k_x^2-U(z_1)\right]\zeta(z_1) =2\pi\rho_0\frac{2mE}{\hbar^2}a(z_1)\int dz_2\,a(z_2)\zeta(z_2) \times\int h\,dh\,J_0(k_x h)c(z_1,z_2,h)\ . \tag{21}$$

This integro-differential equation and its equivalent Eq. (20), was first given by Saam.[9] It is identical to the Schrödinger equation (6) when $a(z_1)$ is negligibly small, i.e., when z_1 is outside the liquid. This means that the symmetrized theory is identical to the unsymmetrized theory in the vacuum region where both theories reduce to the motion of a single particle moving in the potential $U(z)$.

The solutions to the integro-differential equation (21) are of several types, depending on their asymptotic behavior for $z\to-\infty$ and $z\to\infty$. For a horizontal free surface near $z=0$, we can assume

$$\zeta(z)\to e^{-iq_z z},\quad z\to-\infty\ , \tag{22}$$

where q_z may be real and positive, or imaginary. For large z,

$$\zeta(z)\to Ae^{-ik_z z}+Be^{ik_z z},\quad z\to\infty\ , \tag{23}$$

where

$$2mE/\hbar^2=\beta^2+k_x^2+k_z^2=q^2/2mS(q) \tag{24}$$

and

$$q^2=q_z^2+k_x^2\ . \tag{25}$$

The solutions we are primarily interested in have q_z and k_z real. These correspond to states in which there is a phonon of wave vector $q=(q_z^2+k_x^2)^{1/2}$ in the bulk liquid or an evaporated atom of momentum $k=(k_x^2+k_y^2)^{1/2}$ in he vacuum region. States in which q_z is real but k_z is imaginary, so that the wave function decreases exponentially with z in the vacuum region, correspond to phonons which are incapable of single-particle conversion to an evaporated atom and which are totally reflected at the surface. Their energy is below the threshold $(\hbar^2/2m)(\beta^2+k_x^2)$ for the evaporation of an atom with transverse momentum hk_x.

There are also solutions to Eq. (20) or (21), which are localized in the surface region, i.e., both q_z and k_z are imaginary and the function $\zeta(z)$ decays exponentially for large negative z and large positive z. These correspond to the ripplon excitations. Solutions of this type have been sought by Chang and Cohen,[29] Miller,[30] and Woo[31] to determine the theoretical ripplon energy spectrum, i.e., the dependence of E on k_x for these states. The ripplon energy spectrum can also be determined at low k_x by classical or quantum hydrodynamics[32] and this is shown in relation to the phonon and free particle states in Fig. 2. In the context of the present theory the possibility of an incoming atom being absorbed and generating a single ripplon of the same energy and transverse momentum is clearly ruled out.

Finally, there are solutions to Eq. (21) which have k_z real but q_z imaginary so that the wave function decreases exponentially in the liquid region. These states correspond to atoms which are totally reflected, i.e., they have $R(k,\theta)=1$. Total reflection will occur when the atom cannot convert to a single excitation (phonon) of the same energy and conserve transverse momentum $\hbar k_x$. The conditions under which this will happen can be examined using Fig. 2. If we ignore the possibility of producing a roton for the moment, total reflection occurs when

$$q < k_x \ , \tag{26}$$

where q is the wave number of the phonon which has the same energy as the incoming atom of wave vector $\vec{k}$. Equation (26) implies that $q < k$ so that the energy must be above the intersection of the free atom and phonon curves in Fig. 2, i.e., k must be larger than 0.47 Å^{-1}. For any given angle θ, the critical value of k above which total reflection occurs k_c can then be calculated from the condition $q=k_x=k\sin\theta$ using the energy equation $\hbar cq=(\hbar^2/2m)(\beta^2+k^2)$. The result is

$$k_c=(mc\sin\theta/\hbar)-[(mc\sin\theta/h)^2-\beta^2]^{1/2} \ . \tag{27}$$

Values of k_c are marked on Fig. 6, which also shows the experimental $R(k,\theta)$ as a function of k.

