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Scattering of ^{4}He atoms from the surface of liquid ^{4}He at 30 mK

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Abstract. The experimental elastic reflectivity of ^{4}He atoms from liquid ^{4}He surfaces behaves as if the ^{4}He atoms are reflected from a very smooth elastic potential. We explain this anomalous result in terms of Van der Waals forces and coupling to the ripplons which swamps all other processes. Interpretation of the experiments confirms that the surface of liquid ^{4}He is diffuse on a scale of 5 Å.

1. Introduction

In recent years there has been considerable interest in the surface properties of liquid ^{4}He (Anderson 1968, Balibar 1975, Cole 1972, Griffin 1970, Kaplan and Glasser 1971), in particular the process of condensation and evaporation, and its relation to $\omega(q)$, the excitation spectrum of the liquid. Experiments that measure the elastic reflection coefficient $R(k, \theta)$ for ^{4}He atoms incident on a liquid ^{4}He surface are related to this problem and these are the experiments with which we shall be concerned in this paper. Certainly the process of reflection is a complicated one, but at first inspection one might suppose that reflection arises from the incident atom colliding with the hard cores of the liquid atoms. Some atoms would be elastically reflected, and some inelastically reflected or absorbed by the liquid, giving up their energy to characteristic excitations. We should expect that at low incident momenta the reflectivity would rise towards unity as the wavelength became large compared with the onset of the surface potential, and with increasing incident momentum, more inelastic scattering could be expected, especially above the threshold for roton excitation at which a sharp fall in elastic reflectivity should be observed.

In a remarkable experiment, Edwards *et al* (1975) produced results completely at variance with this simple picture. They measured the elastic reflectivity of low-energy ($0{\cdot}1\ \mathrm{K} \leqslant (\hbar^2k^2/2mk_B) \leqslant 3\ \mathrm{K}$) ^{4}He atoms incident on the free surface of superfluid ^{4}He at ~ 30 mK, as a function of angle of incidence and momentum $\hbar k$ of the incident atoms. We summarize their results:

(i) The probability of elastic reflection remained small ($<0{\cdot}05$) even at small momenta.

(ii) The inelastic and diffuse scattering probability was very small, less than the noise level in the experiment (2×10^{-3}) and much less than the elastic scattering probability.

(iii) No structure was observed in $R(k, \theta)$ near the roton threshold, corresponding to an incident momentum of $k = 0{\cdot}5\ \text{Å}^{-1}$.

(iv) The elastic reflection coefficient, $R(k, \theta)$, within small deviations was a function only of the momentum perpendicular to the surface, $k \cos \theta$.

In fact we summarize the situation by observing that the results behave as if the ^{4}He atom were incident on a very smooth potential step (several models will do but they must be smooth on a scale of 5–10 Å to reproduce the data). Figure 1 shows their results.

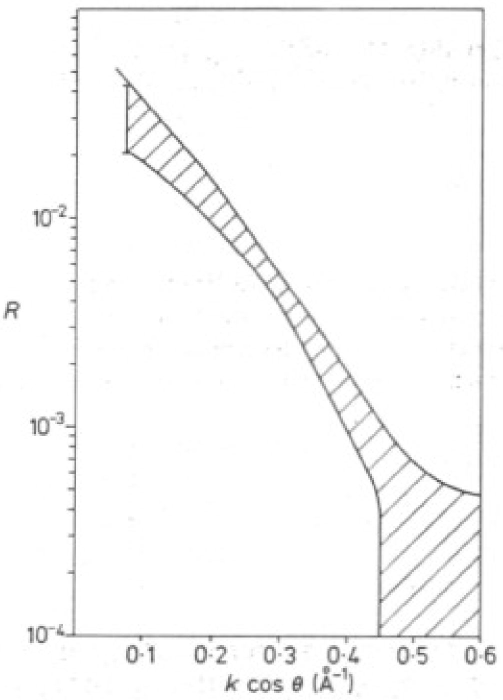

Figure 1. The experimental probability of specular reflection $R(k, \theta)$ as a function of momentum perpendicular to the surface as determined by Edwards *et al.* The shaded area encloses the experimental points.

