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Ultimate fate of a gas of atomic hydrogen in a liquid-helium chamber: Recombination and burial

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In this article we consider the evolution in time of a gas of atomic hydrogen. Gaseous atomic hydrogen is produced in a cell in which the walls are coated with a film of liquid helium to reduce surface recombination. "Stabilized" spin-polarized hydrogen is at best long lived, slowly decaying away by recombination to the molecular state. We describe an experiment which leads to the conclusion that the resultant H_2, either in the form of single molecules or molecular clusters, penetrates the surface of the liquid helium and finally attaches to an underlying wall. The experimental technique developed for this study introduces a new method for producing atomic or molecular clusters at low temperature, either in a helium "background" gas or in liquid helium.

I. INTRODUCTION

Recently, a gas of atomic hydrogen has been experimentally stabilized[1] by polarizing the electron spins of the hydrogen atoms. This was done at low temperatures, $T \lesssim 0.5$ K, and in high magnetic fields, $B \approx 10$ T, so that the Boltzmann factor, $\exp(-\mu B/k_B T)$, where μ is the electronic magnetic moment, overwhelmingly favors the electron-spin-down state. In addition, it was necessary to cover all solid surfaces with a film of liquid helium, the thickness of which was a few hundred Å (some 50 atomic layers) to suppress the otherwise rapid surface recombination. It is now known that a gas of spin-polarized hydrogen ($H\downarrow$) is never absolutely stable, but slowly recombines to the molecular state. In this paper we investigate the fate of these resultant molecules. This subject is interesting independent of the implications for spin-polarized atomic hydrogen, as it deals with many poorly understood phenomena and certainly merits further research. A rather extensive introduction will be presented, in which many of these problems are discussed.

A specification of the state of the mass of molecular hydrogen involves the energy levels and the spatial distribution of the molecular mass. The former involves the internal molecular vibrational-rotational energies as well as the many-body energies due to H_2-H_2 and H_2-He interactions. The latter includes the macroscopic state of the matter, i.e., gaseous, liquid, or solid. Since the temperatures of interest are $T \lesssim 1$ K, the gas and liquid phases are excluded. Microscopically, we are interested in knowing if the hydrogen is clustered together with itself, bound to the helium surface or its underlying substrate, or if it dissolves into the bulk liquid helium. In this paper we do not attempt to answer all of these questions, although a number of interesting results have been obtained experimentally.

Particular attention will be paid to the state of H_2 with regard to the helium film, since experiments have shown[2] that the interaction of $H\downarrow$ with the He surface dominates many properties of the three-dimensional (3D) $H\downarrow$ gas or the two-dimensional (2D) $H\downarrow$ gas bound to the surface. In particular, it controls the density of the 2D and 3D gases, since at very low temperatures recombination and nuclear relaxation between hyperfine states takes place principally at the surface. In addition, the thermal equilibrium of the $H\downarrow$ with the container and thermal bath is controlled by H-He surface interactions, and the gas-liquid interface is responsible for a thermal-boundary Kapitza resistance (R_K) which impedes the flow of recombination energy out of the gas and into a thermal reservoir (refrigerator).[3]

The experimental objective is to determine the final spatial distribution of the molecules, i.e., on, in, or under the liquid-helium surface. If the H_2 is on or mixed into the helium it can strongly affect many physical processes, whereas, if it rapidly sinks under the helium surface and attaches to the underlying substrate, it will no longer influence these processes and the He film, in particular the surface, remains "clean."

