

RECENT EXPERIMENTS ON THE SURFACE OF LIQUID ⁴He: *Conf-750130-1*ELASTIC SCATTERING OF ⁴He ATOMS*

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ABSTRACT

The first part of the paper concerns the experimental determination of the two-dimensional effective interaction potential between ³He atoms adsorbed on the surface of liquid ⁴He. Measurements of the velocity of surface sound and of the surface tension indicate that, for concentrations of ³He corresponding to a small fraction of a monolayer, the interaction is very weak and predominantly repulsive. This is unfavorable for a low-temperature superfluid condensation.

The bulk of the paper discusses the implications of a recent experiment on the elastic scattering of free ⁴He atoms at the surface of pure liquid ⁴He. The elastic scattering probability $R(k, \theta)$ has been measured as a function of the momentum of the atom $\hbar k$ and of the angle of incidence θ . In addition the probability of inelastic scattering was found to be very small (less than 2×10^{-3}).

With the simplifying assumption that the scattered atoms can be treated as distinguishable from those in the liquid it is found that $R(k, \theta)$ can be calculated from the density profile $\rho(z)$ at the surface of the liquid. The relation between the scattering data and the spectrum of atoms evaporated from the liquid at finite temperature is also discussed.

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MASTER

Before starting on the main subject of this paper--the elastic scattering of neutral ^4He atoms from the surface of liquid helium--we would like to append some remarks to C.W. Woo's paper¹, particularly about the possibility of observing 'two-dimensional' superfluidity in ^3He adsorbed on the surface of liquid ^4He . We have studied the effective interaction between ^3He quasiparticles on the surface of ^4He by making simultaneous measurements of the depression of the surface tension, which is due to the two-dimensional pressure of the ^3He , and the velocity of 'surface sound', which is a longitudinal ^3He density wave in which the wave motion takes place parallel to the surface.²

A description of the experimental method and of the theory can be found in Ref. 3, and a more detailed account will soon be published.⁴ The measurements were made on two samples with ^3He number densities corresponding to small fractions of a monolayer. We have made a least squares fit between the data and a theoretical model in which the effective interaction is treated in the same way as the Bardeen-Baym-Pines interaction between ^3He quasiparticles in three-dimensions.^{5,6,7} The interaction is represented by the Fourier transform $V^S(k)$ which is expanded in even powers of k : $V^S(k) = V_0^S (1 + \alpha k^2 + \dots)$. Owing to the weakness of the interaction (the system is almost an ideal 2D Fermi gas) we are only able to determine the first term in this expansion, V_0^S . The results of the fit are shown in Fig. 1.

This shows the " χ^2 probability", which is the probability that the deviations from the theory are due purely to random experimental errors, as a function of the assumed values of M , the effective mass of a single surface quasiparticle, and V_0^S , the interaction parameter. (A high χ^2 probability corresponds to a good fit to the data.) We see that V_0^S is very small and probably positive. A positive V_0^S corresponds to a repulsive interaction which is unfavorable for superfluidity. The smallness of the interaction can

be illustrated in the following way: In 3D it is customary to compare V_0 with the quantity $m_4 s^2 / n_4$ where s and n_4 are the velocity of sound and number density in pure ^4He . At the saturated vapor pressure one finds $V_0 \approx 0.08 m_4 s^2 / n_4$. In comparison, the corresponding quantity for the surface $0.08 m_4 s^2 / n_4^{2/3} = 8 \times 10^{-31} \text{ erg cm}^2$, an order of magnitude larger than the most probable value in Fig. 1.

The fact that V_0^S is probably positive is not, of course, conclusive, as Woo has pointed out, since higher order terms in the expansion of $V^S(k)$ could be attractive and could lead to a superfluid condensation, perhaps in pair states with $\lambda \neq 0$. At present we are trying to make a systematic study of the surface tension and surface sound velocity over a much wider range of surface ^3He density so as to narrow down some of these possibilities. Unfortunately, because of damping by the ^2He dissolved in the bulk of the liquid, surface sound cannot be propagated in densities over ~ 1 monolayer. Hence the interesting region⁸ where the thickness of the sample approaches the coherence length in bulk superfluid ^3He (a few hundred angstroms) can only be investigated by measuring the surface tension. There are also formidable difficulties in cooling ^2He - ^3He solutions to temperature of the transition in pure ^3He , since the Kapitza boundary resistance is much larger⁹ than in pure ^3He .

