

Quantum Evaporation from Liquid ^4He by Rotons

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We have shown that rotons as well as phonons can evaporate ^4He atoms in a single-quantum process. Measurements of the time of flight and the angular distribution of the evaporated atoms clearly distinguish between evaporation by phonons and rotons. The results indicate that energy and the parallel component of momentum are conserved at the free liquid surface.

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We have recently shown that He atoms can be evaporated by phonons in liquid ^4He in a single-quantum process in which one phonon evaporates one He atom.¹ In this Letter we report measurements of the angular distribution of ^4He atoms evaporated by a beam of phonons and rotons which are incident at an angle of 13° to the normal to the free surface. This distribution can be understood in terms of the boundary conditions at the surface. This, together with the good agreement between the measured and calculated times of flight, allows us to conclude that ^4He atoms are evaporated by rotons as well as phonons in a quantum interaction in which an excitation in the liquid is annihilated and an atom in a free state is created.

The evaporation of liquid ^4He has been discussed for many years.^{2,3} Interest in it was rekindled by the work of Johnston and King⁴ who attempted to measure the velocity distribution of atoms evaporating from liquid ^4He at 0.6 K, although their results turned out to be incorrect.⁵ They apparently saw a spectrum of atoms with characteristic temperature (1.5 K) well above that corresponding to the liquid temperature (0.6 K). This stimulated^{6,7} explanations in terms of a roton, at the roton minimum energy Δ , giving its energy to an atom in the liquid which after evaporation would have kinetic energy equal to $\Delta - E_B = 1.5$ K, where E_B is the latent heat per atom. The large number of rotons at this energy (8.65 K) where the density of states is infinite was thought to give rise to a dominant number of atoms with energy 1.5 K. However, Cole⁸ pointed out that these rotons, although numerous, have a low attack frequency on the interface because their group velocity tends to zero. In fact, the effect of the density of states and group velocity cancel each other. More recent measurements^{5,9,10} have shown that evaporated ^4He atoms do have energies commensurate with the temperature of the liquid.

The important suggestion made by Anderson⁶ and Hyman, Scully, and Widom⁷ that a single excitation could evaporate a single atom was tested by

Balibar *et al.*¹¹ However, at the low temperatures $T < 0.3$ K which are needed for long phonon mean free paths, they were unable to detect atoms evaporated by phonons. The signals which they ascribed to roton-atom desorption lead to a maximum velocity for these rotons of 160 msec^{-1} . This is a mysterious result for which no convincing reason could be given. We now believe that they used heater powers which were too high, as we find that the detected atom-signal changes shape for powers above 10 mW mm^{-2} and it appears slower than those for lower powers. We can reproduce the signals seen by Balibar *et al.* by increasing our heater power, but at low powers we see signals from both phonons and rotons that can be understood in terms of the known excitation spectrum.

Previously, we have shown that phonons above the three-phonon cutoff energy can evaporate either ^3He or ^4He atoms, in a single process with energy conservation.¹ In that experiment the phonons and atoms traveled on a path normal to the interface. Here we have phonons and rotons at an angle to the normal and we measure the angular spectrum of evaporated atoms. This enables us to measure the change in momentum at the interface and distinguish between the effects of phonons and rotons. We find that energy and parallel momentum are conserved.

The experiment is carried out in a cell partially filled with liquid ^4He and cooled to 0.1 K with a dilution refrigerator. At this temperature the vapor pressure of ^4He is so low that there is a negligible chance of the atom beam being scattered by the ambient concentration of atoms. The excitations are produced by a thin-film heater in the liquid ^4He which is pulse heated for a period of $\leq 10 \mu\text{sec}$ and they are collimated into a beam directed at the free surface. A bolometer and collimator points at this area of liquid surface and detects atoms which are liberated. The bolometer can move in an arc centered at the point on the surface where the excitations are incident so that the angular distribution of atoms can be measured. The excitations travel 6.5

mm in the liquid and the atoms the same distance in the vacuum above the liquid. The number of atoms evaporated is so low that there is a negligible chance of collisions between them. The signal created by the atoms condensing on the bolometer is recorded as a function of time.

The experimental arrangement is shown schematically in the inset in Fig. 3. The heater and bolometer are 1 mm² each, the collimator c_1 is a circular hole 0.4 mm in diameter, and c_2 is an elongated hole with dimensions 0.4 mm in the plane of the figure and 1.0 mm perpendicular to the paper. θ and ϕ could be altered with superconducting stepping motors. The measured angles are correct to $\pm 2^\circ$.

The thin-film heater (h) is pulse heated with a power of -17 dB (referred to 0.5 W/mm²). This is in the lower power regime where the emitted phonon energy is proportional to the pulse input energy,¹² and where a ballistic phonon pulse can propagate through liquid ⁴He. The heated film injects a broad spectrum of excitation frequencies into the liquid.