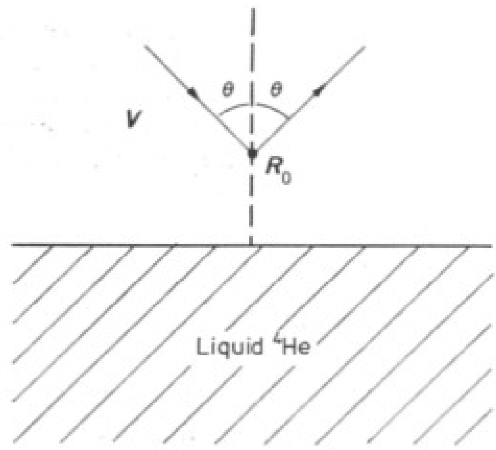

Figure 2. The dominant trajectories in the Feynman formalism are assumed to be those in which the particle travels in a straight line to the point of reflection, $\boldsymbol{R}_0$, with uniform velocity V.

Edwards *et al* pointed out that several ingredients are missing from the simple theory and our theory goes to the extreme of neglecting all short-range interactions: we assume that the relevant interaction with the surface is via the Van der Waals potential, which extends beyond the mass-density cut-off. This potential also couples the incident ^{4}He atom to the elementary excitations of the liquid, in particular to the ripplons, the quantized surface tension waves. The coupling to ripplons, though extending beyond the density limit, does not extend so far into the vacuum as the Van der Waals potential itself.

A ^{4}He atom approaching the surface is scattered first by the weak Van der Waals tail, then meets the ripplon coupling region. We shall show that the coupling switches on sufficiently rapidly that scattering from the potential close in to the surface without losing several quanta to ripplons is extremely unlikely, hence the low elastic reflectivity. The low inelastic reflectivity is explained in a similar manner: so quickly does the ripplon coupling switch on that in all but a very narrow region of space multi-ripplon loss is the most probable course of events, the incident atom being slowed down as if by frictional forces, finally being incorporated into the liquid. Roton losses occur much closer into the surface because the energy involved is greater, and in any case to give up sufficient energy near threshold the incident atom must make a transition to the ground state of the liquid. Thus the atom has already lost several ripplon quanta before it couples to the rotons which are therefore irrelevant in determining elastic reflectivities. The same argument applies to interaction with the hard-core potential.

Our model might be described as the weak interaction limit and be expected to give low elastic reflectivities. To our surprise we found that reflectivities were still too high, *unless* we took account of the diffuse nature of the surface. A sharp discontinuity in density at the surface of liquid ^{4}He is incompatible with the results of Edwards *et al.* A surface diffuse on a scale of 5 Å gives agreement with experiment in accord with theoretical calculations of the surface density by Lieu *et al* (1975).

2. Formalism

The model we adopt in the first instance is of an infinitely sharp surface to the liquid helium. The surface tension waves are taken to have a dispersion relationship given by the classical formula

$$\omega^2 = q^3 T/\rho \tag{1}$$

where T is the surface tension (0·35 dyn cm^{-1} at 0 K) and ρ is the density (0·14 g cm^{-3}). We assume that wavelengths involved are sufficiently short for gravity to be neglected. The incident ^{4}He atom is taken to couple to the density of the liquid by a Van der Waals potential, the potential energy between an element of liquid at $\boldsymbol{r}$ and the particle at $\boldsymbol{R}$ being

$$-\frac{\alpha}{|\boldsymbol{r}-\boldsymbol{R}|^6}\,\mathrm{d}^3\boldsymbol{r} \tag{2}$$

where $\alpha = 3{\cdot}232 \times 10^{-38}$ cgs units. The liquid is assumed incompressible, i.e. we assume that the surface tension waves excited are much 'softer' than sound waves which are neglected. In the absence of surface waves the particle moves in a potential

$$-\lambda/|Z|^3 \tag{3}$$

$$\lambda = \pi\alpha/6 \tag{4}$$

where we have chosen the origin at the surface and the Z axis normal to the surface. A surface wave disturbs this simple result, and results in a coupling between particle and surface mode.