We now describe the results of an experiment that we have recently completed¹⁰ to study the surface of pure ^4He : the measurement of the probability of elastic scattering $R(k, \theta)$ for ^4He atoms striking the surface of the liquid as a function of their momentum $\hbar k$ and angle of incidence θ . During the measurements the temperature of the liquid was varied between 0.025K and 0.125K and no temperature dependence was found, so that the values of $R(k, \theta)$ are characteristic of the ground state of the liquid and its

surface. The experimental method and results are described in Ref. 10 and an account of some preliminary experiments is given in Ref. 11. The present paper is confined to a discussion of the results and their theoretical interpretation. More complete references to previous work can be found in Ref. 10.

When we began the experiment some years ago¹¹ there were some definite theoretical predictions¹²⁻¹⁶ to test, predictions which had been made in connection with the theory of evaporation from liquid helium. The way in which our experiment is related to evaporation is illustrated by Fig. 2. There are three possibilities for an atom striking the surface: (a) Specular reflection (elastic scattering) in which no energy is transferred to the liquid. We define the probability of this occurring as $R(k, \theta)$. (b) Inelastic scattering in which the atom rebounds back into the vacuum above the liquid but with loss of energy to the liquid. We call the probability of this process $D(k, \theta)$. (c) Absorption or condensation (probability $f(k, \theta)$) in which the kinetic energy of the atom $\hbar^2 k^2 / 2m$ plus the binding energy L_0 (latent heat at the absolute zero, $L_0 / k_B = 7.16K$) is completely converted into excitations of the liquid. The excitations produced in (b) and (c) may include quantized surface waves ('ripples') as well as phonons. Clearly

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

The quantity $f(k, \theta)$ is related by the principle of detailed balance to the flux of evaporated atoms with momentum k and angle θ when the liquid is in equilibrium with its vapor at finite temperature. The evaporated flux of atoms with given momentum is exactly balanced by the flux of vapor atoms condensing with the same momentum and

direction. The condensing flux is the appropriate Maxwellian ideal-gas expression multiplied by the probability of condensation $f(k, \theta)$. The well-known "accommodation coefficient" $\bar{f}(T)$ ¹⁷, which is the average probability of condensation, is just $f(k, \theta)$ averaged over the Maxwellian distribution at temperature T :

$$\bar{f}(T) = \frac{\iint f(k, \theta) e^{-\hbar^2 k^2 / 2mk_B T} k^3 dk \sin \theta \cos \theta d\theta}{\iint e^{-\hbar^2 k^2 / 2mk_B T} k^3 dk \sin \theta \cos \theta d\theta} \quad (2)$$

An important result of our experiment is that $D(k, \theta)$, the probability of inelastic scattering is very small. Within our experimental sensitivity, we could detect no inelastically scattered atoms at all. Making the rather conservative assumption that if inelastic scattering does occur it will be completely diffuse, we were able to establish¹⁰ that $D(k, \theta) \leq 2 \times 10^{-3}$. Since $R(k, \theta)$ is also quite small (see the results in Fig. 3) this means that $f(k, \theta)$ is very close to unity for most k and θ and therefore that the spectrum of evaporated atoms is very close to Maxwellian when the liquid is in equilibrium with the vapor. To give a concrete example, at $T=0.6K$ integrating over the values of $R(k, \theta)$ in Fig. 3 gives $\bar{f}(0.6) = 0.989 \pm 0.002$ where the uncertainty is mainly due to the uncertainty in $D(k, \theta)$.

The theoretical predictions that we mentioned earlier¹²⁻¹⁶ are connected with a possible threshold in the condensation process (c). When $k = 0.5 \text{ \AA}^{-1}$ the combined kinetic energy and latent heat ($\hbar^2 k^2 / 2m + L_0$) of an incident atom is equal to the 'roton' energy 8.65K, that is the energy at the minimum in the well-known phonon spectrum of liquid ^4He , shown in Fig. 4. It was predicted^{12, 13} that at $k=0.5 \text{ \AA}^{-1}$ there would be a discontinuity in the spectrum of evaporated atoms and also in $R(k, \theta)$, since below

this momentum there would be insufficient energy to produce a roton excitation. Surprisingly enough there is no sign of this predicted jump in the experimental data and in fact $R(k, \theta)$ seems to depend only on the perpendicular component of the momentum $\hbar k \cos \theta$, rather than on the total momentum $\hbar k$ or kinetic energy $\hbar^2 k^2 / 2m$.

