UNIVERSITY OF AMSTERDAM

UvA-DARE (Digital Academic Repository)

# Stabilization of Atomic Hydrogen at Low Temperature

Silvera, I.F.; Walraven, J.T.M.

DOI
10.1103/PhysRevLett.44.164
Publication date
1980

Published in
Physical Review Letters

Link to publication

Citation for published version (APA):
Silvera, I. F., & Walraven, J. T. M. (1980). Stabilization of Atomic Hydrogen at Low Temperature. *Physical Review Letters*, *44*(3), 164-168. https://doi.org/10.1103/PhysRevLett.44.164

General rights
It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations
If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: https://uba.uva.nl/en/contact, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

*UvA-DARE is a service provided by the library of the University of Amsterdam (https://dare.uva.nl)*

Download date:25 Aug 2022

critical velocity[9] for the process. Above a characteristic value of $\bar{v}$ (or, equivalently, of $E$), $n_3$ may therefore be expected to decrease rapidly, leading to a correspondingly rapid decrease in $\nu$ towards the value found in pure $^4$He, as observed (Fig. 2).

We conclude that vortex nucleation in pure $^4$He below 0.6 K can be well described in terms of a generalized Landau argument. Further work will be needed to establish whether or not the phenomenon can in fact be treated on the same basis in the cases of higher temperatures and of $^3$He-$^4$He solutions. We hope that our present results will provide an impetus towards the development of a microscopic theory of the breakdown of superfluidity through vortex nucleation in liquid $^4$He.

We acknowledge gratefully the assistance of Professor W. M. Fairbairn in developing the technique used in these experiments. This work was supported by the Science Research Council.

[a]Permanent address: Department of Physics, University of Missouri, St. Louis, Missouri 63121.
[b]Present address: School of Mathematical and Physical Sciences, University of Sussex, Falmer, Brighton BN1 9QH, Sussex, United Kingdom.

[1]G. W. Rayfield and F. Reif, Phys. Rev. 136, A1194 (1964).
[2]T. C. Padmore, Phys. Rev. A 5, 356 (1972).
[3]A. L. Fetter, in *The Physics of Liquid and Solid Helium*, edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1976), Pt. I, Chap. 3.
[4]R. J. Donnelly and P. H. Roberts, Philos. Trans. Roy. Soc. London, Ser. A 271, 41 (1971).
[5]K. W. Schwarz and P. S. Jang, Phys. Rev. A 8, 3199 (1973). See also E. I. Blount and C. M. Varma, Phys. Rev. B 14, 2888 (1976).
[6]R. M. Bowley, J. Phys. C 9, L367 (1976).
[7]J. A. Titus and J. S. Rosenshein, Phys. Rev. Lett. 31, 146 (1973).
[8]R. Zoll and K. W. Schwarz, Phys. Rev. Lett. 31, 1440 (1973).
[9]D. R. Allum and P. V. E. McClintock, J. Low Temp. Phys. 31, 321 (1978).
[10]P. C. E. Stamp, P. V. E. McClintock, and W. M. Fairbairn, J. Phys. C 12, L589 (1979).
[11]P. V. E. McClintock, Cryogenics 18, 201 (1978).
[12]D. R. Allum, P. V. E. McClintock, A. Phillips, and R. M. Bowley, Philos. Trans. Roy. Soc. London, Ser. A 284, 179 (1977).
[13]R. M. Bowley and F. W. Sheard, Phys. Rev. B 16, 244 (1977).
[14]R. J. Donnelly, Phys. Lett. 39A, 221 (1972).
[15]R. M. Ostermeier, Phys. Rev. A 8, 514 (1973).
[16]D. R. Allum and P. V. E. McClintock, J. Phys. C 9, L273 (1976).