A few important processes which H_2 can affect are recombination and nuclear relaxation. Since the minimum of the attractive well for the H-H_2 potential is substantially deeper than that for H-He, if recombined H_2 collects on the surface of the helium, spatial correlations between pairs of H atoms attracted to the H_2 can strongly enhance the surface-recombination or nuclear relaxation rate. The latter rate has been measured to be 30 times faster than expected from theoretical predictions using a binary collision model with free particle correlations. Another situation for which it is important to know the surface condition is for the low-temperature hydrogen maser. It has been estimated that this can have a frequency stability 300 times greater than a conventional room-temperature maser.[4] One of the limits on the stability of the maser is the wall shift. Hydrogen atoms have a short residency time on the surface of the container which gives rise to a wall shift of the frequency. If H_2 builds up on the surface of the helium during operation, the wall shift can change, reducing the stability.

Let us first discuss the energy state of the H_2 molecules. Two unbound H atoms in the spin-polarized ${}^3\Sigma_u^+$ state recombine to the ${}^1\Sigma_g^+$ bound molecular state, ultimately releasing 4.6 eV of recombination energy. The atoms recombine to a highly excited vibrational-rotational state near the continuum level[5,6] and via collisional deexcitation, rapidly cascade down to the ground state. The details of the cascade are poorly understood, but each step releases a few to several hundred kelvins of energy. The ground-state molecules will be either para or ortho, i.e., in the rotational $J=0$ or metastable $J=1$ states, respectively; the final rotational state depends on the initial hyperfine states of the atoms.[5-7] The time τ required for the cascade is estimated to be $\tau \lesssim 20$ msec from experiments in which a gas of $H\downarrow$ is triggered to recombine, and the time required for the recombination energy to flow out of the system is measured using microcalorimetric techniques.[8,1] Thus for our purposes we can consider the H_2 to be either ortho or para in the ground vibrational state. The spatial distribution of the H_2, which is the experimental objective of this paper, still remains to be determined.

In the following section I shall discuss a number of properties of H and H-He considered to be important for the problem at hand. In Sec. III I will describe the experiment performed and in Sec. IV I will discuss the results and draw conclusions.

II. RELEVANT PROPERTIES OF HYDROGEN AND HELIUM

A. Interaction potentials

The H atom has four hyperfine states which are shown in Fig. 1. Spin-polarized hydrogen is a mixture of atoms in the $|a\rangle$ and $|b\rangle$ hyperfine states.[2] Note that state $|b\rangle = |\downarrow\Downarrow\rangle$ ($\downarrow$ is the electron-spin projection and $\Downarrow$ is the nuclear-spin projection) has both the electronic and nuclear-spin projections down and is a pure Zeeman state. In contrast, state $|a\rangle \approx -|\downarrow\Uparrow\rangle + \alpha|\uparrow\Downarrow\rangle$ has an admixture α of the reversed electron-spin state. For a field $B=10$ T, $\alpha \approx 2.54\times10^{-3}$, so that the admixture is quite small. A gas with greater than 99% of the atoms in the $|b\rangle$ state can also be produced.[9,10] In this gas, called double-polarized hydrogen ($H\downarrow\Downarrow$), both the electronic and nuclear spins are polarized.

