Reflectivity of ^{4}He Atoms from Liquid ^{4}He: Direct Observation of the Effect of Phonons and Rotons

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It was suggested some years ago that the reflectivity of ^{4}He atoms, reflecting from the free surface of liquid ^{4}He, should show evidence of rotons being created by some of the condensing atoms. However, the measured reflectivity showed no change as the atom energy was scanned through 1.45 K, the energy which is necessary to create a roton of the minimum energy. This was strange as rotons, albeit of higher energy, were detected in the liquid from condensing atoms. We report here clear evidence for the creation of phonons and rotons in the reflectivity data. This enables us to estimate the probabilities for condensing atoms to create ripplons (92%) (upper limit), phonons (1.5%), and rotons (3%), (both lower limits), at an angle of incidence of 83°.

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Evaporation and condensation are commonplace phenomena and are well understood on a macroscopic scale. However, if one poses the question, asking what are the microscopic processes that are involved in an atom or molecule leaving the liquid state and entering the vapor state, or vice versa, then there are few answers. For classical liquids we may have to be content with a statistical description, but for quantum liquids we can answer in more detail. We know that a single phonon or roton incident on the free surface can eject an atom in a one-to-one process [1–3]. The phonon or roton is annihilated and an extra atom in the vapor is created. This process of quantum evaporation can be studied because the phonons and rotons in liquid ^{4}He have very long lifetimes at low temperatures so they can propagate to the free surface without decaying or scattering. Also the free surface is translationally invariant so parallel components of momentum are conserved, therefore evaporation by phonons and rotons can be clearly distinguished. These conditions do not occur in any other system so it is important that we understand evaporation and condensation with ^{4}He in as much detail as possible.

The surface of liquid helium is important in these processes. It can be studied by measuring the reflectivity of ^{4}He atoms from the surface, and this has been done extensively by Edwards and co-workers [4,5]. The reflectivity is small as most of the atoms in the incident beam condense. However, the reflectivity is found to be a strong function of the perpendicular component of the incident atom's wave vector $k_\perp$. This is explained as a quantum reflection from the van der Waals potential well at the surface of the liquid ^{4}He, and, generally, good agreement between the model and the measurements is found [6]. The atoms are reflected from the top part of the potential well created by the liquid, and do not approach very near to the atoms in the liquid [7]. The incident atoms that are not reflected are accelerated into the liquid by the potential. They condense into the liquid and their excess energy (the initial kinetic energy far away from the surface plus the latent heat per atom) goes into excitations. These might be ripplons on the surface or phonons and rotons in the bulk of the liquid ^{4}He.

It was recognized [4] that if the incident atom has less excess energy than any roton energy (minimum roton energy is 8.61 K [8]) and if roton creation is a significant channel for condensation, there should be a change in the reflectivity as the excess energy of the incident atom is scanned through 8.61 K. However, all the measurements of reflectivity showed no significant feature at this energy [4,5], so it was concluded that the creation of rotons on condensation does not occur and that ripplon production is the only energy loss mechanism. This conclusion did not seem to happily coexist with the results from quantum condensation experiments. When bulk excitations were looked for in a condensation experiment, there was clear evidence that rotons were being produced [9,10]. However, the rotons that were detected were well above the energy of the roton minimum so it remained arguable that the probability of roton production decreased with decreasing roton energy and became insignificant at the roton minimum. This behavior would then explain why no step change in the reflectivity occurred at the roton minimum energy. So the situation was that the atom reflection experiments showed no evidence for condensation to bulk excitations, while the transmission experiments showed that at least some of the atoms condensed, producing rotons. Although there was no direct conflict of evidence, there was concern that the situation was not fully understood.

We report here new measurements of the atomic reflectivity which show evidence of a phonon and roton channel. The measurements suggest it is weak compared to the ripplon channel for the glancing angles which we have used.