We can now write a Lagrangian for the system

$$\mathscr{L} = \sum_{\boldsymbol{q}}\left(\frac{1}{4}\frac{\rho}{|\boldsymbol{q}|}\dot{A}_{\boldsymbol{q}}^2 - \frac{1}{4}\frac{\omega^2\rho}{|\boldsymbol{q}|}A_{\boldsymbol{q}}^2\right) - \phi_{\boldsymbol{q}}(\boldsymbol{R})A_{\boldsymbol{q}} + \tfrac{1}{2}m|\dot{R}|^2 + \frac{\lambda}{|Z|^3} \tag{5}$$

where A is the amplitude of the surface wave in the Z direction

$$\xi = \Omega^{-1/2}A_{\boldsymbol{q}}\,{\cos \atop \sin}\,(\boldsymbol{q}\cdot\boldsymbol{r}) \tag{6}$$

and we take the $\boldsymbol{q}$ summation to be over both cos- and sin-like waves. $\phi_{\boldsymbol{q}}(\boldsymbol{R})$ is the interaction between the wave and the particle, m is the mass of the particle, and Ω is the area of surface considered.

We wish to calculate the probability that a particle will be reflected without having lost energy to a surface wave. The problems facing us are that if we switch to a straight quantum mechanical treatment, enormous mathematical difficulties appear caused by the low energy of the ripplons which enables the particle to excite several of them simultaneously, and also results in a highly retarded interaction with the surface. The usual technique for dealing with this situation is to treat the particle classically on a time scale short compared with $1/\omega$ (Ritchie 1957, Echenique and Pendry 1975), but clearly this approach is blocked to us by the fact that reflection of the particle is an essentially quantum mechanical process.

A much more sophisticated means of straddling the classical and quantum cases is given by Feynman and Hibbs (1965) and their theory provides the basis for our calculation. Their formula states that if at t, the coordinates $\boldsymbol{R}$, $A_{\boldsymbol{q}}$ take values represented by $x_1(t_1)$, the probability *amplitude* of the system arriving at $x_2(t_2)$ is given by

$$F(x_2, x_1) = \int_{x_1}^{x_2} \exp\left[\mathrm{i}S(x_2, x_1)/\hbar\right] \mathscr{D}x(t) \tag{7}$$

where the $\int \mathscr{D}x$ stands for a weighted sum over all possible classical trajectories between x_1 and x_2, and the action for a given trajectory, $x(t)$, is

$$S = \int_{t_1}^{t_2} \mathscr{L}(\dot{x}, x, t)\, \mathrm{d}t. \tag{8}$$

Given an incident wavepacket $\psi^-(\boldsymbol{R})$, momentum $\boldsymbol{k}^-$, at $t_1 = -\infty$, and the surface in its undisturbed state

$$\Psi_0(A_{\boldsymbol{q}}) = \prod_{\boldsymbol{q}} \left(\frac{2\beta_{\boldsymbol{q}}}{\pi}\right)^{1/4} \exp(-\beta_{\boldsymbol{q}} A_{\boldsymbol{q}}^2) \tag{9}$$

$$\beta_{\boldsymbol{q}} = \frac{\omega\rho}{4\hbar q}, \tag{10}$$

we wish to calculate the amplitude of the emerging wavepacket $\psi^+(\boldsymbol{R})$, momentum $\boldsymbol{k}^+$, associated with the unexcited state of the surface. This is

$$\psi^+(\boldsymbol{R}) = \left[\prod_{\boldsymbol{q}} \int \mathrm{d}A_{2\boldsymbol{q}} \Psi_0^*(A_{2\boldsymbol{q}})\right] \prod_{\boldsymbol{q}'} \int \mathrm{d}A_{1\boldsymbol{q}'} \int F(x_2, x_1)\, \Psi_0(A_{1\boldsymbol{q}'}) \psi^-(\boldsymbol{R}_1)\, \mathrm{d}^3\boldsymbol{R}_1. \tag{11}$$