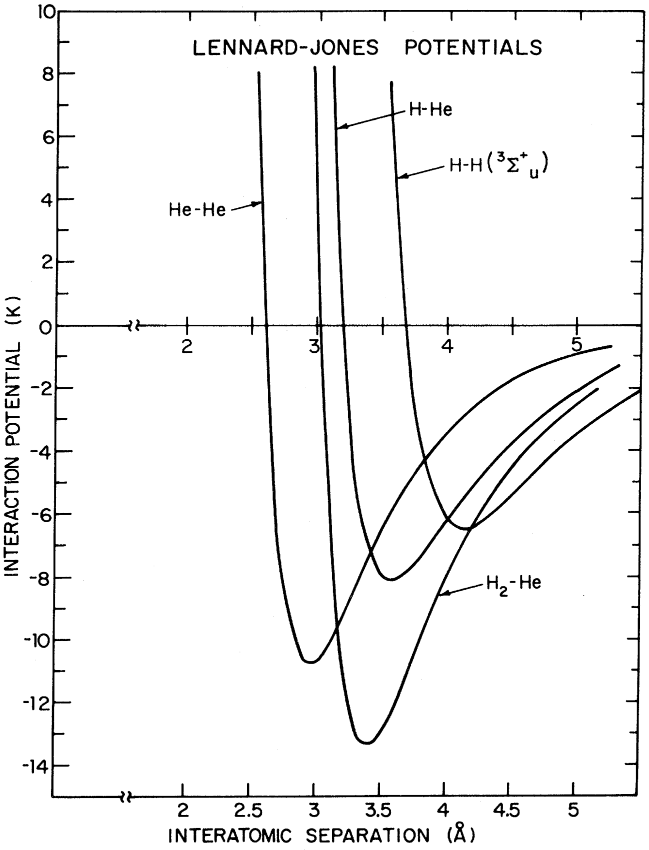

FIG. 2. Lennard-Jones potentials of some weakly interacting pairs. The parameters are given in Table I.

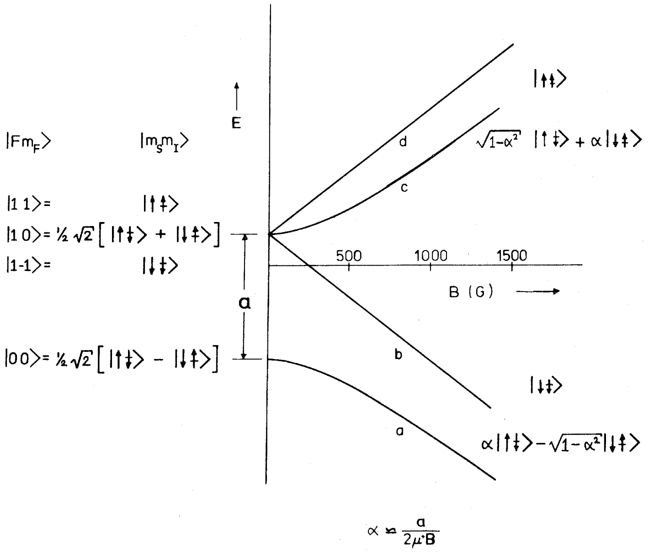

FIG. 1. Energies of the four hyperfine states of atomic hydrogen in an applied magnetic field B. The levels are eigenvalues of the Hamiltonian $H=(g_e\mu_B\vec{S}+g_N\mu_N\vec{I})\cdot\vec{B}+a\vec{I}\cdot\vec{S}$, where $\vec{S}$ and $\vec{I}$ are electron and nuclear spins $\frac{1}{2}$, g_e, and g_N are the electron and nuclear g factors, μ_B and μ_N are the electron and nuclear Bohr magnetons, and a is the hyperfine constant. Also, $\mu^{\pm}=g_e\mu_B \pm g_N\mu_N$.

For this study it is important to know the interaction potentials of hydrogen and helium. The weakest of all is H-H on the $^3\Sigma_u^+$ potential.[11] Next comes H-He,[12] then He-He,[13] and finally H_2-He.[14] In Fig. 2 we show these po-

TABLE I. Lennard-Jones well (ϵ) and range parameters (σ) and the quantum parameter $\eta=\hbar^2/2M\epsilon\sigma^2$ for pairs of interest in this article. These parameters have been adapted from more precise potentials given in the references.

	ϵ (K)	σ (Å)	η	Ref.
H-H $^3\Sigma_u^+$	6.46	3.68	0.547	11
H-He	8.10	3.20	0.363	12
He-He	10.8	2.65	0.159	13
H_2-He	13.34	3.03	0.148	14
H_2-H_2	34.3	3.04	0.0761	17