When the energy is above the roton threshold (corresponding to $k > 0.5$ Å^{-1} for the experimental excitation curve) the atom can always convert into one of the two roton states with the same energy. However, even in this case the present theory indicates that the reflection coefficient should still be very high when k is above the critical value k_c. Above the roton threshold for a given energy E there will be three degenerate solutions of the integro-differential Eq. (21) corresponding to the three possible values of the excitation momentum q_1, q_2, and q_3 in Fig. 2. If $k > k_c$, of these three degenerate solutions, the value of q_z will be imaginary for the first phonon solution and real for the roton solutions 2 and 3. The incoming beam will be composed of three independent components of equal amplitude corresponding to the three solutions labeled by the quantum numbers q_1, q_2, and q_3. The first component will be reflected with probability R_1 equal to unity because of the imaginary value of q_{1z}. The total reflected current for the whole beam will be the sum of the currents for the three components since they correspond to different quantum numbers q_1, q_2, and q_3, and there will be no interference effects. This means that the reflection coefficient for the whole beam will be

$$R=\tfrac{1}{3}(R_1+R_2+R_3) \ . \tag{28}$$

When $k > k_c$, the reflection coefficient must be larger than $\frac{1}{3}$, $R(k,\theta) \geq \frac{1}{3}$, despite the fact that the two roton channels are open for absorption of the atom.

The preceding argument, which is qualitative and general, should hold despite the fact that the simple Feynman wave function does not give a good value for the roton energy. The same reasoning could be applied to more accurate trial functions which would give agreement with the experimental roton energy curve.

To calculate theoretical values of $R(k,\theta)$ below k_c we must solve Eq. (21). This requires knowledge of the correlation function $c(z_1,z_2,h)$ in the region of the surface. Shih and Woo[11] and Chang and Cohen[12,13] have proposed that $c(z_1,z_2,h)$ may be approximated by $c(\bar{\rho},|\vec{r}_1-\vec{r}_2|)$, a function depending only on the distance between the two points $\vec{r}_1$ and $\vec{r}_2$ and on the local density at some mean point between them: $\bar{\rho}=\rho(\bar{z})$. For instance, $\bar{\rho}$ can be chosen to be $[\rho(\vec{r}_1)(\vec{r}_2)]^{1/2}$ or $\frac{1}{2}[\rho(\vec{r}_1)+\rho(\vec{r}_2)]$. This approximation could have been incorporated into our numerical integration of Eq. (21) relatively easily, but would have lengthened the computation time. Instead we made the simplest possible approximation by writing

$$c(z_1,z_2,h)=c_0[(z_1-z_2)^2+h^2]^{1/2} \ ,$$

where $c_0(r)$ is the correlation function in the bulk liquid of density ρ_0. We further simplified the integral with respect to h in Eq. (21) by representing the experimental values of $c_0(r)$ by ten points, with a linear interpolation in r^2 between them. The resulting structure factor $S(q)$, and theoretical phonon spectrum $\hbar\omega(q)=\hbar^2q^2/2mS(q)$ are close to the experimental curves in the region of interest between ~0.4 and ~0.7 Å^{-1}. To complete Eq. (21) we also need $U(z)$ and $a(z)$; for these we used the model potential and profile described in Sec. III.

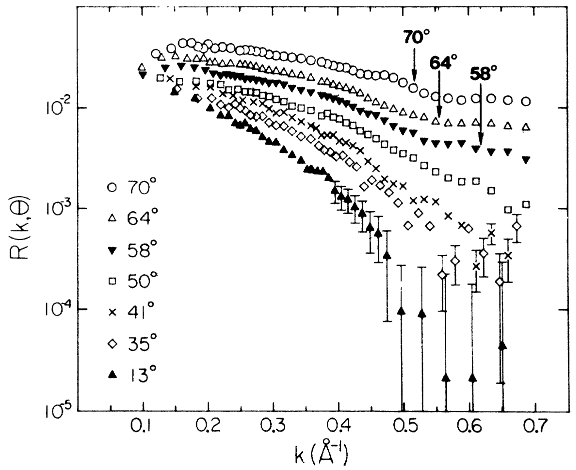

FIG. 6. Probability of elastic scattering $R(k,\theta)$ for free ^{4}He atoms striking the surface of liquid ^{4}He as a function of the incident momentum $\hbar k$. The points are experimental values from Ref. 2 at the angles of incidence shown. The arrows mark the theortical momenta k_c for angles of incidence of 70°, 64°, and 58° at which total reflection could occur (see text).