At this stage we make an approximation to enable us to solve equation (11). It is the approximation of extreme retardation in which the particle traverses all important trajectories so quickly that, although it gives $\dot{A}_{\boldsymbol{q}}$ a finite value, not until the particle is well clear of the surface does $A_{\boldsymbol{q}}$ itself become substantial, and it is to $A_{\boldsymbol{q}}$ not $\dot{A}_{\boldsymbol{q}}$ that the interaction couples. Thus the complicated formula (11) is replaced by a much simpler one

$$\psi^+(\boldsymbol{R}_2) = \int \tilde{F}_1(\boldsymbol{R}_2, \boldsymbol{R}_1) \psi^-(\boldsymbol{R}_1)\, \mathrm{d}^3\boldsymbol{R}_1 \tag{12}$$

where $\tilde{F}_1$ is the Feynman propagator for a single particle moving in a potential $-\lambda/|Z|^3$, but modified from the usual definition by an attenuation factor for each trajectory

$$\tilde{F}_1(\boldsymbol{R}_2, \boldsymbol{R}_1) = \int_{\boldsymbol{R}_1}^{\boldsymbol{R}_2} \exp\left[\mathrm{i}S_1(\boldsymbol{R})/\hbar\right] \mathscr{D}\boldsymbol{R}(t) \times \gamma(\boldsymbol{R}) \tag{13}$$

$$S_1(\boldsymbol{R}) = \int \left(\tfrac{1}{2} m \dot{R}^2 + \frac{\lambda}{|Z|^3}\right) \mathrm{d}t. \tag{14}$$

The attenuating factor, γ, is given simply by the overlap integral between Ψ_0 after the impulse conveyed by the trajectory $\boldsymbol{R}(t)$ has acted upon it, and Ψ_0^*, i.e. the probability amplitude that the trajectory excites no surface waves.

The impulse is given by $\bar{\phi}_q$:

$$\sum_q \int_{-\infty}^{\infty} \phi_q(\boldsymbol{R}(t)) A_q \,\mathrm{d}t = \sum_q \bar{\phi}_q A_q; \tag{15}$$

therefore

$$\begin{aligned} \gamma(\boldsymbol{R}) &= \prod_q \int \mathrm{d}A_q (2\beta_q/\pi)^{1/4} \exp(-\beta_q A_q^2) \exp -(\mathrm{i}\bar{\phi}_q A_q/\hbar)(2\beta_q/\pi)^{1/4} \exp(-\beta_q A_q^2) \\ &= \prod_q \int \mathrm{d}A_q (2\beta_q/\pi)^{1/2} \exp[-2\beta_q A_q - (\mathrm{i}\bar{\phi}_q A_q/\hbar)] \\ &= \prod_q \int \mathrm{d}A_q (2\beta_q/\pi)^{1/2} \exp\{-2\beta_q[A_q + (\mathrm{i}\bar{\phi}_q/4\beta_q\hbar)^2]\} \exp(-2\beta_q \bar{\phi}_q^2/16\beta_q^2\hbar^2) \\ &= \exp \sum_q (-\bar{\phi}_q^2/8\beta_q\hbar^2) = \exp \sum_q (-q\bar{\phi}_q^2/2\rho\hbar\omega). \end{aligned} \tag{16}$$

Finally, transforming the summation over $\boldsymbol{q}$ to an integral,

$$\gamma(\boldsymbol{R}) = \exp\left(-\frac{\Omega}{2\pi} \int \frac{\bar{\phi}_q^2}{2\hbar\rho\omega} q^2 \,\mathrm{d}q\right). \tag{17}$$

3. Ripplon creation

We shall assume that the dominant trajectories of the incident particle are those shown in figure 2, which is as near as we can approach to the classical limit in the reflection situation. For simplicity we shall choose

$$\boldsymbol{R}_{0\parallel} = 0 \tag{18}$$

so that only the cos-like ripplons are excited.

The particle–ripplon interaction is then

$$V_{\mathrm{PR}}(\boldsymbol{R}) = -\alpha \int \mathrm{d}^2 \boldsymbol{r}_\parallel \int_0^{u(\boldsymbol{r}_\parallel)} \frac{\mathrm{d}z}{[|\boldsymbol{R}_\parallel - \boldsymbol{r}_\parallel|^2 + |Z - z|^2]^3}. \tag{19}$$

To first order in u/Z, V_{PR} becomes

$$V_{\mathrm{PR}}(\boldsymbol{R}) = -\alpha \int \frac{\mathrm{d}^2 \boldsymbol{r}_\parallel \, u(\boldsymbol{r}_\parallel)}{[|\boldsymbol{R}_\parallel - \boldsymbol{r}_\parallel|^2 + Z^2]^3}. \tag{20}$$