## Stabilization of Atomic Hydrogen at Low Temperature

Isaac F. Silvera and J. T. M. Walraven
*Natuurkundig Laboratorium der Universiteit van Amsterdam, 1018 XE Amsterdam, The Netherlands*
(Received 19 November 1979)

Atomic hydrogen has been stabilized at temperatures of 270 ± 20 mK and in magnetic fields up to 7 T. It is believed to be gaseous. A sample displayed no measurable decrease in density after 532 sec. Covering of exposed surfaces with a film of $^4$He is essential.

Atomic hydrogen (H) with one proton and one electron is the simplest atomic system provided by nature. Just as helium and its isotopes display fascinating properties at low temperature and high density, atomic hydrogen and its isotopes are expected to exhibit spectacular phenomena. Until now it has not been possible to study condensed atomic hydrogen experimentally, as under normal circumstances it is highly unstable with respect to recombination to $H_2$ which has a binding energy $\epsilon_b/k_B = 52\,000$ K in the singlet spin state, $^1\Sigma_g^+$. However, in the spin-polarized state, $^3\Sigma_g^+$, a pair of atoms have no bound state. We refer to a dense gas of atoms with electron spins polarized, so that all pairs interact within the $^3\Sigma_u^+$ potential, as polarized hydrogen ($H\uparrow$). We have created a gas of H in a cell at $T = 270 \pm 20$ mK and in magnetic fields up to 7 T. Using specially developed detectors we have established that after loading the cell, the density did not change to within experimental error for periods up to 532 sec, the longest period investigated. Without our special conditions, we calculate that the sample would have a decay-time constant of ~80 $\mu$sec, due to surface recombination if kinetic effusion of the gas to the surface is rate limiting. The lifetime of this sample was increased by at least a factor $7 \times 10^7$ in this sense. We shall refer to samples with increased lifetimes of $> 10^6$ as stabilized. In most cases we observed stabilized

 © 1980 The American Physical Society

samples for times of order 10–60 sec before destroying them. The highest density of a stabilized sample was $n > 1.8 \times 10^{14}$ atoms/cm$^3$.

The first published discussion of some of the exciting phenomena expected for stable H↑ is due to Hecht,[1] who, in 1959, suggested that superfluidity should exist in this boson gas. His article was evidently overlooked by researchers who became actively interested in H↑ in the 1970's, including ourselves. Etters and colleagues[2] and Stwalley and Nosanow[3] reconsidered the properties of the hydrogen isotopes. The stability of H↑ was first considered by Jones *et al.*,[4] who found that it would be unstable at very high densities. Stwalley[5] suggested that H↑ would be stable in a magnetic field $B$ for ratios $B/T > 10^2$ T/K. However, Berlinsky *et al.*[6] showed that his reasoning was incomplete and that even at $T = 0$, for a given magnetic field there is a critical density above which the H↑ gas will become spontaneously unstable.

Our research program to create and stabilize H began in 1972 and was directed at stabilizing H by polarizing an atomic beam and depositing it on a cold inert surface in a weak magnetic field, with the eventual objective of studying Bose-Einstein condensation in a quasi-two-dimensional system. The results were discouraging as the atoms rapidly depolarized on the surface and recombined.[7] On the basis of the stability conditions found in Ref. 6 we decided to redirect our efforts to stabilization of H↑ as a three-dimensional gas in high magnetic fields at low temperatures.

Our design aims were based on an applied magnetic field $B = 10$ T and a temperature $T \simeq 300$ mK. We recently developed a high-flux source of He-temperature atomic hydrogen[8] required for the ultimate goal of loading a cold cell to densities of $10^{18}$–$10^{19}$ atoms/cm$^3$. Our earlier experience with rapid recombination of H on surfaces suggested that any stabilization attempt using currently available fields would be doomed because of condensation of the H on the surface followed by recombination. We conceived of the idea of coating all surfaces with a (superfluid) helium film,[9] believing that of all known materials this would have the weakest adsorption potential for H. This turns out to be vital for stabilization.

