THE STABILIZATION OF ATOMIC HYDROGEN : A NEW BOSE GAS

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Résumé.- Dans les circonstances habituelles, un gaz atomique d'hydrogène (H) est très instable car il tend à former par recombinaison des molécules d'hydrogène (H_2). Nous avons réussi à stabiliser un gaz d'hydrogène à forte polarisation de spin (H↓) à T = 300 mK dans des champs magnétiques atteignant 10 Tesla, dans une cellule à parois recouvertes d'hélium superlfuide. Nous avons obtenu des densités atomiques de 10^{16} cm^{-3}, et observé que le gaz H a une durée de vie surprenante par sa longueur, même en champ magnétique nul. Nos résultats tendent à indiquer que H↓ reste sous forme gazeuse, sans se liquéfier ou se condenser sur les surfaces de façon appréciable. On discute les aspects cruciaux des techniques expérimentales pour stabiliser H↓, ainsi que quelques unes des propriétés observées ou attendues de ce nouveau gaz de Bose.

Abstract.- Under normal circumstances a gas of atomic hydrogen (H) is highly unstable with respect to recombination to molecular hydrogen, H_2. We have succeeded in stabilizing a gas of spin polarised hydrogen (H↓) at T = 300 mK in magnetic fields up to 10 Tesla in a superfluid helium coated cell. Densities of at least 10^{16}atoms/cc have already been achieved. H is found to have a surprisingly long life even in zero magnetic field. Evidence points to H↓ remaining in the gaseous form and not liquifying or condensing out on the surfaces to any large extent. The crucial experimental techniques used to stabilize H↓ will be discussed as well as some of the observed properties and expectations of this new Bose gas.

In the past half century low temperature physicists have been fascinated by the unravelling and elucidation of the many marvelous properties of the quantum fluids ^{4}He and ^{3}He. In this paper we shall discuss our recent experiments in making new quantum fluids: atomic hydrogen and deuterium /1 - 3/.

First many of the prospective exciting properties of these gases will be briefly discussed. We shall then describe the techniques and difficulties involved in producing the gases, and finally the possibilities of achieving high density at low temperatures to observe the effects of quantum degeneracy. Although most of the discussion will be devoted to atomic hydrogen, where differences occur, appropriate comments will be made about deuterium.

Since the majority of the participants of this conference come from a helium background it is useful to make some comparisons.

Upon cooling a gas of ^{4}He at 1 atmosphere pressure at 4.2 K, it becomes unstable to condensation and liquifies. Below 2.17 K it becomes a superfluid. Since the early work of London /4/ it has been believed that this superfluidity is intimately related with the phenomenon of Bose-Einstein condensation (BEC), i.e. the macroscopic population of the ground state at a finite temperature. This is most easily demonstrated for the weakly interacting Bose gas. However for helium a serious problem arises in the theoretical analyses. In the liquid state the mean He - He distances are so small that the effects of He-He interactions play a dominant role in the hydrodynamic phenomena: helium is a strongly interacting boson superfluid, with inherent theoretical difficulties. This problem probably will not exist for atomic hydrogen.

Atomic hydrogen (H) has been stabilized in a special state called spin polarized hydrogen (H↓) /5/. In this state all hydrogen atoms interact pairwise on the almost repulsive $^3\Sigma_u^+$ potential, shown in fig. 1, and thus do not form bound states.

As discussed by Nosanow /6/, due to the weak interactions and light mass, H↓ will not form a many-body bound state at T = 0 K, i.e. as a result of the large zero-point energy H↓ remains a gas to T = 0 K. Hydrogen is predicted to have a solid phase for pressures P $\gtrsim$ 50 bar. As a consequence, ignoring the problems of recombination to H_2, at T $\approx$ 0 K the density of H↓ can be varied almost arbitrarily, so that at low densities it will behave as a weakly interacting gas.

Since H has an electron spin of ½ and a nuclear spin of ½ it is a spin 1 or 0 particle, i.e. a composite boson. H↓ will thus enable a study of the weakly interacting Bose gas and the relation between BEC and superfluidity. Since D↓ has an electron spin ½ and nuclear spin 1, it is a composite fermion. It is expected that this will provide an ideal system for the study of superfluidity due to fermion-pairing /7/. Since the mass of D is twice that of H it may be a liquid at T = 0 K, although theoretical predictions have some uncertainty.