Another interesting feature of the data in Fig. 3 is the fact that $R(k, \theta)$ is so small, less than 5% even for the smallest values of $k \cos \theta$ measured, about 0.05 \AA^{-1} . By analogy with the simple quantum mechanical problem of a very slow particle approaching a stepped, attractive potential one might have expected that $R(k, \theta)$ would approach unity as $k \rightarrow 0$. At the present time we believe that this will indeed happen, but at values of $k \cos \theta$ much smaller than 0.05 \AA^{-1} . This is because of the attractive van der Waals potential outside the liquid. This varies as $-1/z^3$ and writing the Schrodinger equation (for normal incidence) in this region in the form:

$$d^2 \Psi / dz^2 + (k^2 + \lambda / z^3) \Psi = 0 \quad (3)$$

we see that the edge of the potential will not behave like an abrupt step until $k \lambda \ll 1$. For the helium van der Waal's potential, $\lambda = 20 \text{ \AA}^3$ so that $R(k, \theta)$ should approach unity when k is small compared to $\lambda = 0.05 \text{ \AA}^{-1}$.

The theoretical interpretation of the experiment has not been completed but we have developed an approximate theory of elastic scattering that gives a very satisfactory fit to the data. The approximation is to use the Feynman¹⁸ variational method and to treat the scattered atom as distinguishable from those in the liquid, i. e. . the variational wave function is not symmetrized with respect to the scattered atom. The neglect of symmetry is a serious error but perhaps not in the region above the surface where the liquid density is quite low and which we believe has a dominant effect on the reflection coefficient.

The Feynman trial wave function of a state in which one of N ^4He atoms has been replaced by an 'impurity' He atom is $\Psi = f(\vec{r}_1) \phi(\vec{r}_1, \dots, \vec{r}_N)$ where ϕ is the ground state for N ^4He atoms: $H\phi = 0$ where H is the Hamiltonian and we are now measuring energies from the ground state ($-NL_0$ compared to the vacuum). If the impurity has the same interaction potential but a different mass from ^4He , $m' \neq m$, the Hamiltonian becomes $H' = H - \frac{\hbar^2}{2} \left(\frac{1}{m'} - \frac{1}{m} \right) \nabla_1^2$.

When the expectation value of the energy

$$E = \int \Psi^* H' \Psi \, d\vec{r}_1 \dots d\vec{r}_N / \int \Psi^* \Psi \, d\vec{r}_1 \dots d\vec{r}_N \quad (4)$$

is minimized. Feynman showed that, in bulk helium, $f(\vec{r}_1) = e^{i\vec{K} \cdot \vec{r}_1}$ and the state Ψ has energy $\hbar^2 K^2 / 2m$ above the ground state and momentum $\hbar\vec{K}$. When the theory is applied to helium with a free surface it is convenient to write $f(\vec{r}_1) = \psi(\vec{r}_1) \sqrt{\rho(\vec{r}_1)}$ where $\rho(\vec{r}_1)$ is the single particle density in the ground state ϕ :

$$\rho(\vec{r}_1) = N \int \phi^2 \, d\vec{r}_2 \dots d\vec{r}_N. \quad (5)$$

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

$$\nabla^2 \psi(\vec{r}_1) + [E - U(\vec{r}_1)] \psi(\vec{r}_1) = 0 \quad (6)$$

i.e., a single particle Schrodinger equation with an effective potential $(\hbar^2 / 2m) U(\vec{r})$ which is related to the particle density $\rho(\vec{r})$ and kinetic energy density $T(\vec{r})$ in the ground state ϕ . This result, which was derived by Lekner,¹⁹ has been applied with considerable success²⁰⁻²² to the calculation of the binding energy of ^3He to the surface the potential $U(\vec{r})$ for ^3He having a minimum at the surface.