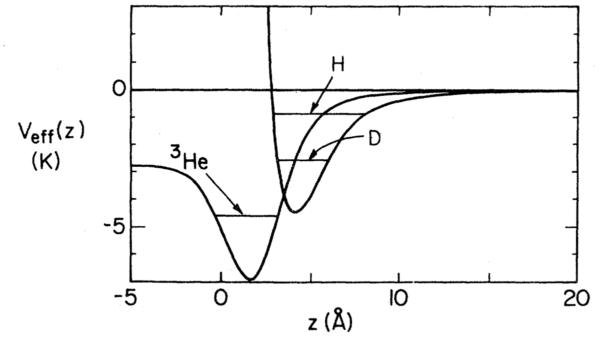

FIG. 3. Surface adsorption potentials of H, D, and ^{3}He on ^{4}He (after Ref. 21) with the bound states $\epsilon_a/k_B = 0.89$, 2.6, and 4.64 K, respectively.

tentials as represented by the Lennard-Jones form, $V = 4\epsilon[(\sigma/R)^{12} - (\sigma/R)^6]$. Here, ϵ is the well depth and σ is the range R at which $V = 0$. The Lennard-Jones parameters we have used are given in Table I. We also give values of the modified de Boer parameter,[15] or quantum parameter, $\eta = \hbar^2/2M\epsilon\sigma^2$, where M is the reduced mass. The quantum parameter gives a measure of the importance of zero-point energy relative to potential energy. For example, H-H with a large value of η has no bound states; nor does H-He. He-He may have one bound state[16] (from studies on more precise forms of the potential) with dissociation energy $D \ll 1$ K; however, the He-He dimer has never been observed. From this scaling parameter and the law of corresponding states[15] we can predict that H_2-He may have a bound state. We also give the potential parameters[17] of H_2-H_2 which is known to form a bound dimer state[18] with a dissociation energy $D/k_B \approx 3.5$ K.

The situation is quite different if we consider the interaction of atoms with a ^{4}He surface. In Fig. 3 we show potentials of H, D, and ^{3}He on ^{4}He. H is weakly bound to the ^{4}He surface with a measured adsorption energy $\epsilon_a/k_B \approx 1$ K.[19,20] We show the theoretical potential of the H-^{4}He surface due to Mantz and Edwards.[21] According to Miller,[22] the chemical-potential difference of H replacing a ^{4}He atom in liquid helium is 42 K, so that H↓ does not penetrate the surface, behaving as an almost perfect 2D gas. It is speculated that the heavier D↓ which is bound by $\epsilon_a/k_B \approx 2.6$ (Ref. 23) can dimerize in two dimensions.[24] An interesting case in which an impurity atom with a bound surface state dissolves into the bulk liquid is ^{3}He on the ^{4}He surface. In Fig. 3 we show the relevant potential from Ref. 21.

A single H_2 molecule will certainly be bound to a many-body He system at $T = 0$ K. We can only speculate as to whether it will be confined to surface states or if it can also penetrate into the liquid. If its chemical potential (difference for replacement of an He atom) is negative, it will penetrate the bulk. Ristig *et al.*[25] have recently made a theoretical study of impurities in helium. In a preliminary unpublished calculation[26] they find that H_2 can penetrate the helium surface. Even if this were not the case, then single H_2 molecules could reside in He surface states. Then, dimers, trimers, etc., of H_2 could form on the surface, and these might be able to penetrate into the liquid helium since their kinetic energy and effective quantum parameters are smaller than those of the monomer.

B. Recombination

Recombination requires the collision of two H atoms in the presence of a third body X ($H + H + X \rightarrow H_2^* + X^*$), which can also be an H atom. The rate of decay of a volume density n_i of H atoms in the ith hyperfine state is

$$\frac{dn_i}{dt} = \sum_j -K_{ij}^{v} n_i n_j n_x \,, \tag{1}$$

where n_i and n_j are densities in the i and j hyperfine states, and n_x is the density of species X. K_{ij}^{v} depends, in general, on temperature, magnetic field, and the initial states of the collision partners.