To envisage the phonon and roton channels, consider an incident atom that is not reflected by the potential well but goes on to condense, so producing a phonon or a roton. If these processes are not allowed for some atom energies,

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then these atoms must go elsewhere and so some or all are reflected. This is shown schematically in Fig. 1. So if we scan through the atom energies, we should see a larger reflection coefficient where the channels are closed.

The reason for the roton channel closing is the atom not having enough energy to create a roton. As rotons have energy ≥8.61 K and as the latent heat of condensation is 7.16 K [11], the minimum kinetic energy of the incident atom must be 1.45 K to create a roton. When the atom energy is less than 1.45 K we expect the reflectivity to rise.

The condition for the phonon channel to be open or closed is not based on energy considerations as any condensing atom has enough to create one or more phonons. However, there is a limitation based on the component of momentum parallel with the surface of the liquid, $\hbar k_{\parallel}$. Because of the translational invariance of the surface this momentum must be conserved. If the atom has a larger $k_{\parallel}$ than the phonon wave vector q with the corresponding energy, then it is not possible to satisfy $k_{\parallel} = q \sin\theta$, where θ is the angle between the phonon momentum and the surface normal. On the contrary, if $k_{\parallel} < q$ then the phonon direction can be chosen so that $k_{\parallel} = q_{\parallel} = q \sin\theta$. So for low energy atoms at a given angle of incidence ϕ, i.e., atoms with energy less than a critical energy (we shall see below that this is 1.24 K at $\phi = 83°$), the phonon channel is possible, but above this energy it is closed. So there is a small range of atom energies, at large angles of incidence, where neither the phonon nor roton channel is open. For these atoms we expect the reflectivity to increase. The amount of the increase will depend on the probability of an atom entering these condensation channels when they are open.

To calculate the phonon cutoff we study the plot shown in Fig. 2, where we show the atom energy against the component of wave vector parallel to the surface, $k_{\parallel}$, and the phonon energy against the full phonon wave vector q [12]. The atom dispersion curves are offset from the phonon dispersion curve by the enthalpy per atom needed for evaporation, i.e., 7.16 K. If an atom curve lies on the high wave vector side of the phonon curve, then it is impossible for a phonon to be created by that atom in a condensation process and conserve the parallel component of wave vector. In contrast, where the atom curve is on the low wave vector side of the phonon curve, then one or more phonons can be created. The phonon(s) in this case can have an angle <90° to the surface normal so that the component of parallel momentum is conserved. For an angle of incidence of 83° for the atom, we find that the maximum energy for an atom to produce a phonon is 1.24 K. The energy threshold for the phonon channel varies with angle of incidence, whereas for the roton channel it is constant. At 70° the two energies are coincident at 1.45 K. At this angle the reflectivity change at this atom energy will be a minimum, and for smaller angles there should be a dip in the reflectivity.

We summarize the position in Fig. 3 and we see that for atom energies between 1.24 and 1.45 K neither phonons nor rotons can be produced. Also shown in Fig. 3 is the effect of the incident atom losing a small amount of energy (0.1 K) to ripplons as well as creating a bulk excitation. We see that it raises both the phonon and roton thresholds so that the energy gap where no bulk excitations are produced is narrowed. We also see that if there is a small energy loss to ripplons then both the phonon and roton thresholds are broadened on their high energy sides only.

An incident atom has several possible final states or channels. We define the probabilities $P_{\rm ref}$, $P_{\rm rip}$, $P_{\rm phn}$, and $P_{\rm rot}$ as the probabilities for the atom to specularly reflect, create a ripplon(s), phonon(s), or roton, respectively. We ignore the creation of, say, both a phonon and a ripplon explicitly at this juncture. We could imagine that we add