In this experiment we wanted a clean ⁴He surface. As any ³He atoms in the liquid ⁴He will go to the free surface until a monolayer is formed,^{13,14}

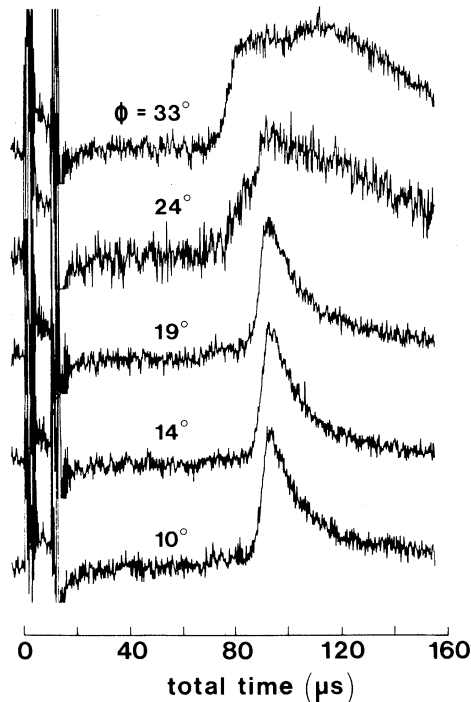


FIG. 1. Recorded signals as a function of time for different angles ϕ with $\theta = 13^\circ$. The time scale begins at the start of the crosstalk from the 10- μ sec pulse. At $\phi = 10^\circ$, 14° , and 19° the atom signal is due to phonons; at 33° it is due to rotons.

we designed the experimental cell with a considerable amount of copper sinter above the free liquid surface. We estimate that this was sufficient to reduce the amount of ³He on the surface to negligible proportions. Indeed we saw no difference between natural well gas and purified ⁴He¹⁵ with a concentration of one part of ³He in 6.5×10^{10} parts of ⁴He.¹⁶

The experimental results shown in Fig. 1 are a small sample of an extensive set of results. For incident angles $\phi = 10^\circ$, 14° , and 19° a phonon-atom signal can be seen with a minimum total time of 87 μ sec. As the angle ϕ is increased to 24° a faster signal is apparent and by $\phi = 33^\circ$ a roton-atom signal with a well-defined takeoff at 70 μ sec is established. At this angle there is no sign of a phonon-atom signal. The phonon-atom pulse is much narrower than the roton-atom one.

In order to show the angular separation of the phonon-atom and roton-atom signals we have integrated the detected signals, shown in Fig. 1 and others, over the first 17 μ sec, starting at 70 μ sec for the roton atoms and 87 μ sec for the phonon-atom signals. These are shown in Fig. 2. It can be seen that the two effects are easily resolved and the peaks occur within a few degrees of the angles predicted by the simple analysis which we now give.

Consider an excitation in liquid ⁴He traveling towards the free surface at an angle θ to the normal to the surface. Let the energy of the excitation be $\hbar\omega$, its momentum to be $\hbar\vec{q}$, and its group velocity be v . At the surface we assume that the excitation is either reflected or gives all its energy to a surface atom. The atom is bound to the liquid with a binding energy E_B . If $\hbar\omega > E_B$ then the atom has sufficient energy to escape into free space and if energy is conserved in the excitation-atom system the atom will have kinetic energy given by

$$\hbar^2 k^2 / 2m = \hbar\omega - E_B, \quad (1)$$

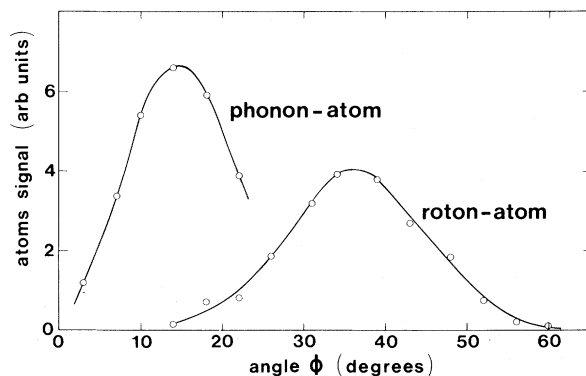


FIG. 2. The atom signals integrated over the first 17 μ sec are shown as a function of angle ϕ for $\theta = 13^\circ$.

where $\hbar\vec{k}$ and m are the momentum and mass of the free atom.

The direction of \vec{k} will depend on the boundary conditions. If we approximate the wave packet of the excitation by a plane wave with wave vector \vec{q} and similarly the atom by a plane wave with wave vector \vec{k} , and we recognize the translational invariance of the surface, there can be no change in momentum parallel to the surface, so that

$$|\vec{q}|\sin\theta = |\vec{k}|\sin\phi, \quad (2)$$

where ϕ is the angle between \vec{k} and the normal. Combining Eqs. (1) and (2) we obtain

$$\sin\phi = \hbar|\vec{q}|\sin\theta/[2m(\hbar\omega - E_B)]^{1/2}, \quad (3)$$

where ω and q are related by the dispersion curve for liquid ^4He at the saturated vapor pressure.^{17,18} For an excitation of given frequency ω , all the quantities on the right-hand side of Eq. (3) are known and so $\sin\phi$ can be calculated.