Atoms also physisorb on the helium surface with a surface density n_{si} in state i; the dominant recombination decay rate of this density is

$$\frac{dn_{si}}{dt} = \sum_j -K_{ij}^{s} n_{si} n_{sj} \,. \tag{2}$$

Here, the surface plays the role of the third body. We shall confine our attention to the helium surface, in which case adsorbed hydrogen forms a 2D gas with a single bound state with adsorption energy ϵ_a.[27] In thermodynamic equilibrium the relation between gas and surface density is, for low density,[27]

$$n_{si} = n_i \lambda \exp(\epsilon_a / k_B T) \,, \tag{3}$$

where $\lambda = (2\pi\hbar/mkT)^{1/2}$ is the thermal de Broglie wavelength. The adsorption energy ϵ_a/k_B of H on He has been measured to be about 1 K.[19,20]

Experimentally, at low temperature, surface recombination has been found to dominate the volume term for densities $n \lesssim 1 \times 10^{18}$ cm^{-3}. Clearly, from Eq. (3) the surface-coverage term will generally be dominant at low temperature since coverages grow exponentially with T^{-1}. Surfaces of ^{3}He or ^{3}He-^{4}He mixtures have a value of ϵ_a about a factor of 3 lower than for ^{4}He.

C. Spatial distribution of H_2

The suggested picture at this point is that H↓ recombines to form H_2 which interacts with the helium surface. Single molecules of H_2 [or N-mers, $(H_2)_N$] can penetrate into the bulk helium. The H_2 is not dissolved in a thermodynamic equilibrium state; if it collides with another molecule or N-mer, it can combine to form a larger cluster. If it strikes the container wall it will physisorb to lower its energy. In this way the walls become solid hydrogen.

Even in a wall-free situation such as a spherical drop of liquid helium in gravity-free space, H_2 molecules diffusing in the interior would eventually collide and form H_2 clusters until there was a complete phase separation of solid H_2 enclosed in liquid helium.

If large clusters of H_2 are formed in a gravitational field, the buoyant forces will be important and the cluster will float up to the surface of the denser helium. Just the opposite effect is expected for solid D_2, which is more dense than helium.

III. EXPERIMENT

The ideal experimental condition to determine the state of the H_2 is one in which a gas of H is produced in a helium-covered chamber at subkelvin temperatures. As the hydrogen recombines, the state of the H_2 is monitored as a function of time. The experiment is greatly simplified if we ignore the path by which H_2 gets to its ground energy state and concentrate on its spatial distribution. As already discussed, when H recombines, the H_2 rapidly cascades down to its ground internal molecular state, thermalizes, and then forms an aggregate many-body state. From this point of view, to determine the H_2 spatial distribution, we can omit the step of producing H and just introduce H_2 from an approximately thermal low-temperature source into a helium chamber. A temperature, $T \lesssim 1$ K, of the helium chamber should be adequate. This temperature is somewhat lower than the expected characteristic temperatures of the bound states. Finally, since H is usually studied in the presence of saturated He films, a much thicker layer of helium should not affect the surface physics.

Our experimental apparatus is shown in Figs. 4 and 5. A double-walled liquid-helium glass cryostat with strip windows in the opposing silvered walls is used. The liquid helium can be pumped down to a temperature of about 1.1 K. A Pyrex glass tube (10 mm diam) with a flat bottom plate is suspended in the superfluid liquid helium. At the bottom of the tube, a small disk with a 4-mm-diam hole in the middle defines a small cylindrical chamber. A second Pyrex tube of 3.5 mm o.d. and 2 mm i.d. protrudes a few millimeters through the hole in the disk without touching the walls (to prevent superfluid flow of liquid He into the tube). This tube, connected to an external gas manifold, is used as a molecular expansion source of H_2 or D_2. Since the source tube will normally cool down to the temperature of the helium and plug up with solid H_2 or D_2, a tightly wound spiral wire heater is pressed to the inner walls of the lower part of the tube. When operated as an expansion source, H_2 is first condensed onto the walls of the tube; the flow rate out of the bottom is controlled by regulating the electrical power dissipated in the spiral heater.