There are three important considerations for the selection of the operating temperature. First of all, as discussed in the accompanying Letter,[10] compression of a gas of H from a field-free region to a region of magnetic field $B$ is proportional to $\exp(\mu B/kT)$ ($\mu$ is the magnetic moment of the H atom), which favors lowest possible temperatures. The region $T \to 0$ is also favored to minimize recombination due to spin-flip mechanisms[11,4] and to minimize the density of thermally populated spin-flipped states, which depends on $\exp(-2\mu B/kT)$. However, as a third point, the low operating temperatures may be limited by condensation of H↑ on the liquid-helium–covered surfaces, where critical densities may be exceeded. Recent calculations[12,13] suggest that a surface state exists with a binding energy possibly greater than 0.6 K.

The heart of our low-temperature apparatus is shown in Fig. 1. The copper hydrogen stabilization cell (HSC) is in the center of a superconducting solenoid (maximum field, $B = 11$ T). This is connected by thin-walled German silver (GS) tubing to a helium vapor compressor (HEVAC). This is actually a miniature vapor diffusion pump which uses a reservoir of superfluid helium as its pumping fluid. Both the HSC and the HEVAC are cooled by sorption-pumped $^3$He refrigerators. During operation the HSC runs at $T = 270$ mK and has an estimated residual background helium vapor density of $\sim 5 \times 10^8$ atom/cm$^3$. The HEVAC which operates at $T = 480$ mK is connected by GS tubing to a copper H accomodator pinned to $^4$He temperatures. A Teflon-lined GS tubing continues out of the vacuum to a room-temperature hydrogen discharge. The hydrogen atoms are fed in

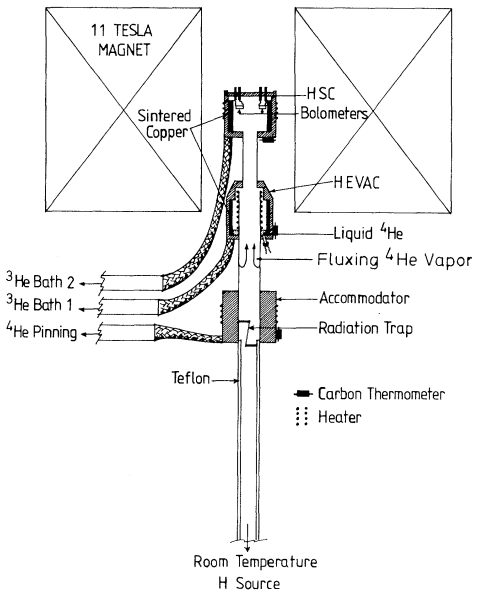


FIG. 1. Low-temperature part of stabilization apparatus. See text for an explanation.

from the bottom of the cryostat to minimize the distance and thus the recombination of H during transportation to the HSC; the atoms are cooled to liquid-helium temperature by the accomodator as described elsewhere[8] and are still unpolarized at this point. The electron spin-down atoms are drawn into the HSC by the field gradient, whereas the spin-up states are retarded and either relax to spin down or recombine. The system is prepared as follows. All surfaces above the Teflon lining are coated with molecular $H_2$. The $^3$He refrigerators are activated and $^4$He gas is condensed in the HEVAC and HSC.

We shall briefly describe the operation of the HEVAC. A superfluid helium film is driven from the reservoir of the HEVAC, up, then down the GS tube towards the accommodator. As the film flows down it comes into increasingly warmer regions, vaporizes and fluxes back to recondense at the HEVAC. Without this fluxing gas the density of $^4$He gas in the region of the HEVAC would be $\sim 10^{14}/\mathrm{cm}^3$ and the collisional mean free path ($\lambda$) for H-He would be about 2 cm.[14] The back-fluxing gas is much denser (we estimate $\lambda \sim 0.36$ mm for H-He collisions) so that hydrogen atoms will be directed into the HSC by momentum transfer from the four times more massive helium atoms. The HEVAC not only guides the atoms into the HSC, but also retards their leakage out when the discharge is turned off. As discussed in the accompanying Letter, the cell should empty when the density of H in zero field is zero. We estimate that the emptying time constant is increased by a factor 79 by the HEVAC.