Now expanding in terms of normal modes

$$u(\boldsymbol{r}_\parallel) = \frac{1}{\Omega^{1/2}} \sum_q A_q \cos(\boldsymbol{q} \cdot \boldsymbol{r}_\parallel), \tag{21}$$

we get

$$V_{\mathrm{PR}}(\boldsymbol{R}) = \sum_q \phi_q(\boldsymbol{R}) A_q \tag{22}$$

where

$$\phi_q(\boldsymbol{R}) = -\frac{\pi\alpha}{4\Omega^{1/2}} \cos(\boldsymbol{q}\,.\,\boldsymbol{R}_{\|}) \frac{q^2}{Z^2} K_2(qZ). \tag{23}$$

K_2 denotes the second-order modified Bessel function of the second kind (Abramowitz and Stegun 1965).

For the trajectory shown in figure 2 we calculate the impulse to the $\boldsymbol{q}$th mode

$$\begin{aligned} \bar{\Phi}_q &= \int_{-\infty}^{\infty} \phi_q(\boldsymbol{R}(t))\,\mathrm{d}t \\ &= -\frac{\pi\alpha}{2\Omega^{1/2}} \int_0^{\infty} \cos(\boldsymbol{q}\,.\,\boldsymbol{V}t) \frac{q^2}{(Z_0 + Vt\cos\theta)^2} K_2[q(Z_0 + Vt\cos\theta)]\,\mathrm{d}t \\ &= -\frac{\pi\alpha}{2\Omega^{1/2}} \frac{1}{V\cos\theta} \int_{Z_0}^{\infty} \cos\left[\frac{\boldsymbol{q}\,.\,\boldsymbol{V}(r - Z_0)}{V\cos\theta}\right] \frac{q^2}{r^2} K_2(qr)\,\mathrm{d}r. \end{aligned} \tag{24}$$

The function K_2 decays rapidly with increasing argument and in consequence the largest contribution to the integral comes from the region of $\boldsymbol{R}$ near to $\boldsymbol{R}_0$ and in order to proceed with analytic formulae we make use of this fact to replace cos of the expression in square brackets in equation (24) by unity. The approximation will not be valid for angles of incidence around 90° but will be exact for normal incidence. The experimental range of angles lies between $\theta = 13°$ and $\theta = 70°$. It is in consequence of this approximation that our reflectivities depend only on $k\cos\theta$, as observed in the experiments.

Our result for γ is

$$\gamma = \exp(-\delta\epsilon/k_z^2 Z_0^{15/2}) \tag{25}$$

where

$$\delta = \pi\alpha^2 m^2/16\hbar^3(\rho T)^{1/2} \tag{26}$$

and

$$\epsilon = \int_0^{\infty} \mathrm{d}x\, x^{13/2} \left[\int_x^{\infty} \mathrm{d}y\, \frac{K_2(y)}{y^2}\right]^2 \doteq [0.39] \tag{27}$$

Finally, by numerical integration of (27)

$$\gamma = \exp(-1{\cdot}075 \times 10^3/K_z^2 Z_0^{15/2}) \tag{28}$$

where K_z and Z_0 are in Å^{-1} and Å respectively. Thus the weight of a path increases with momentum perpendicular to the surface, and is also a very rapidly varying function of Z_0, the closest distance of approach.

We can make appeal to the rapid variation of γ to explain why experiments mainly observe elastic flux reflected from the surface. As soon as the particle enters the absorption zone the most likely course of events is that it loses many ripplons and becomes assimilated into the liquid without reflection. To make this point we plot in figure 3, $|\gamma_1(Z_0)|^2$ the probability of completing a trajectory with the loss of one and only one ripplon. It will be seen that there is only a very narrow domain in which one-ripplon-loss reflection processes can take place. Hence the inelastic spectrum will not be intense relative to the elastic spectrum.