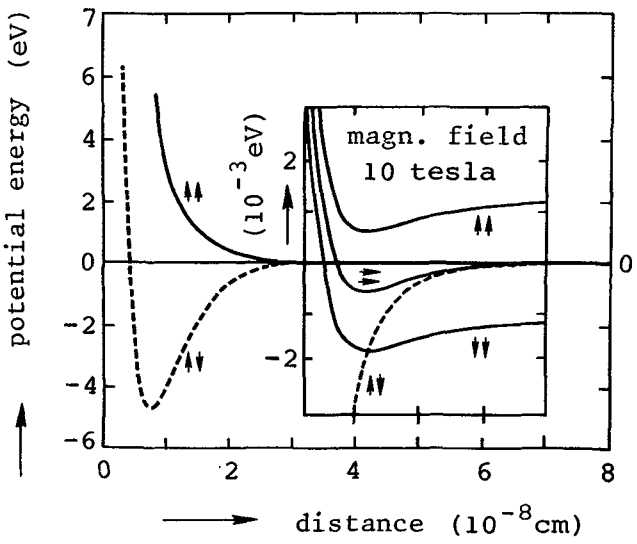

Fig. 1 The interatomic interaction potentials for singlet $^1\Sigma_g^+$ (dashed curve) and the triplet $^3\Sigma_u^+$ (solid curve) hydrogen as calculated by Kolos and Wolniewicz. The effect of a magnet field is shown in the inset on a scale which is expanded by approximately 2000.

Dynamical properties of H↓ in the Bose condensation phase are predicted to display many new phenomena. Berlinsky /8/ has predicted an interesting density-spin excitation spectrum and Siggia and Ruckenstein /9/ have conjectured that the slow nuclear relaxation rate enables the occurrence of two Bose condensates, distinguished by the nuclear spin states. They predict that coherent nuclear relaxation occurs and may be observed as spontaneous emission of rf radiation. Spatial localization of the condensate in a magnetic field gradient has been suggested as a characteristic of the Bose condensed gas at low densities where interactions are unimportant /10/. Some possible practical applications of H↓ are as an improved maser for time and frequency standards, a cryogenic refrigerant, and as an exotic form of chemical fuel used as a rocket propellant. Although this latter application seems somewhat remote at present, one can imagine this as a fuel for future space travel, with spaceships gathering their fuel with huge collectors as they travel through intragalactic clouds of H.

Recombination

The stable form of a gas of hydrogen atoms is with all atoms pairwise bound in the molecular form. This is the lowest energy state for the $^1\Sigma_g^+$ potential of fig. 1. A gas of H↓ can get to this state by three-body recombination processes, H↓ + H↓ + X → H_2^* + X* where X is a third body, including surfaces, and the * indicates an excited state. The rate of decay of a density n of H is governed by the rate equation

$$-\frac{dn}{dt} = K_v\, n^3 + K_s'\, n_s^\delta \qquad (1)$$

where K_v is the volume recombination rate, K_s' the surface rate and n_s the surface coverage with δ = 1 or 2. Since in general $n_s \sim n$, we can write $K_s'\, n_s = K_s\, n$. K_v is known for the process H + H + He → H_2^* + He* : at room temperature $K_v = 1.2 \times 10^{-32}$ cm^6/atom-s /11/, with a rate about

1/5 this at about 1 K /12/. At room temperature a density of $n \sim 10^{16}$ at/cc would have a lifetime of about 1 sec. The surface term is less well known. At room temperature H can have many thousands of bounces off of a teflon surface before recombining. Metal surfaces have a probability of order one for recombination upon collision. Since H will have long range attractive interactions with all surfaces, at temperatures low with respect to the surface adsorption energy, ε_a, H will condense, rapidly building up the surface coverage and recombining due to the second term in eq. (1). In stabilizing H↓, this is the most important term to control. With "normal" surfaces at low temperatures the lifetime of a gas of H↓ is of the order of tens of microseconds. We have found that by covering all surfaces with a film of ^{4}He, surface recombination is strongly suppressed, since ε_a/k_B is probably less than 1K /13/. The rapid recombination rate on helium free surfaces provided us with a sensitive technique for detecting the presence of the H↓, as shall be described.