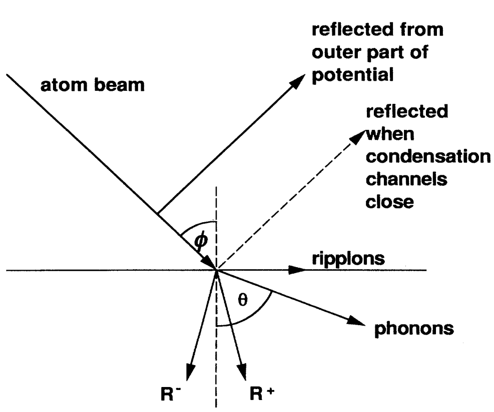

FIG. 1. An atom approaching the liquid surface is either reflected quantum mechanically from some height above the surface or condenses into the liquid to form ripplons and/or bulk excitations. If any of these condensation channels are closed, there will be an additional probability of reflection.

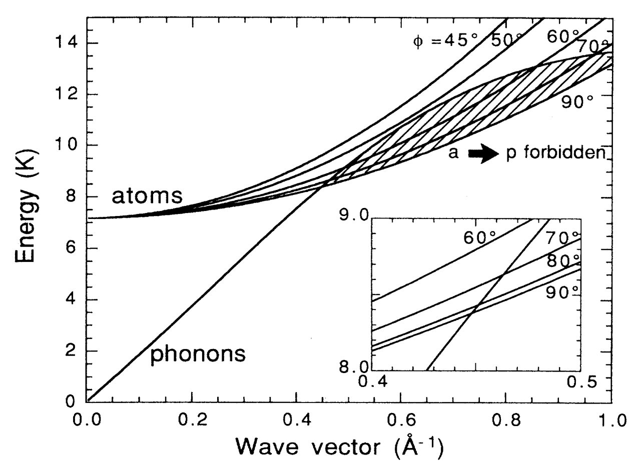

FIG. 2. The dispersion curve of phonons and atoms. Energies are relative to the liquid ground state. For atoms, the wave vector is multiplied by $\sin\phi$ for various values of ϕ, where ϕ is the angle of incidence for atoms. The condensation process, atom → phonon(s), is forbidden kinematically to the right of the phonon dispersion curve. Inset: The region of interest on a larger scale.

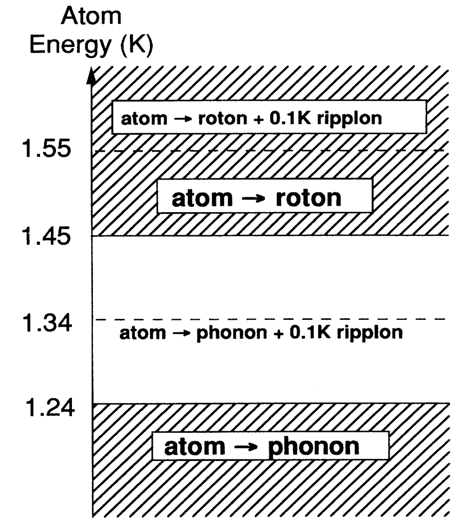

FIG. 3. Kinetic energy diagram for atoms showing allowed quantum condensation processes for an incident atom angle of 83°. If $E_k < 1.24$ K, phonon(s) can be created, but rotons can only be created if $E_k > 1.45$ K. Atoms within this energy range $1.24 < E_k < 1.45$ K can only condense and create ripplons. If ripplon production is coupled with the creation of bulk excitations, these energy thresholds are increased by the energy of the ripplons, and are therefore softened by the statistical nature of ripplon production. As an example, the effect of a 0.1 K ripplon on the two thresholds is shown by the dashed lines.

such a process to the phonon probability if the ripplon has a very small energy compared to the photon energy but otherwise add it to the ripplon process. The probabilities add to unity so we see that $P_{\text{ref}} = 1 - P_{\text{rip}} - P_{\text{phn}} - P_{\text{rot}}$. If the phonon or roton channel is not available, i.e., $P_{\text{phn}} = 0 = P_{\text{rot}}$, then we expect P_{ref} to increase as might P_{rip}. Thus we might expect changes in reflection probability as a function of energy at the thresholds for the phonon and roton creation processes.