In our scattering problem the 'impurity' is the scattered ^4He so that $m' = m$ and the effective potential is given by

$$\begin{aligned}
 U(\vec{r}) &= a'' / a \\
 a(\vec{r}) &= \rho(\vec{r}) \\
 a''(\vec{r}) &= \nabla^2 a(\vec{r}) - \frac{d^2 a}{dz^2}
 \end{aligned}
 \tag{7}$$

Since $\psi^* \psi$ is the probability density for the scattered atom and ψ obeys a single-particle Schrodinger 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 $U(\vec{r}) = U(z) = a'' / a$. The probability for elastic scattering is thus directly related to the density profile of the liquid $\rho(z) = [a(z)]^2$.

The reader will notice two features of the theory which agree very well with the observations: (a) The predicted reflection coefficient can only depend on the perpendicular component of the incident momentum $\hbar k \cos \theta$. (b) There can be no inelastic scattering, only specular reflection or absorption. It is also clear that the theory can be applied to ^3He scattering from ^4He although this has not yet been measured.

In calculating the reflection coefficient the proper asymptotic behavior of $a(z)$ outside the liquid must be taken into account. Far from the surface, where the liquid density drops off exponentially, the effective potential must be identical to the real, van der Waals potential:

$$\left. \begin{aligned} a &\rightarrow \exp \left[-\left(\beta z + \frac{\lambda}{4\beta z^2} \right) \right] \\ U &\rightarrow \beta z^2 - \lambda / z^3 \end{aligned} \right\} \text{as } z \rightarrow \infty \quad (8)$$

where $k^2 = \beta^2 / 2m = L_D^{-2}$.

When this condition is satisfied it is relatively easy to find analytic forms for $a(z)$ and $U(z)$ which give excellent agreement with experiment. In this way we can use the scattering data to determine the density profile of the liquid at the surface $\rho(z) = [a(z)]^2$. However it is not yet clear how the neglect of symmetry will impair the derived density profile or whether this approximation can be eliminated. The details of the fit and of the theoretical formalism will be reported elsewhere.

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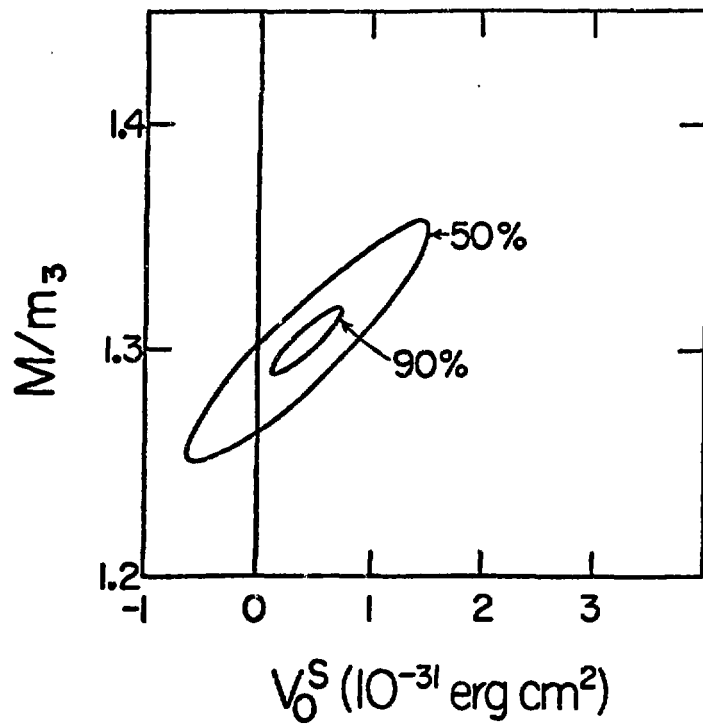
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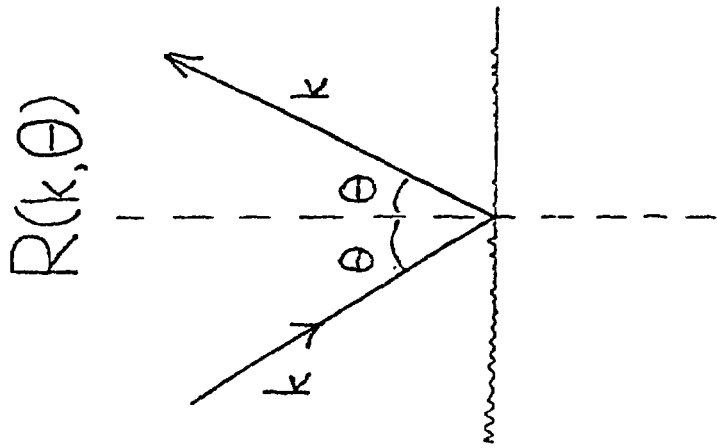
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Figure Captions