Experimental Stabilization of H

Our experimental work on atomic hydrogen started in 1972. At that time we believed that it would be difficult to collect and confine large quantities of stable atomic hydrogen and such studies might best be carried out on surfaces. Atomic beams of H could be made with axial fluxes of $\sim 10^{14}/\mathrm{cm}^2/\mathrm{sec}$ which corresponds to about one monolayer/sec if deposited on a substrate. Clean, well characterized substrate surfaces of closed shell atoms or molecules (Ar, H_2, N_2 etc.) - insulators - could be deposited or grown in situo. In this case the hydrogen would be physisorbed and remain as electrically neutral atoms. Because H is a composite boson, if it was in a sufficiently mobile state on the surface, one could study the possible superfluidity of H films. To prevent the surface recombination the deposited H would have to be spin-polarized so that atoms would interact via the non-binding $^3\Sigma_u^+$ potential of fig. 1. An apparatus was built to polarize a beam of hydrogen and déposit this H↓ on a cold substrate ($T \approx 2$ K) in a moderate magnetic field. After a substantial build-up period experiments were performed, with very discouraging results. The H↓ apparently rapidly depolarized on the surface followed by recombination or desorption /14/. After some consideration /15/ it was decided that the temperature and magnetic field were inappropriate for stabilizing H. Furthermore we developed a rather negative propensity toward the surface due to its continual presence as the third body in recombination processes. We decided to lay down the attempt to stabilize H on surfaces and build an apparatus for stabilization of a three dimensional gas. We felt that the recombination processes could be better controlled if we could prevent the H↓ from condensing on the surfaces. A description of this approach and apparatus follows.

In order to stabilize H↓ , magnetic fields and low temperatures are required. This is most easily seen from fig. 1. First of all we see that in zero field the $^1\Sigma_g^+$ state is lower in energy than the $^3\Sigma_u^+$ for all ranges. Thus as $T \to 0$, the gas can lower its energy by reversing spins which leads to subsequent recombination. A magnetic field can split the $^3\Sigma_u^+$ level so that the $m_s = -1$ state lies below the singlet state for part of the range. This leads to what is called static stability /15/, i.e. in a given field the gas is metastable below a certain density. The gas will still decay to H_2 by three-body recombination but at a reduced rate in the field. The low temperature is also required so that the equilibrium Boltzmann population of the reversed spins will be negligible in the applied field.

Experimental Techniques

The gas of H↓ was stabilized in the apparatus shown in fig. 2. A hydrogen stabilization cell (HSC) sits in a solenoidal magnetic field of maximum value 11 Tesla. The HSC can be cooled to ∿ 270 mK by a ^{3}He evaporation refrigerator. Atomic hydrogen arising from a room temperature microwave discharge is guided into the cryogenic environment by teflon lined tubing and, before entering the HSC, is cooled to ∿ 4.2 K by contact with the H_2 covered walls of a copper accomodator /16/.

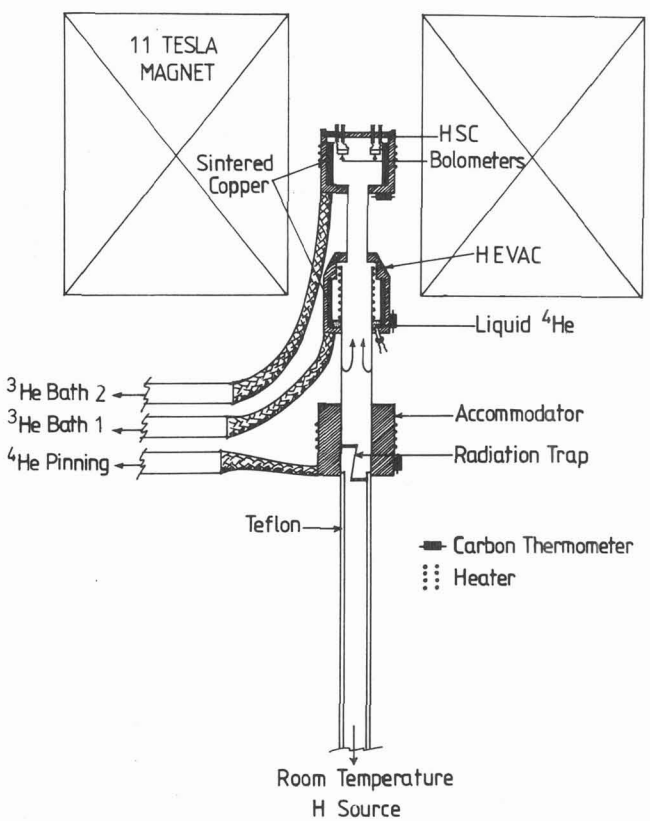

Fig. 2 The low temperature part of the apparatus used for stabilizing atomic hydrogen.