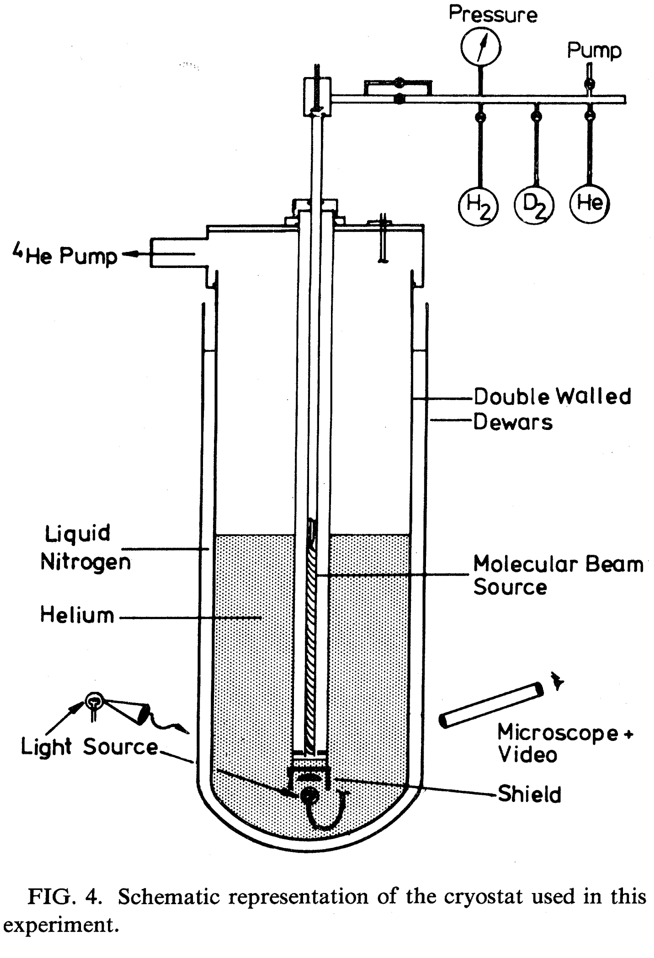

FIG. 4. Schematic representation of the cryostat used in this experiment.

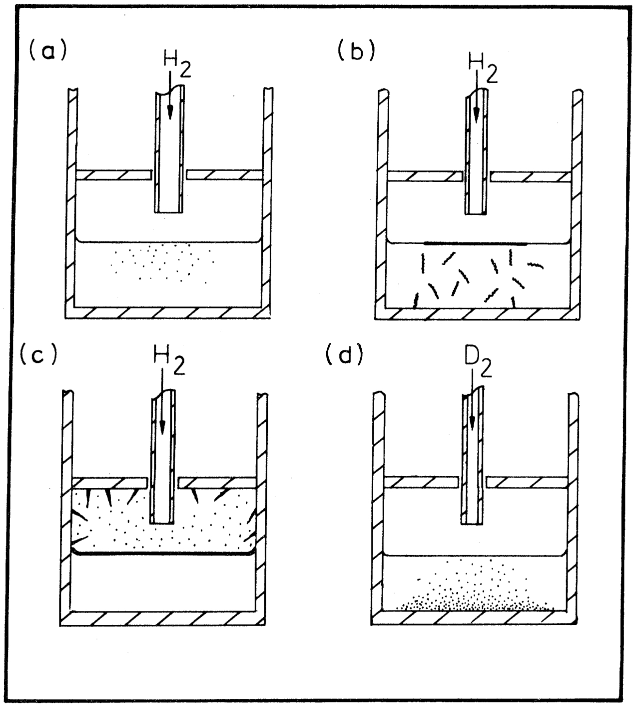

FIG. 5. Results of visual observations: (a), (b), and (c) H_2 flowing into liquid helium under low-, medium-, and high-flow conditions, respectively; (d) D_2 flowing into liquid helium.