To find the reflection coefficient, $\zeta(z)$ was assumed to be $e^{-iq_z z}$ deep inside the liquid, i.e., for z large and negative. Then Eq. (21) was integrated numerically to $z \sim 200$ Å where $a(z)=0$, and $U(z)=\beta^2$, and $\zeta(z)$ has the form $Ae^{-ik_z z}+Be^{ik_z z}$ with $R=|B/A|^2$. The numerical integration was performed by writing the integral with respect to z_2 in terms of Simpson's rule, and the differential with respect to z_1 in terms of the Numerov expression, then solving the resulting set of linear equations by elimination. The interval of integration in z was 0.2 Å. For z larger than several angstroms, where the integral term is negligible, the differential equation was integrated by the Numerov method as in Sec. III. The final results for $R(k,\theta)$ are shown in Fig. 7.

Before discussing the numerical results we ought to comment on the physical interpretation of the "single-particle" function $\psi(\vec{r})$ and the corresponding trial function Ψ. In contrast with the situation where the scattered atom is distinguishable and the trial wave function is unsymmetrized, the current $\vec{j}$ evaluated for the symmetrized function Ψ is not conserved. This was checked by calculating the current from the numerical solution in the bulk liquid region and in the region very far above the surface where $z>200$ Å. In the bulk liquid region where $\psi(\vec{r})=e^{ik_x x-iq_z z}$ it can be shown that the expectation value of the mass current operator for the state Ψ is given by $C\vec{q}S(q)$ where C is a constant which depends on the normalization constant of the ground-state wave function. The z component of this current, which we call $\vec{j}^{\phi}$, is therefore

$$j_z^{\phi}=-Cq_zS(q), \quad z\to-\infty \quad . \tag{29}$$

In the vacuum, above the liquid, where

$$\psi(\vec{r})=e^{ik_x x}(Ae^{-ik_z z}+Be^{ik_z z}) \quad ,$$

the z component of the mass current is

$$\begin{aligned} j_z^{\rm atom} &= -Ck_z(|A|^2-|B|^2) \\ &= -Ck_z|A|^2(1-R); \quad z\to+\infty \quad . \end{aligned} \tag{30}$$

It was found that the numerical values of these currents agreed with the following simple physical picture: The net mass current of atoms approaching the surface and being absorbed is $-j_z^{\rm atom}$, therefore the number of atoms per unit area being absorbed is $-j_z^{\rm atom}/m$. Each absorbed atom produces a single phonon moving with group velocity $-(q_z/q)(d\omega/dq)$ in the z direction so that, if the number density of phonons is n^{ϕ},

$$n^{\phi}\frac{q_z}{q}\frac{d\omega}{dq}=j_z^{\rm atom}/m \quad . \tag{31}$$

The mass current or momentum density corresponding to this flux of phonons is $j_z^{\phi}=n^{\phi}(\hbar q_z)$ so that

$$j_z^{\phi}=\hbar n^{\phi}q_z=\frac{\hbar q}{m d\omega/dq}J_z^{\rm atom} \quad . \tag{32}$$