The separation of the electron spin-up and spin-down states is performed by the magnetic field gradient that exists at the entrance of the solenoid. The gradient field exerts a force on the atomic magnetic moments so that the atoms in spin-up states are repelled and the spin-down states are attracted into the field center. Since the atoms are confined by the guide tube, the spin-up atoms either recombine on the walls or relax to spin-down and are drawn into the field. The atoms in the HSC come into thermodynamic equilibrium with the cell walls and are confined to the field center by the magnetic gradient /10/. The atoms are prevented from condensing on the surfaces by covering all walls with a saturated film of ^{4}He which is a superfluid at these low temperatures. Although this prevents wall recombination it leads to a serious cryogenic problem: due to the fountain effect, the film flows towards warmer regions, in this case the accomodator: The heat flux from the warmer regions of the guide tube evaporates the film; a pressure gradient drives the ^{4}He gas back to the colder HSC where it condenses to liberate its heat of condensation and warm the cell. To suppress this heating we built an intermediate ^{3}He refrigeration stage called a HEVAC. The fluxing ^{4}He vapors condense out on the HEVAC; residual fluxing vapors are of such low density that they do not present a serious cryogenic problem.

The HEVAC which stands for helium vapor compressor serves a second role. It is actually a miniature vapor diffusion pump, the fluxing ^{4}He imparting its momentum to the H gas and confining it to the cell. It operates with a compression c_H ($1/c_H$ is the probability for a particle entering the HEVAC to escape) of approximately 50.

With this system the HSC can be loaded with a gas of H↓ that is stable and confined for long

periods of time. The presence of the gas was detected with specially developed bolometer detectors made of carbon composition from a Speer resistor for which the resistance drops rapidly with increasing temperature. Two such bolometers are suspended in the HSC by fine wires as shown in fig. 2. The idea is to selectively cause the $H\downarrow$ to recombine and determine its presence by measuring the warming due to the liberated recombination energy. Under normal conditions the bolometers are covered by the film of ^4He so that they are inert with respect to the $H\downarrow$ gas. By passing a sufficiently large current through a bolometer, the ohmic heating will cause the film to evaporate more rapidly than it can be replenished along the wires. When the surface is clean of ^4He, the $H\downarrow$ condenses and rapidly recombines. The surface of the bolometer is probably covered with H_2 which has an adsorption energy for H of 38 K /17/, or other molecules (O_2, N_2, H_2O etc.) which have even stronger adsorption energies. We expect that for a gas of $H\downarrow$ with $kT \ll \varepsilon_a$ there will be a probability of about 1 that an atom that collides with the surface will stick.

To determine the time constant for recombination of the $H\downarrow$ after "triggering" by heating the bolometer, we assume that the rate limiting process is the thermal effusion of atoms to the bolometer surface of area A_b. Then solving the equation $dN/dt = \frac{1}{4} n\bar{v}A_b$ where N is the total number of atoms and $\bar{v}$ the average velocity gives $N = N_0 \exp(-t/\tau)$ with $\tau = 4\, V_{eff}/\bar{v}A_b$ where V_{eff} is the effective volume of the gas $H\downarrow$ (10). Typical values of τ are 10 - 20 msec /1/.