The presence of H in the HSC is detected with special bolometers. The principle is to build up a density of H in the cell and then trigger its recombination by selectively making the bolometers active recombination surfaces. The concomitant heating due to the recombination energy is easily

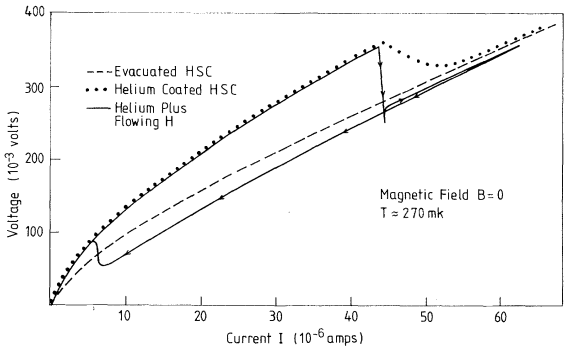


FIG. 2. Current-voltage characteristics of a hydrogen bolometer detector.

detected and provides a measurement of the H density. The bolometers were made by cutting $2\times1\times0.2$-mm$^3$ chips from Speer resistors. They are connected to the top plate of the HSC by fine copper wires which serve not only as current-voltage ($I$-$V$) leads but also as the path which allows the $^4$He film to cover their surfaces. In Fig. 2 we show the $I$-$V$ curves. When $^4$He is introduced into the HSC the $I$-$V$ curves develop a peak. The $^4$He on the bolometer surface is evaporated as a result of Ohmic heating. Fresh $^4$He will flow along the wire leads to replenish the film; however, this flow is limited by the wire circumference.[15] The peak in the $I$-$V$ curve represents the point where the $H_2$-covered surface is first exposed. This surface is an active source for recombining H.[16] When H is present while tracing an $I$-$V$ curve, arrival at the desorption peak will trigger recombination. This destroys the H gas with a time constant $\tau \simeq 4V/\bar{v}A$, where $V$ is the volume of the HSC, $\bar{v}$ is the average velocity, and $A$ is the total area of the bolometer; we assume a probability of 1 for recombination when a particle strikes the surface. For our system at $T = 270$ mK, $\tau = 8$ msec. Thus we see a sharp downward-going spike on the $I$-$V$ curve and a return to a level determined by the steady-state recombination on the bolometer of H flowing into the HSC. The $I$-$V$ curve has hysteresis because of the extra heating after desorption. From earlier work[16] it is known that the recombining atoms deposit only a small portion of the recombination energy on the bolometer surface, since the $H_2$ molecules are desorbed upon recombination. The second bolometer, operated below its desorption peak, will absorb part of this energy from the "hot" molecules. The rest will be absorbed in the copper walls of the HSC (or blown out of the filling tube) and is detected with a thermometer. By calibrating the heating of the HSC with an electrical heater, a lower bound for the number of recombinations, or the H density, can be determined.

Proof of stabilization was obtained as follows. With the magnetic field and discharge on, we measured the recombination spike height as a function of cell loading time. As the loading period was increased the size of the spike increased, indicating that larger densities of H were being built up. We then loaded the cell for a period of time $t_L$($\sim$30 sec), turned off the discharge, and waited a time, $t_s$, before triggering. Starting with $t_s \simeq 1$ sec we continually doubled the wait time, finding no measurable attenuation in

the size of the spikes. Periods longer than 532 sec were not measured as we chose to use our limited experimental measuring time to investigate other aspects of the system. This provides proof of long-term stability of H. This was performed with a field of 7 T at $T = 270 \pm 20$ mK. Although no detailed systematic studies have been performed we found that the recombination spike height decreased with both decreasing fields and increasing temperature for a constant filling rate, as is to be expected. The largest density we observed was at 6 T (270 mK) with a *very conservative* lower bound of $1.8 \times 10^{14}$ atom/cm$^3$. No effort has been made as yet to determine a maximum achievable density.