This relation between j_z^{ϕ} and $j_z^{\rm atom}$ was confirmed by the numerical values of $|A|^2$ and R when substituted in Eqs. (29) and (30). The phonon mass current j_z^{ϕ} therefore is just what one would expect but it is not equal to $j_z^{\rm atom}$. The discrepancy is made up by a supercurrent. Instead of using the stationary ground state Φ in constructing the variational wave function we should use

$$\Phi\exp\left[\frac{im}{\hbar}\vec{V}_s\cdot\sum_i\vec{r}_i\right] \quad ,$$

where the superfluid velocity $\vec{V}_s$ is in the z direction and makes up the difference between j_z^{ϕ} and $j_z^{\rm atom}$:

$$\rho_0 mV_s+j_z^{\phi}=j_z^{\rm atom} \quad . \tag{33}$$

The magnitude of V_s is microscopically small and the additional energy involved is negligible. The modification to the Euler-Lagrange equation is also negligible. In this way we conserve mass current and

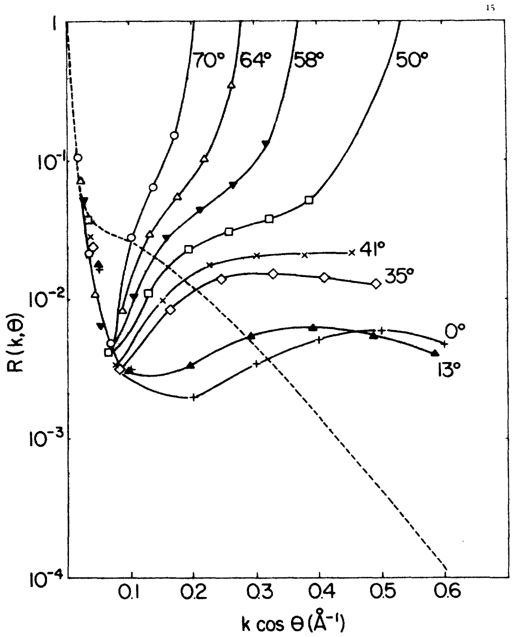

FIG. 7. Probability for elastic scattering $R(k,\theta)$ for free ^{4}He atoms striking the surface calculated from the symmetrized theory of Sec. IV. The full curves, which are labeled with the angle of incidence θ, have been drawn through the numerical results from the computer program, shown as the points. The dashed curve is the unsymmetrized theory, which is a very good fit to the experimental data. The symmetrized theory allows only the conversion of an atom into a single energetic phonon. It does not agree with the experiments, and it predicts total reflection, $R(k,\theta)=1$, at certain momenta and angles.

we agree with a conventional two-fluid picture in which the current of absorbed atoms $\vec{j}$ is converted into a normal current $\vec{j}^{\phi}$ due to the phonons and a super current $\vec{j}_s = \rho_0 m \vec{V}_s$.

We now return to consideration of the numerical values of $R(k,\theta)$ in this theory, shown in Fig. 7. For each angle of incidence we can distinguish three ranges of k_z. Initially for low k_z, below ~ 0.03 Å^{-1} the symmetrized and unsymmetrized theories are identical: in this range the reflection coefficient is insensitive to the integral on the right-hand side of Eq. (21). In the intermediate region, up to ~ 0.1 Å^{-1} depending on the angle of incidence, the results for different angles fall on the same curve, i.e., R depends only on k_z and not on k_x. However, R is quite different from the curve obtained from the unsymmetrized theory, and it is in poor agreement with experiment. In this range the integral in Eq. (21) has an important effect but the argument of the Bessel function J_0 in the integral is small enough that $J_0 \simeq 1$ and the integral does not depend on k_x. The poor numerical agreement with experiment in this range of k_z could perhaps be corrected by using a density-dependent correlation function and by a better choice of $U(z)$ and $a(z)$ in the liquid region. A serious attempt to adjust the potential to fit the data would have been quite costly in computer time, and probably quite useless, since the absence of total reflection in the data indicates that multiple excitation processes are important. Since the integral in Eq. (21) is proportional to $a(z_1)$ it has a negligible effect where this factor is negligible. According to the model profile $a(z)$ has fallen to approximately 0.01 at $z \simeq 6.4$ Å. Above this height the symmetrized and unsymmetrized theories are equivalent and so, above 6.4 Å, $U(z)$ should not require any correction.