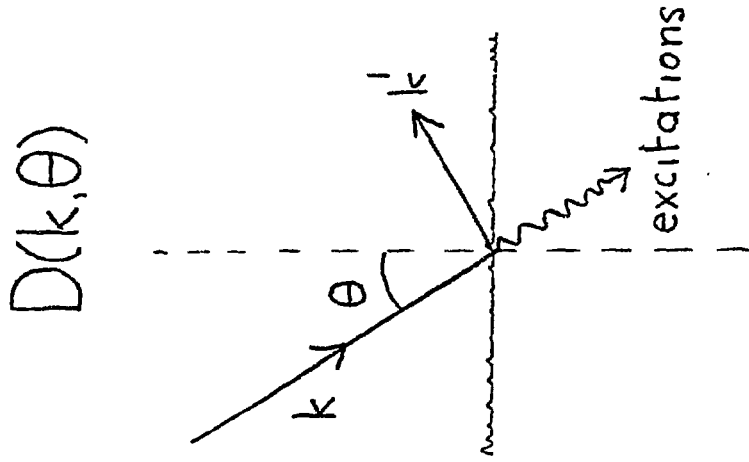
- Fig. 1 Least-squares fit of the interacting 2D gas model to data on the velocity of surface sound and on the surface tension in two samples of ^3He adsorbed on ^4He . The graph shows contours of the χ^2 probability as a function of the quasi-particle effective mass M and the interaction parameter V_0^S (see text). A positive value of V_0^S corresponds to an interaction which is predominantly repulsive at large distances. (From Ref. 4).
- Fig. 2 The three possible fates for a free ^4He atom striking the surface of the liquid and their probabilities: (a) elastic scattering (specular reflection) (b) inelastic scattering, converting part of the atom's kinetic energy into excitations (phonons and ripplons) in the liquid (c) absorption, creating excitations whose total energy is equal to the kinetic energy of the atom plus the binding energy, L_0 .
- Fig. 3 The probability of specular reflection $R(k, \theta)$ for ^4He atoms striking the surface of liquid helium as a function of $\hbar k \cos \theta$, the perpendicular component of their momentum (from Ref.10). The different symbol shapes refer to the angles of incidence θ shown in the left hand corner. The arrows show the roton threshold ($k = 0.5\text{\AA}^{-1}$) for each angle. Overlapping points have been omitted for the sake of clarity
- Fig. 4 The energy-momentum relation for free atoms $\epsilon(k)$ relative to their ground-state energy in the liquid, compared with the phonon-roton excitation spectrum. The finite energy for the atoms at $k=0$ is the binding energy

$L_0/k_B = 7.16K$. The energy of an atom is equal to the phonon energy at the roton minimum when $k = 0.50 \text{ \AA}^{-1}$. (From Ref. 10).

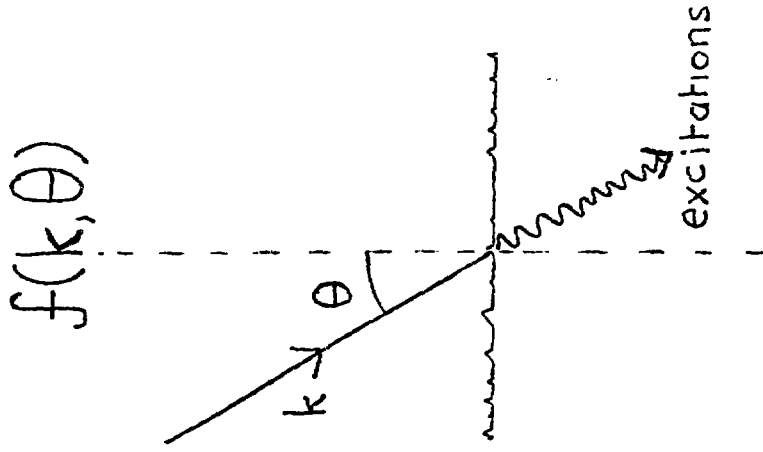




a)



b)



c)