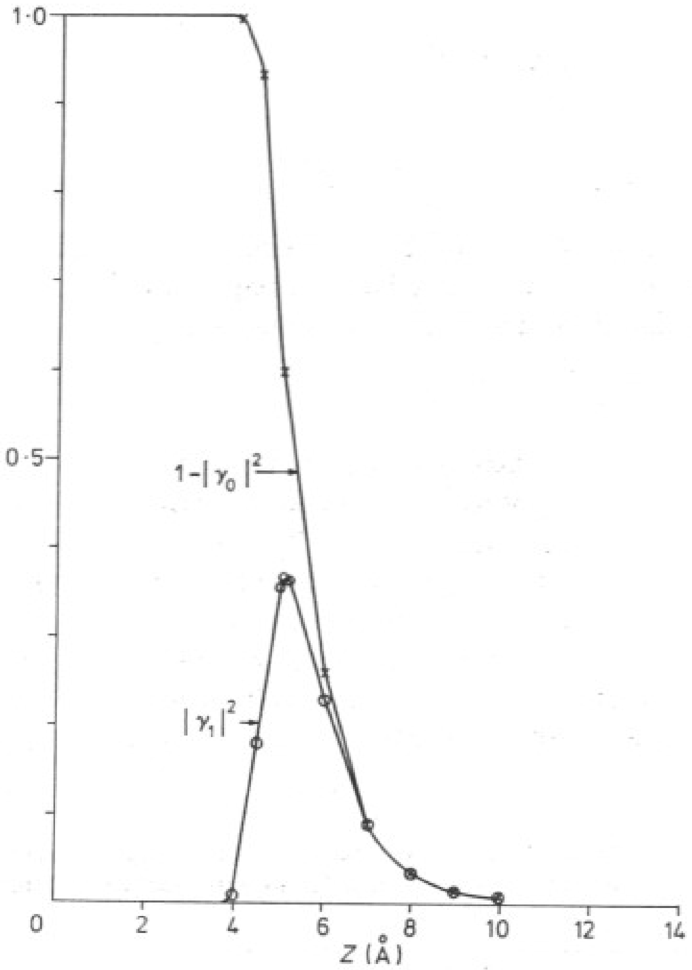

Figure 3. The probability of completing a trajectory, point of reflection at Z_0, $|\gamma_1(Z_0)|^2$ with the loss of one and only one ripplon. For comparison the probability of losing at least one ripplon is shown, $1 - |\gamma(Z_0)|^2$, demonstrating that once close to the surface a particle tends to lose many ripplons.

4. Surface reflectivity

The residual problem is to solve for the reflectivity of the Van der Waals potential $-\lambda/|Z|^3$ corrected for the attenuation of elastic flux caused by ripplon interaction. The Feynman path integral method is very useful for identifying the dominant processes and making approximations to them, but it is a clumsy tool for solving the simple problem of reflection by a potential step for which it is much easier to integrate the Schrödinger equation. We have calculated the corrections from the ripplon interaction in terms of their effects on trajectories, but we can easily recast this correction as an absorptive self-energy to be included in the Schrödinger equation. According to equations (13) and (18) an increase in the point of reflection of ΔZ_0 increases the probability of the trajectory by

$$\Delta\gamma = \gamma(Z_0)\left(\frac{15}{2}\frac{\delta\epsilon}{k_z^2 Z_0^{17/2}}\right)\Delta Z_0. \tag{29}$$

Returning to equations (13) and (14) we see that this effect can be simulated for the dominant paths by including in the Lagrangian an imaginary term

$$\mathrm{i}\Sigma_{\mathrm{i}}(Z_0) = -\frac{\mathrm{i}\hbar k_z}{2m}\frac{15}{2}\frac{\delta\epsilon}{k_z^2|Z_0|^{17/2}} = -\mathrm{i}\frac{15}{4}\frac{\hbar\delta\epsilon}{mk_z|Z_0|^{17/2}} \tag{30}$$

and we can now solve the equation,

$$-\frac{\hbar^2}{2m}\frac{\mathrm{d}^2\Psi}{\mathrm{d}Z^2} + \left(\frac{-\lambda}{|Z|^3} + \mathrm{i}\Sigma_{\mathrm{i}}(Z)\right)\Psi = \frac{\hbar^2 k_z^2}{2m}\Psi, \tag{31}$$

exactly by conventional means. We do this by observing that for some positive value of Z, Z_c, Σ_i is so large that little elastic flux reaches that region. For Z closer to the surface than this value the potential terms are given a constant value

$$-\frac{\lambda}{|Z_c|^3} + \mathrm{i}\Sigma_{\mathrm{i}}(Z_c) \qquad Z < Z_c. \tag{32}$$

It does not matter what value is taken since hardly any flux penetrates here: we only do this for computational reasons of avoiding the singularity at $Z = 0$. The wavefield is then integrated numerically from a value of $Z < Z_c$ where it is a pure decaying wave, to a sufficiently large positive value for the potential and self-energy to be substantially zero. Here the wavefunction is matched to incident and reflected plane waves.