When the atoms recombine to H_2 on the bolometer surface only a fraction of the recombination energy is delivered to the bolometer /14/. Most likely the molecules desorb in a highly excited vibration-rotation state, also desorbing ground state H_2 molecules from the first layers of the bolometer under the helium. This energy is mostly adsorbed in the walls of the cell; part of it is probably blown out of the HSC through the HEVAC. Although the bolometer is an extremely sensitive $H\downarrow$ detector and can probably detect as little as 10^8 atoms, it is a highly non-linear detector, less suited for density measurements of the $H\downarrow$. To measure the density we actually measure the temperature rise of the HSC with a (Speer) thermometer attached to its wall. This has a slightly slower response than the bolometer and is less sensitive, but well matched to our densities. It can be calibrated with electrical heat pulses. It has the rather nice feature that its response, as monitored by the resistance change of the Speer resistor, is linear with density. There are two shortcomings to this technique. First it only provides a lower bound to the density since some of the atoms or available energy can escape out of an open ended system. We shall see the problem this can create at higher densities later. Second, it is a destructive technique - to measure the density, the gas of $H\downarrow$ must be destroyed. As a result measurements of time dependent effects are extremely tedious. A non-destructive type measurement would be very useful. At densities of 10^{15} - 10^{16} at/cc one can use a capacitance manometer to measure the pressure, which is proportional to the density for low densities. We have already used the bolometers as Pirani gauges to non-destructively detect the $H\downarrow$ gas by measuring a signal proportional to its thermal conductivity. This should provide a linear detector up to densities where the mean free path is of order of the dimensions of the cell. Other possible techniques are NMR, ESR, viscosity, dc magnetization etc. The bolometer has proven to

be a vital link in the initial stabilization of H↓. It will eventually be replaced by non-destructive detectors for studies at high densities, but will remain as an easy to operate diagnostic tool.

When triggered the shape of the heating pulses are of some interest. In fig. 3 we show the response of the cell thermometer for a low density of H↓.

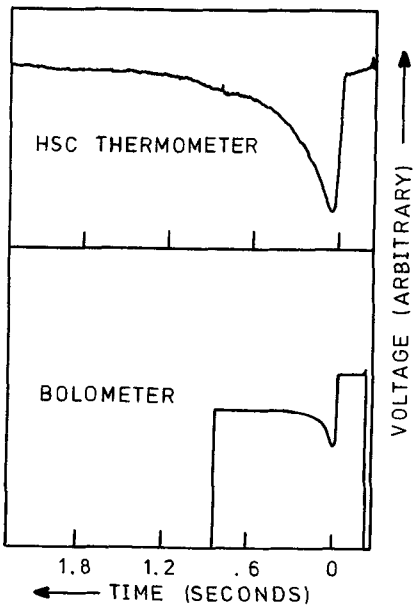

Fig. 3 The response of the HSC thermometer and the bolometer used to trigger the recombination of hydrogen. The bolometer is heated with a current square wave. The voltages are proportional to the resistances. Long after the H↓ is burned, the bolometer approaches a lower value of resistance than before the triggering because the helium film is broken. The voltage of the thermometer has a zero offset.

This was recorded on a fast response (100 Hz) strip chart recorder. In fig. 4 we show the response (on a slow recorder ∿ 5 Hz response) of the secondary bolometer in the cell, after triggering by the other bolometer. This bolometer receives its heat primarily by being struck by recombined molecules that desorb from the primary bolometer.

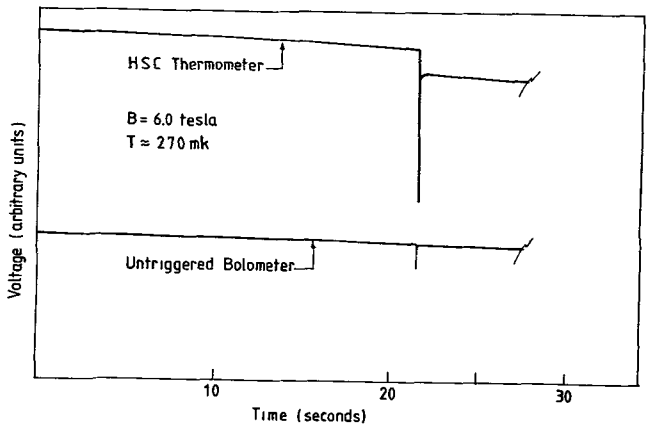

Fig. 4 Voltage responses of the HSC thermometer and the secondary (untriggered) bolometer. Due to ohmic heating after the triggering, the HSC voltage remains low.