At higher values of k_z, the value of k_x becomes important and the reflection coefficient rises; for the angles of incidence greater than 41° the reflection coefficient reaches unity as the critical momentum k_c for total reflection is reached. Since the theoretical roton energy in the Feynman theory is so large, we are not able to calculate the effects of roton production on the reflection coefficient in this model but, as we have already demonstrated in the discussion of (28), the prediction of a large reflection coefficient near or above the critical angle for total reflection will not be affected.

In principle, the symmetrized theory which we have described above could be elaborated to include multiple excitation processes. The trial wave function would contain products of single-particle functions

$$\Psi = \Phi \sum_{ij} \psi_1(\vec{r}_i)\psi_2(\vec{r}_j) \cdots \; .$$

The appropriate Euler-Lagrange equation would then be formidable. However, we can deduce an important fact about the excitations poduced by absorbed atoms without further calculation. If the only excitations produced were phonons we would still observe total reflection for $k > k_c$ below the roton threshold. This follows because k_x would still be larger than the combined momenta of all the phonons produced: $k_x > q_1 + q_2 + \cdots$. Since no trace of total reflection is observed in the experimental data we deduce that ripplon excitations must be important in dissipating the energy of an absorbed atom. Low-energy ripplons because of their $k_x^{3/2}$ spectrum can carry away an almost unlimited amount of transverse momentum for a given total energy. The conclusion that ripplon production has a dominant effect was also reached by Echenique and Pendry[23] by direct calculation.

V. DISCUSSION

The calculations in Sec. IV and those of Echenique and Pendry demonstrate that the production of multiple low-energy excitations, in particular ripplons, must be the fate of the majority of incident atoms, which are subsequently absorbed into the liquid. How then can one understand the success of the simple unsymmetrized theory (Sec. III) in fitting the reflection coefficient? Apart from the excellent quantitative agreement when the model potential is used, the unsymmetrized theory, which treats the incoming atom as moving in a one-dimensional effective potential, explains the lack of any dependence on the transverse momentum k_x and the absence of any measurable inelastic scattering.

The explanation is that the specular reflection coefficient is mainly determined by the static van der Waals potential outside the liquid where the effects of symmetry, inelastic scattering, etc., are negligible. An atom which penetrates beyond this region is then effectively lost as far as the original beam is concerned because, as shown by Echenique and Pendry, it begins to produce low-energy excitations and is incoherently scattered. The effective potential $U(z)$ in the liquid region is therefore just an approximate way of terminating the real van der Waals potential outside the liquid. As we have seen in Sec. III, the reflection coefficient is insensitive to $U(z)$ in the region where $\rho/\rho_0 = a^2$ is appreciable and has only a weak dependence where $a(z)$ is appreciably different from zero. In addition, a large imaginary component in the potential has a negligible effect where $a^2(z)$ is large and a small effect where $a(z)$ is appreciable. This shows that an incident atom which has penetrated this far towards the liquid has a negligible chance of being reflected. We can assume that the effective potential $U(z)$ has very little physical significance in the liquid region but it must be a fair representation of the real, static van der Waals potential in the vacuum region beyond some critical distance z_c. According to Echenique and Pendry, the critical distance from the liquid, at which ripplons begin to be produced, is approximately 5 Å. In their theory the calculation of the reflection coefficient reduces to the solution of a one-dimensional Schrödinger equation with a potential composed of the real van der Waals potential plus an imaginary term, proportional to $1/k_z z^{17/2}$, calculated from the rate of ripplon production.

Further evidence confirming this physical picture is provided by the measurements[4] of the angular distribution of excitations produced in the bulk of the liquid by an incoming ^{4}He beam. The experiments are consistent with the idea that an incident atom, which is absorbed by the liquid, loses some of its energy to low-energy ripplons as it approaches the surface, but the rest of its energy and all of its transverse momentum $\hbar k_x$ are given up in the creation of a single energetic phonon or a single roton if the remaining energy is sufficient. The hypothesis that the production of ripplons takes place without much loss of transverse momentum is consistent with the calculation of Echenique and Pendry in which the ripplon modes are standing waves, with zero momentum.