To look for these changes we set up two reflection experiments similar in principle to that of Edwards *et al.* [4]. The main differences are in the atom beam generator, detector, and electronics. The atom beam is generated by a thin film Au heater which evaporates some of the helium film covering it in a short heating pulse. The atom beam is collimated into a beam ~2° wide and is incident on the liquid ^{4}He surface at 83° to the surface normal. The total atom path length is 50 mm in one experiment and 53 mm in another. The reflected beam passes through a horizontal collimating slit so that the angular width of the beam can be measured. The reflected beam is detected by a superconducting Zn bolometer biased at the transition edge by a constant temperature feedback circuit. This gives a short time constant of ~10 μs for the bolometer in the vacuum but covered with a superfluid ^{4}He film. The experiments were done at $T < 200$ mK where there is an excellent vacuum above the liquid. The signals are found to be independent of temperature. The signal from the bolometer is amplified and averaged up to 10^6 times. The signal is essentially proportional to the deposited energy. Ultrapure ^{4}He is used in the experiment [13]. The heater is heated with a short (5 μs) low power (1 mW) pulse. This creates essentially a delta function pulse of atoms and, because they travel ballistically, their time of flight can be used to find their energies.

A typical result is shown in Fig. 4 for a path of 50 mm. We see an approximately Maxwellian distribution modified by the energy dependent reflectivity, but at 600–700 μs there is an increase in reflectivity. It is this feature that we ascribe to the closing of the phonon and roton channels. It is a persistent feature and has been seen in two separate experimental arrangements. It is not particularly sensitive to the atom flux. We checked that it is a signal from atoms reflected from the helium surface by spoiling the translational invariance of the surface and seeing that the signal decreased. The time of flight for atoms with energy 1.24 K (the phonon threshold) is 700 μs, and we see that there is a distinct change in the reflectivity at this point. (The graph should be read from right to left to see the effect of increasing atom energy.) The roton threshold at an atom energy of 1.45 K corresponds to an arrival time of 645 μs, and, although the feature is declining at this time of flight, there is not a sharp threshold. This may well be due to low energy ripplon production as discussed above. We have also measured the signal from an atom beam over the same propagation distance from a similar heater but with the same bolometer (Fig. 4). The reflection coefficient is computed as the ratio of the two signals at the same time. The result of this is shown in Fig. 5.

The general values of the reflectivity as a function of atom wave vector ($k_\perp$) are similar to those found by

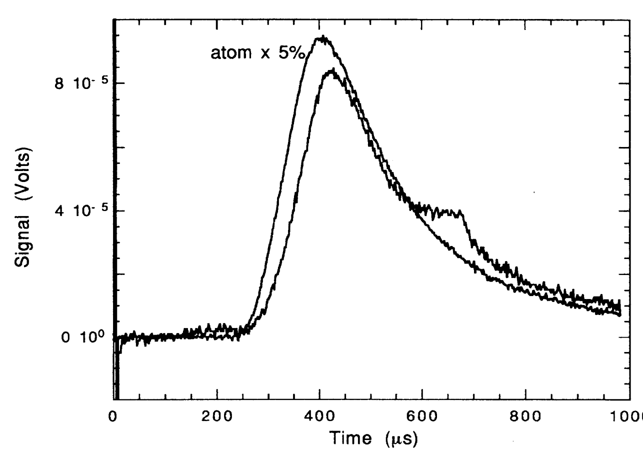

FIG. 4. Comparison of the reflected atom signal with the direct atom signal (reduced by a factor of 20). The feature at $600 < t < 700$ μs is very clear, and corresponds to both the phonon and roton condensation channels being closed. Phonon creation is allowed for $t > 700$ μs, and roton creation for $t < 645$ μs.