Evidence for the gaseous nature comes from the (non-quantitative) thermal conductivity measurements. Furthermore time constants are in rough accordance with what would be expected for a gas. Theoretical calculations of adsorption isotherms /18/ also suggest that very little H↓ would be condensed on the surfaces. If the trigger signals arose only from H↓ adsorbed on the bolometers, then with a maximum surface coverage of order 10^{14} at/cm^2, one would be unable to explain the size of the observed signals.

Finally experiments were performed in which bolometers 1 and 2 were rapidly sequentially heated. Only the first bolometer heated resulted in a recombination signal. If the H↓ were predominantly in an adsorbed state we would expect signals from both bolometers.

Measurements

In fig. 5 we show the results of the first measure-

ments on H↓ /1/. At the time of these observations the most important questions were: could a gas of H↓ be stabilized and for how long. In these experiments the magnetic field was set to 7 Tesla and the temperature was 270 mK. The cell was filled for a certain period of time at a fixed filling flux and the discharge was turned off. After waiting a short period of time, recombination of the H was triggered. This period was continually doubled until a wait time of 532 sec. After each triggering the signal was unattenuated, to within experimental error. The conclusion that could be drawn was that the lifetime τ was much greater than 532 sec. Densities of order 10^{14} at/cc were stabilized, with no effort exerted at that time to increase this number. The results showed that the main stabilization was due to suppression of surface recombination /19/ as the expected volume recombination rates would not be limiting at these densities.

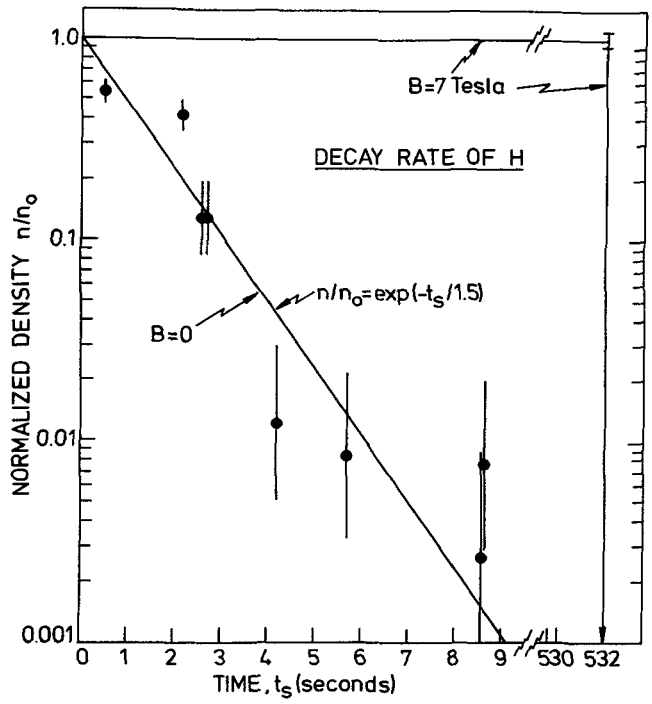

Fig. 5 The decay rate of H for T = 270 mK with magnetic field B = 7 Tesla and 0. t_s is the elapsed time after loading the cell to density n_o.

In the same figure we show the decay of the density in zero magnetic field. This decay rate could be fit to an exponential with a time constant of $\tau \approx 1.5$ sec. This decay is attributed to thermal diffusion of the gas out of the HSC. To calculate this time constant we can again appeal to simple kinetic theory,

$$- dN/dt = \frac{K}{c_H} \frac{1}{4} n\bar{v}A \qquad (2)$$

Here K is the Clausing factor of the tube of area A leading from the HSC to the HEVAC, n the density and c_H the HEVAC compression. This leads to a time constant

$$\tau = 4\, c_H V/K\bar{v}A \qquad (3)$$

By measuring τ one can determine $c_H \approx 50$ since all other factors are reasonably well known. When the magnetic field is turned on we can replace $n(B_H) = n(B_o)/c_m$ which defines the magnetic compression $c_m = \exp\{\mu(B_o - B_H)/kT\}$ where B_o is the field in the HSC and B_H that in the HEVAC /10/. With density n_o at B_o we define the effective volume as $V_{eff} = N/n_o$. This then leads to an expression for the time constant in magnetic field

$$\tau_m = 4\, c_H\, c_m\, V_{eff}/K\bar{v}A \qquad (4)$$

More recently we have carried out more extensive measurements on the time dependence and the magnetic equation of state of H↓. Samples of H↓ have been maintained for as long as 47 minutes before triggering. The decay in a magnetic field was studied at densities $\sim 3 \times 10^{14}$ at/cc and found to be exponential in accordance with the thermal diffusion mechanism. The dependence of eq. (4) is shown in fig. (6B) for two different magnetic fields.