Returning now to the problem of finding the density profile from the data on the reflection coefficient, if we define the van der Waals potential in the vacuum outside the liquid, where $\rho(\vec{r}) \simeq 0$, as $\hbar^2 V(\vec{r})/2m$, then

$$\frac{\hbar^2 V(\vec{r})}{2m} = \int_{\rho(\vec{r}\,') > 0} v(|\vec{r}\,' - \vec{r}|)\rho(\vec{r}\,')\, d\vec{r}\,', \quad \rho(\vec{r}) \simeq 0 \ , \tag{34}$$

where $v(r)$ is the helium interatomic potential. The integral in (34) is taken over the region in $\vec{r}\,'$ where the liquid density is substantially different from zero. In the vacuum the model potential, measured with respect to the energy of an isolated atom, $\hbar^2/2m[U(z)-\beta^2]$, should be identical to the real van der Waals potential

$$U(z) - \beta^2 = V(z), \quad z > z_c \ . \tag{35}$$

In comparing their theory with the reflection coefficient data, Echenique and Pendry tried two $V(z)$ derived from two different density profiles. The first, $-\lambda/z^3$, corresponds to a step-function profile giving the result labeled "0" in Fig. 3. The second, $-\lambda z/(z^2-l^2)$, corresponds to a profile which falls linearly from the bulk density to zero over a width $2l$ of 5 Å. The resulting reflection coefficient, labeled "5 Å" in Fig. 3, is in better agreement with the data but the fit is still not quite satisfactory.

It is interesting to compare our effective potential to the $V(z)$ used by Echenique and Pendry, although it is difficult to do so unambiguously. This is because the origin of z is arbitrary for $U(z)$, since a displacement of the origin makes no difference to the calculated reflection coefficient. For a van der Waals potential derived from a profile using Eq. (34) it is logical to define $z=0$ at the dividing surface for the density profile, however, since the model profile has no physical significance, it does not make sense to do this for $U(z)$. We have tried choosing the origin of z so as to make $U(z)-\beta^2$ coincide with the Echenique and Pendry potentials at some given distance, $z=10$ Å say. The effective potential looks fairly different from either of them, considering that all three are asymptotic to $-\lambda/z^3$ at large values of z. We have also tried fitting the potential $V(z)$ corresponding to a linear profile of adjustable width to $U(z)-\beta^2$. The fitted width depends on the range of z over which the fit is made. Fitting at $z=6.5$, 8, 10, and 15Å gives a width of 6.5 Å, but, if larger values of z are used, the width is smaller. If a more complicated type of profile is assumed, the fits tend to favor one with a very pronounced maximum in density just below the surface; however, we have been unable, so far, to obtain an unambiguous result for the density profile by these methods. Probably refinement of the Echenique and Pendry calculation would be more fruitful in yielding the profile.

In using Eq. (34) to obtain $V(z)$ from $\rho(z)$, it is perhaps worth noting that there is some latitude in the choice of the interatomic potential $v(r)$. For instance the usual values[33] of the parameters in the Lennard-Jones potential give for the asymptotic potential at large distances $-\hbar^2\lambda/2mz^3$ with $\lambda=21.5$ Å for a ^{4}He atom. The Bruch-McGee[34] potential, which uses the theoretical value for the $1/r^6$ term in $v(r)$, gives $\lambda=19.2$ Å for ^{4}He. The value that we, and Echenique and Pendry, have used, $\lambda=20$ Å, is a compromise between these two values. There is also the problem that, at large distances, retardation effects cause the $1/r^6$ term to gradually change to $1/r^7$, so that at large distances from the surface the interaction is $1/z^4$ instead of $1/z^3$. An accurate interpolation formula[27] for the interatomic potential can be integrated to give the ratio between the retarded and unretarded potential for a step profile. To study the possible effect of retardation on the reflection coefficient we multiplied our effective potential $U(z)-\beta^2$ by this correction factor and calculated the reflection coefficient. The results were very little changed and we conclude that retardation probably has an unimportant influence on the scattering probability.