The cylindrical chamber is filled with about 5 mm of liquid ^{4}He which is cooled to about 1.1 K by contact to the outer walls. The surface was usually 2–5 mm below the molecular expansion source. The experimental intent is to visually observe the region around the gas-liquid interface while scattering H_2 molecules from the source off of the ^{4}He surface. This region is viewed by means of a long-focal-length microscope and is illuminated by two lamps. An external light beam is incident at an angle approximately equal to the viewing angle of the microscope so that the surface of the liquid helium is accentuated by the reflected light. Without this the surface is quite difficult to see, except at the tube wall, because of the small index of refraction of the liquid helium. The second light source is a collimated beam from a flashlight bulb located in the liquid-helium bath under the cylindrical chamber. It is shielded by an opaque screen so that only upward shining light scatters into the microscope. This provides a dark-field illumination for any macroscopic clusters that form in the helium, enabling aggregates with dimensions on the order of micrometers to be observed.

It is difficult to characterize the flow out of the molecular source. First of all, we could not quantitatively determine the flow rates, but could only characterize them as low, medium, or high, depending on the heater power. Second, a beam source or supersonic expansion normally flows into an environment with a low background pressure; in our case, there is a dense helium background gas above the liquid, of stagnation pressure 0.3—0.6 Torr and density $3-5\times10^{18}/\text{cm}^3$. The collision mean free path with H_2 is estimated to be roughly 2 μm. The helium also penetrates into the source tube. When the source heater is turned on there is an initial transient (2—3 sec) depression of the liquid-helium surface. This is due to the flow of the initially cold helium gas out of the source tube. This is not completely thermalized by the background gas and transfers flow momentum to the helium surface. Under steady-state conditions, with flowing H_2 or D_2, a slight depression of the He surface could sometimes be observed. In this case the reflected light from the external lamp beam was particularly useful.

Presumably, the H_2 flowing out of the tube has some mean energy corresponding to $T\approx15-20$ K. It interacts with the background gas and may approach the temperature of the stagnant helium. This implies that the H_2 flow may consist not only of H_2 molecules, but also of H_2 clusters. The percentage of the mass that is clustered would depend on the distance to the surface.[28]

The results of these experiments are restricted to visual observations and are shown in Fig. 5. Under low-flow conditions of H_2, after a few minutes, a swarm of tiny clusters, estimated to have linear dimensions on the order of micrometers, are observed in the interior of the liquid helium. These remain suspended in the interior while flowing, exhibiting Brownian-type motion. Presumably, the visible clusters are being scattered by invisible microscopic clusters. When the flow terminates, they rise to the surface, coagulate, and eventually attach themselves to the tube walls forming a ring at the meniscus. After a long flow period under these conditions, some solid H_2 buildup can be seen on the glass surface below the helium surface level.

At medium-flow conditions two phenomena occur. At the He surface, a thin floating disk-shaped island of H_2 forms, stabilized in position under the source by the flow. Inside the liquid, long (a few millimeters), wormlike, approximately vertically oriented clusters with diameters estimated to be tens of micrometers form. When the flow is terminated the "worms" rise to the surface unless they touch the Pyrex bottom where they remain stuck. The thin disk is attracted to the side wall where it coagulates and sticks.

Under high-flow conditions it is possible to set up a miniature whirling snow storm. Visible clusters grow in the gas-phase helium, forming large whirling eddies. Icicles start growing out of the ceiling and walls above the liquid-helium surface.

Molecular deuterium has also been studied. Under low-flow conditions again, tiny clusters form in the interior of the liquid, but these sink to the bottom Pyrex surface, since solid D_2 is denser than helium. Under higher-flow conditions larger clusters form, which also sink to the bottom surface.