In these measurements it was shown that at low densities the cell loads up to a saturation

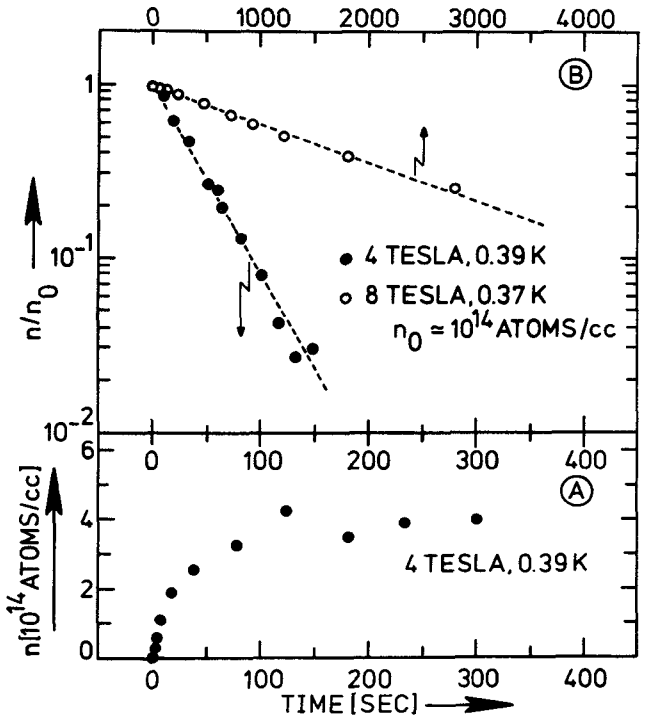

Fig. 6 A. The density of H as a function of time during filling of the cell at constant flux, field and temperature.
B. The decay of initial density n_0 at constant field and temperature.

value that can be roughly described by

$$n_{sat}(B_0) = n_{eff}(B') \exp\{ \mu(B_0 - B')/kT\} \qquad (5)$$

where $n_{eff}(B')$ is some effective density during loading, probably in the throat of the HEVAC. This is shown in fig. (6A). At higher densities there are deviations from this such that densities lower than those predicted by eq. (5) are found. It is not yet clear if this is a fundamental limitation or a problem arising from the cell design.

The highest densities we have achieved are $n > 10^{16}$ at/cc at 10.5 Tesla and $T \approx 500$ mK. The main limitation in achieving higher densities was the refrigeration. As the filling flux was increased the cell would warm up which reduces the compression (see eq. 5). We were forced to work at filling fluxes of order 5 - 10 % of that available. At these high densities deviations from exponential decay are observed, with the time for the density to halve being of order 20 minutes. At these densities the magnetic field is certainly playing an important role in reducing volume recombination (in zero field the time constant would be of order 1 sec). However the reduced lifetimes may be an indication of the increasing importance of three-body recombination processes.

As a final point we discuss some interesting observations on the trigger signals for higher densities. A trace is shown in fig. 7 of the resistance of the cell thermometer and bolometer for a high density trigger. The bolometer is triggered by exciting it with a square wave current source; the thermometer is powered with a low level ac current source, the voltage drops, proportional to the resistance are monitored. The simple theory presented earlier predicts a bolometer time constant independent of density and the cell to heat to higher temperatures for higher densities.