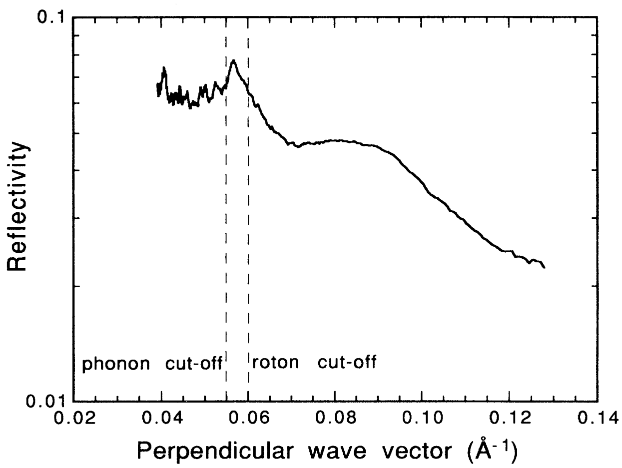

FIG. 5. The reflectivity is the ratio of the two signals shown in Fig. 4. It is plotted as a function of perpendicular wave vector to enable comparison with previously published data. The peak at 0.057 $Å^{-1}$ is due to the phonon and roton channels being closed.

Edwards *et al.* [4]. However, there is a sharp peak at $k_\perp = 0.057$ $Å^{-1}$ which we ascribe to the closing of the phonon and roton channel. There is a broader feature between 0.07 and ~0.10 $Å^{-1}$ which is also not present in previous data. This is possibly due to the probability of the roton channel being energy dependent. The angular dependence of the reflectivity due to the channels closing seems to be present in the data of Nayak [14].

From the reflectivities of the peak and on each side of the peak we estimate that $P_{\text{rip}} = 0.92$, $P_{\text{phn}} = 0.015$, and $P_{\text{rot}} = 0.03$, assuming that these probabilities are independent of each other. If this is not so then 0.92 represents the upper limit of ripplon production and 0.015 and 0.03 represent the lower limits for phonon and roton production, respectively. This indicates that a large majority of the atoms can condense, producing ripplons at this angle of incidence. We expect that these probabilities will vary with angle of incidence, because when the atom comes in at glancing angle it spends more time just above the liquid surface than if it came in nearer to the surface normal, so it has a greater chance of creating a ripplon. When the phonon and roton channels are closed the probability of creating ripplons is indeed high, 0.92. This is consistent with the theoretical model of Echenique and Pendry [7], who found that the incoming atom was strongly coupled to surface excitations. It is interesting to compare this with the quantum evaporation results. Here it is found that a phonon or roton evaporates an atom with small upper limits on any lack of conservation of energy or parallel momentum. These are 0.14 K from the phonon-atom angular width [15] and 0.39 K from the roton-atom time of flight [16]. This implies that the evaporated atom has a low probability of creating a ripplon as it leaves the surface of the liquid ^{4}He. This clearly raises the question why this behavior is different from the one for an incoming atom in a condensation process. We suggest that it is due to the evaporated atom coming from the top of the surface where the density profile has decreased well below the bulk density, whereas the condensing atom penetrates deeply into this profile and creates the ripplons by the other atoms adjusting their position to accommodate that extra atom. In other words, an atom leaving the top of the surface is much less perturbing than an atom joining the liquid where the density is high. Finally, we should say that there is no information on the probability that a phonon or roton incident on the free surface produces ripplons. That is a measurement for the future.

In conclusion, we have seen the effect on the atom reflectivity of condensing atoms creating phonons and rotons in the bulk liquid ^{4}He. We find that the reflectivity increases when the possibility for creating a phonon or roton is prohibited. This removes the apparent inconsistency with the observation that rotons are produced by condensing atoms. In agreement with previous work it is found that an incident atom can have a high probability of creating ripplons. The probabilities of creating phonons or rotons is apparently low at large angles of incidence.

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