IV. DISCUSSION

These experiments provide direct visual evidence that H_2 or D_2 can penetrate into liquid helium. By the time the H_2 hits the He surface, it is probably in the form of both single molecules and clusters. There is no direct proof that a single molecule of H_2 penetrates the surface although this idea has theoretical support. This could probably be obtained by a Raman scattering study of the rotational spectrum of H_2 in the interior of liquid helium.

One might question whether any single molecule ever reaches the He surface or if the H_2 from the source is completely clustered. This would depend on the source-surface distance.[28] This distance has been varied continuously from ~0 to 5 mm without any perceptual difference in the results.

Under low-flow conditions, H_2 certainly penetrates the helium surface in the form of clusters. With increasing flow, the growth of H_2 in the form of a disk takes place at the surface and remains there due to buoyant forces. In the case of experiments with spin-polarized atomic hydrogen in the presence of superfluid films of order 10^2 Å thick, at the current rates of production of H_2 due to recombination of $H\downarrow$, the molecules or clusters which penetrate the film surface will have a higher probability of colliding with the wall and attaching than they will of encountering other clusters and uniting to form an aggregate which can float to the He surface. However, if a high-density gas of $H\downarrow$ is triggered into rapid recombination so that a large flux of molecules is produced, it may be possible that large clusters are formed on the surfaces of an atomic hydrogen cell. In any event, this solid H_2 will then still be covered with a superfluid helium film. We also note that if only $(H_2)_N$ $(N>1)$ clusters can penetrate the He surface, this implies that in the presence of recombining $H\downarrow$ there will be a steady-state density of H_2 on the surface, determined by the H_2 formation rate and the depletion rate due to clustering.

These ideas have implications for the attempt to achieve Bose-Einstein condensation in $H\downarrow$ at high densities and low temperatures. A very serious problem will be the removal of the recombination heat generated in the gas. Calculations[3] indicate that the thermal boundary resistance between the gas of $H\downarrow$ and the He surface varies as $T^{-7/2}$. For $T\approx10$ mK the time constant to achieve thermodynamic equilibrium, R_KC, where R_K is the thermal resistance and C the heat capacity of the gas, will be many hours. If excited molecules can directly penetrate the helium surface, then a large portion of the recombination energy could be directly transferred to the liquid helium. This heat can be removed from the liquid by using a large-surface-area sinter.

In this article a number of new research problems are suggested. Most important is the theoretical and experimental determination of the binding energy of H_2 to a helium surface. The latter is particularly difficult to measure, since surface H_2 will be unstable and either penetrates the bulk as a single molecule or as a cluster.

In a totally different area of physics, that of supersonic expansions, clusters of particles, dimers, trimers, and N-mers, are formed in the expansion. This interesting area of research has been studied intensively in the past decade. In this technique, due to the cooling in the isentropic expansion, collisions result in the formation of clusters. Normally, the background gas in the expansion chamber is at room temperature and the interaction of the expansion clusters with the background gas thermalizes the expansion, destroying the clusters. The present experiments suggest a new technique for forming clusters: expansion of a gas X into a cold helium vapor. Here, *thermalization gives rise to cluster formation:* He-X collisions cool off the translational degrees of freedom of X. The particles can then condense to form clusters of the X species, or He-X. These can be studied either in the gas or liquid phase of helium. Indeed, they may even provide an interesting probe of the properties of liquid helium.

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[28] If the H_2 source is a supersonic expansion, then massive clustering is expected to take place after a few source diameters "downstream" of the expansion tube. [See, for example, H. P. Godfried and I. F. Silvera, Phys. Rev. A 27, 3019 (1983)]. More likely, in our case, the clustering takes place in the region of beam thermalization to $T\approx1.2$ K. For a supersonic expansion this occurs at the so-called Mach disk. Owing to the complex nature of our system it is difficult to reliably estimate its position. In any event, the depression of the He surface indicates that the expansion is not completely thermalized.