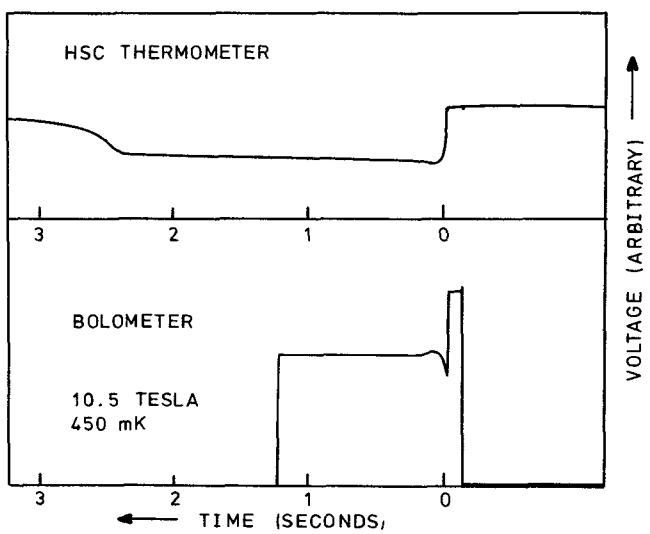

Fig. 7 The response of the HSC thermometer and bolometer for a high density, $n \approx 10^{16}$ atoms/cc. The bolometer is heated with a current square wave which returns to zero before the burning is completed. Note that the temperature of the HSC remains almost constant during the burning for approximately 2.4 sec.

Instead the cell warms to a certain temperature and remains constant until the H↓ is completely burned. The time constant for burning has been observed to be as long as 2 - 3 sec! This can be explained as follows. If the density is above a certain critical value, then after triggering the cell heats up to a temperature above that of the HEVAC. The helium film which flows towards warmer regions now starts flowing up into the HSC and the vapors flux back to condense on the HEVAC. The H↓ in the cell is pumped into the region between the HEVAC condenser and the HSC. This is a feedback controlled process: the H↓ leaks back into the HSC at a constant rate ρ to maintain the temperature of the HSC constant until it is depleted. During this time the H↓ can also leak out of the HEVAC and recombine in warmer regions, thus not contributing to the warming of the cell. The time required to establish this process can be estimated from the bolometer temperature - time trace which is seen to first overshoot the temperature of the cell and then return. The main point here is that we measure the density by the warming of the cell but this is only the lower limit. We can get an idea for the loss out of the HEVAC from the following simple model. After the beginning transient, the decay of the N H↓ atoms in the trapped volume can be calculated from

$$- dN/dt = \rho + KN\bar{v}A / 4 c_H V \qquad (6)$$

Here $N = N_H + N_c$ where N_c is the number of atoms flowing to the cell and corresponds to our measured number of atoms. N_H is the number lost out the HEVAC, K is an effective Clausing factor and V is the volume of the trap. The equation is easily solved to find $N = (N_o + \tau\rho) \exp(-t/\tau) - \tau\rho$ where N_o is the initial number of atoms and $\tau = 4 c_H V / K\bar{v}A$. For an experimental burn time of $t = t_b \approx 2.5$ sec we found $N_c = \rho t_b = 10^{16}$ atoms. We find N_o by setting $N = 0$ or $N_o = \tau\rho\{\exp(t_b/\tau)-1\}$. The results are very sensitive to τ. Using the most conservative value, $\tau = 1.5$ sec, corresponding to the measured value for atoms to empty out of the cell (not the trap) in zero field, we find $N_o \approx 3 \times 10^{16}$ at/cc. For $t_b/\tau << 1$ then $N_o \approx \tau\rho = N_c$. Thus for short burn times the measured value N_c is linearly dependent on the length of the HSC response pulse; however for high densities, in the current geometry, this measurement technique becomes insensitive and can grossly underestimate the initial density if linearity is assumed.

In summary, we have discussed here the difficulties and the techniques used to stabilize H↓. Experiments have also been performed on D↓. For undetermined reasons it has not yet been possible to achieve densities of D↓ above $\sim 10^{14}$ at/cc. H↓ has been stabilized to densities of at least 10^{16} at/cc and perhaps as much as an order of magnitude higher At the higher densities the lifetimes appear to decrease, but this has not yet been studied systematically. An increase of density of one to two orders of magnitude does not seem to be out of reach. With such densities and T < 100 mK interesting phenomena due to quantum degeneracy should be observable.

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/5/ H↓ is produced in a magnetic field. The electron spins are polarized antiparallel (down) with respect to the field direction, so it seems more appropriate to use this symbol rather than H↑ which would correspond to the higher energy spin levels.

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/19/ We use the term stabilization to mean a very large increase of the lifetime of the gas as compared to the lifetime it would have if no helium film, magnetic field or low temperature environment was used.