We remark in passing that our results clearly depend only on k_z, the normal component of momentum, as observed in the experiments. In table 1 we tabulate the reflectivities we obtain and compare with experiment. The numbers are too large by an order of magnitude!

Table 1. The reflectivities of a ^{4}He surface for various incident momenta. The results of calculations for a sharp and diffuse cut-off in density at the surface are compared with experiment.

$k\cos\theta$	Sharp	Diffuse	Experiment
0·1	$7{\cdot}4 \times 10^{-2}$	$6{\cdot}0 \times 10^{-2}$	$2{\cdot}5 \times 10^{-2}$
0·2	$4{\cdot}2 \times 10^{-2}$	$2{\cdot}4 \times 10^{-2}$	$1{\cdot}2 \times 10^{-2}$
0·3	$2{\cdot}0 \times 10^{-2}$	$8{\cdot}5 \times 10^{-3}$	$4{\cdot}5 \times 10^{-3}$
0·4	$9{\cdot}0 \times 10^{-3}$	$2{\cdot}8 \times 10^{-3}$	$1{\cdot}4 \times 10^{-3}$
0·5	$4{\cdot}7 \times 10^{-3}$	$1{\cdot}0 \times 10^{-3}$	$4{\cdot}0 \times 10^{-4}$
0·6	$3{\cdot}8 \times 10^{-3}$	$4{\cdot}7 \times 10^{-4}$	$<4{\cdot}0 \times 10^{-4}$

Several approximations have been made which should be re-examined. Firstly we might ask if our classical dispersion relationship (equation (1)) for the surface waves is satisfactory. Some of the ripplons excited are of quite short wavelength, and several authors (Edwards *et al* 1974, Reut and Phisher 1971, Saam 1973, 1975) have suggested dispersion relationships differing by as much as a factor of 2 from ours in the relevant range. Against this effect being important we can say that the extremely strong $Z^{-17/2}$ dependence of the absorbing term means that even gross changes in the dispersion relationship result only in the onset of absorption being moved forward or backward by a fractional amount. The same holds true of many other corrections one might think of making: most of them are incapable of making so gross a change as an order of magnitude in reflectivity.

We believe that the answer lies in the diffuse nature of the liquid helium surface. This will change the dispersion relationship of the ripplons and the coupling of the incident particle to the ripplons, but these are minor effects compared with the smoothing of the Van der Waals potential achieved. Lieu *et al* (1975) have made a Monte Carlo calculation for liquid ^{4}He with a free surface and find diffuseness on a scale of 5 Å. We approximate their diffuse surface with a linear increase of density from the surface, achieving the bulk value in 5 Å. The corresponding Van der Waals potential is

$$\frac{\lambda}{2Z_s}\left[\frac{1}{(Z_s - Z)^2} - \frac{1}{Z^2}\right] \tag{33}$$

where Z_s takes the value 5 Å. Modifications to Σ_i will come about through changes in the ripplon modes and we can make a very crude estimate of their effect by noting that Σ_i as defined by (30) depends on the density through δ defined in equation (26)

$$\Sigma_i \propto \rho^{3/2} \tag{34}$$

(α also depends on the density). Σ_i varies very rapidly with distance from the surface and we find from equation (30)

$$\frac{\Delta\Sigma_i}{\Sigma_i} = -\frac{17}{2}\frac{\Delta Z_0}{Z_0}. \tag{35}$$

Thus our reduction in density effectively moves the absorption towards the surfaces *but by a vastly reduced factor*. Thus a factor of two reduction in density changes the position of the barrier by only 12%. Since our calculation of Σ_i is probably not accurate to within 12% in the first instance, we shall take Σ_i as being effectively unchanged by the density variation.

The results of our revised calculations appear in table 1 and show good agreement with experiment over several decades, a result that has been achieved without any adjustable parameters.

Acknowledgments

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