Besides the direction of the evaporated atoms we can measure the total time between injecting the excitations into liquid He and detecting the arrival of the atoms. If the distance through the liquid is l_1 and through the vacuum l_2 then the time due to an excitation with wave vector q is

$$t(q) = l_1/v(q) + l_2/v_a(k), \quad (4)$$

where

$$v(q) = d\omega/dq|_q$$

$$v_a(k) = \hbar k/m = [2(\hbar\omega - E_B)/m]^{1/2}. \quad (5)$$

For fixed values of l_1 and l_2 , $t(q)$ is only a function of q and in the ranges of q where $\hbar\omega > E_B$, there is a minimum value of $t(q)$ in each of the three regions of the excitation curve, namely, the phonon region and the two regions with q less than or greater than the roton minimum, which we will refer to as low- and high- q rotons. The values of the minimum time (t_m) are quite different in the three regions and so t_m gives a way of identifying the received signals, in addition to the angular distribution.

So we see that with the assumptions of a one to one interaction and conservation of energy and the parallel component of momentum, the atom evaporated by a particular excitation will travel at angle ϕ to the normal which depends on ω and \vec{q} of the excitation. In addition, the total time for the signal to go from heater to bolometer also depends on ω . Hence, we can calculate the curves shown in Fig. 3 where we have taken E_B to be the latent heat per atom (7.15 K).¹⁹ Points on these curves are labeled with the excitation energy expressed in kelvins.

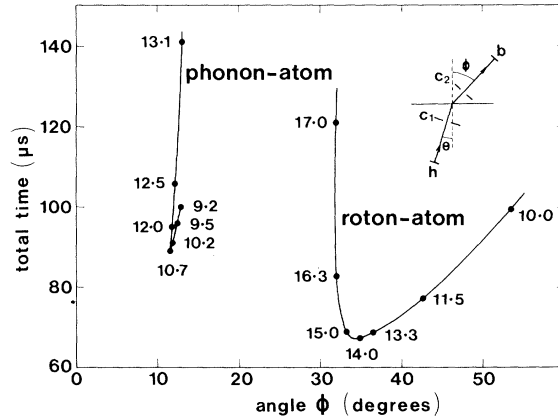


FIG. 3. Calculated total time for phonon-atom and roton-atom signals shown as a function of angle ϕ for $\theta = 15^\circ$. Points on the curves are labeled with the excitation energy in degrees kelvin.

The curves are for a single angle of incidence $\theta = 15^\circ$.

The behavior for high- q rotons and phonons (low- q rotons are not shown in Fig. 3) are quite different both in minimum total time and in refracted angle. The phonons desorb with little refraction (i.e., $\theta \sim \phi$) because the momenta of the phonons and atoms are similar. However, the total time varies rapidly with ω . Phonons with energy less than the critical cutoff for three-phonon processes, which is ~ 9.5 K at zero bar, do not reach the free surface before decaying to an energy below E_B .^{20,21}

In contrast, the rotons desorb over a wide range of angles which increases rapidly as the roton energy is decreased below 14 K. However, for rotons of higher energy the angle does not decrease very much and there are no atoms with $\phi < 32^\circ$ (for $\theta = 15^\circ$). However, for high-energy rotons the total time rapidly gets longer because of the decrease in the roton group velocity.

In the experimental arrangement there is a finite range of incident angles θ that come through the collimator. In fact the range is $\pm 9^\circ$ which means we have a set of curves similar to those in Fig. 3. These curves all have minima at similar total times with only a slight variation due to the different path lengths involved. This means that we expect to see the same time for the fastest signals over a range of angles ϕ . However, the collimation is still good enough to clearly separate in angle the signals due to phonon-atom and roton-atom interactions. The calculated times are 89 and 67 μsec which compare well with the measured times of 87 and 70 μsec for phonons and rotons, respectively.

The different widths of the phonon-atom and

roton-atom signals can be understood in the following way. The fastest phonon-atom signal is due to phonons just above the critical frequency. We would expect these to be much more highly populated than higher phonon states so that there are fewer phonons at energies which give long total times. In contrast the fastest roton-atom signals come from 14.3-K rotons which are well above the roton minimum, so that there are a relatively large number of rotons with lower energies which give longer total times.

The detected energy in the total signals is much higher for the roton-atoms than for the phonon-atoms. It is not clear at present whether this is due to there being many more rotons created by the heater than phonons or that rotons are much more efficient at desorbing atoms. Our prejudice is that the latter is true as it seems unlikely that more rotons of 14.3 K energy are created than phonons of 10.7 K energy.

^4He atoms adsorbed onto solids can also be evaporated by phonons²²⁻²⁴ but in this case the phonons are characteristic of the solid and the binding energy is due to van der Waals forces between the atom and substrate. As there is no phonon dispersion, the different energy phonons cannot be distinguished.

From our experiments we can draw several conclusions. The most important is that single phonons and rotons can desorb single atoms with conservation of energy and parallel component of momentum at the liquid surface. We have found very reasonable quantitative agreement between theory and experiment for both the angle of evaporation and the minimum signal time. The results also show that high-energy rotons (~ 14 K) are generated by thin-film heaters and that these rotons can travel through many millimeters of liquid ^4He without being scattered or decaying. It seems likely that these results will enable us to understand the equilibrium between liquid ^4He and its vapor.²⁵

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