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APPENDIX

Possible methods for measuring the density profile at the free-helium surface, apart from atomic scattering, include elastic neutron scattering, light scattering, and electron scattering. This appendix looks at the first possibility.

Cohen and Feynman[35] showed that inelastic neutron scattering from liquid He II can be used to determine the energy-momentum relation for the elementary excitations (phonons and rotons) in the liquid. They also pointed out that elastic scattering occurs only at the surface, where the density $\rho(\vec{r})$ varies with position. In their theory a contact interaction $\delta(\vec{r}-\vec{r}_i)$ between the neutron at $\vec{r}$ and the atom at $\vec{r}_i$ is assumed. The cross section for a process which leaves the ground state Φ of the liquid unchanged is

$$\sigma=\frac{2a^2}{k}\int|V_{00}(\vec{k}'-\vec{k})|^2\delta(k'^2-k^2)\,d\vec{k}' \ , \qquad \text{(A1)}$$

where $a=3\times10^{-13}$ cm is the s-wave scattering length and $\vec{k}$ is the wave-vector of the incident neutron. The matrix element is given by

$$\begin{aligned}V_{00}(\vec{q})&=\int\Phi^*\sum_{i=1}^{N}e^{i\vec{q}\cdot\vec{r}_i}\Phi\,d\vec{r}_1\ldots d\vec{r}_N\\&=N\int\Phi^2e^{i\vec{q}\cdot\vec{r}_1}\,d\vec{r}_1\ldots d\vec{r}_N\\&=\int\rho(\vec{r})e^{i\vec{q}\cdot\vec{r}}\,d\vec{r}\\&=A\,\rho_0f(q_z)\ ,\end{aligned}\qquad \text{(A2)}$$

where A is the area exposed to the incident beam. The function $f(q)$ is the Fourier transform of the normalized density function $\rho(z)$,

$$f(q)=\frac{1}{\rho_0}\int\rho(z)e^{iqz}\,dz \ , \qquad \text{(A3)}$$

with

$$\rho(\vec{r}_1)=N\int\Phi^2\,d\vec{r}_2\ldots d\vec{r}_N \ , \qquad \text{(A4)}$$

and ρ_0 is the bulk density. Substituting V_{00} into the expression for σ gives

$$\sigma=(a^2\rho_0^2\pi^2A/2kk_z)|f(2k_z)|^2 \ , \qquad \text{(A5)}$$

and thus the reflection coefficient is

$$R=\sigma/A\cos\theta=a^2\rho_0^2\pi^2|f(2k_z)|^2/2k_z^2 \ , \qquad \text{(A6)}$$

so that R is directly related to the Fourier transform of $\rho(z)$. For example, if $\rho(z)=\rho_0/(1+e^{\alpha z})$ with a Fourier transform

$$f(q)=-(\pi i/\alpha)\,\mathrm{csch}(\pi q/\alpha)+\pi\delta(q) \ , \qquad \text{(A7)}$$

the reflection coefficient is

$$R=\tfrac{1}{2}[a\,\rho_0\pi^2/(k_z\alpha)\,\mathrm{csch}(2\pi k_z/\alpha)]^2 \ . \qquad \text{(A8)}$$

As $\alpha\rightarrow\infty$, $\mathrm{csch}(2\pi k/\alpha)\rightarrow\alpha/2\pi kz$ and thus $R\rightarrow\pi^2\rho_0^2a^2/8k_z^4$, the expression for a step profile. Using $a=3\times10^{-13}$ cm and $\alpha\sim1$ Å^{-1} yields

$$R=(2\times10^{-11}/k_z^2)\,\mathrm{csch}^2(6.3k_z) \ , \qquad \text{(A9)}$$

where k_z is expressed in Å^{-1}. Assuming a typical momentum transfer, $2k_z=0.2$ Å^{-1}, we obtain $R\simeq4\times10^{-9}$ which would be very difficult to measure unless the incident beam were exposed to a large number of free helium surfaces, as for example, in helium adsorbed on Grafoil.

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