We have also measured the time dependence of the density in zero field, where the gas is certainly H (not H$^\uparrow$) and should not be stable. The results are shown in Fig. 3. The decay does not appear to be controlled by volume recombination, probably because the density is low. If we roughly fit by an exponential decay, as would be expected for thermal effusion, we find a time constant of 1.5 sec, as compared to a calculated 19 ms without the HEVAC. This implies a compression factor of 79 for the HEVAC.

Indications that H exists in the gas phase are the following. On the basis of the theoretical estimate for the binding energy of H on a He surface, at the employed temperatures we expect that in thermodynamic equilibrium a considerable fraction will be gaseous. Second, the H totally recombines in about 80 msec as measured by the full width at the base of the bolometer spike. Since this can be several time constants long, the recombination rate is of the expected order as determined by gaseous effusion to the bolometer surface. Third, if the bolometer signals were due only to H on its surface, then, assuming a coverage of maximal $10^{14}$/cm$^2$, the signal would correspond to $2 \times 10^{12}$ recombinations, inconsistent with observations; a second triggering without reloading the cell did not result in a recombination spike as would be expected as a result of surface diffusion which would recover the bolometers. Finally, we performed experiments in which we sequentially triggered the two bolometers within 100 msec. Only the first triggered bolometer displayed a recombination spike, which would not be the case if the H resided mainly on the surface. Although no one argument here provides a hard proof of the state of H, altogether the evidence points to gaseous H. We hope to be able to increase the density by some orders of magnitude to study the interesting properties of this almost ideal Bose gas. Already at present densities there are important implications for increased stability of the hydrogen maser.[17]

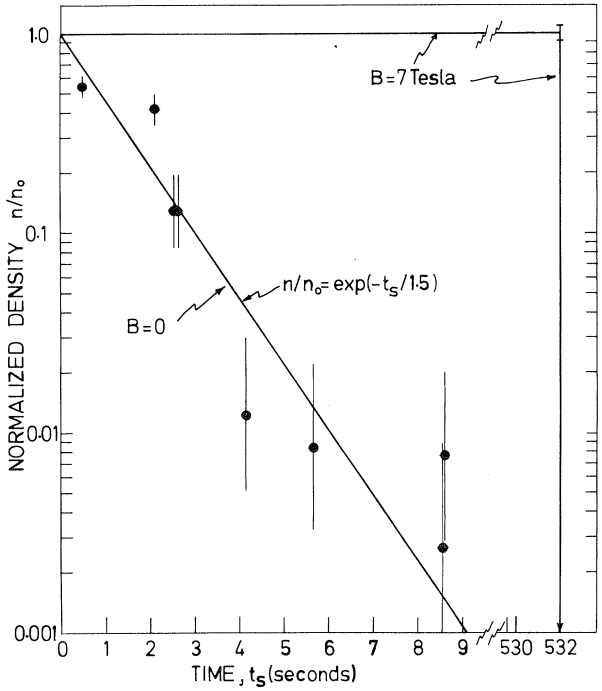


FIG. 3. Decay rate of H for $T = 270$ mK at $B = 0$ and 7 T. $t_s$ is the elapsed time after loading the cell and $n_0$ is the density for $t_s = 0$. The error bars are due to calibration uncertainties only.

We thank A. P. M. Mattheij, R. Sprik, and M. B. J. Diemeer for their help with the apparatus and measurements. We warmly acknowledge discussions with A. J. Berlinsky and E. R. Eliel at the beginning states of these experiments. The rapid and expert constructional work by O. H. Höpfner has been of inestimable value. We thank the Stichting voor Fundamental Onderzoek der Materie for their great flexibility and understanding in supporting this research of a more speculative nature.

[1]C. E. Hecht, Physica 25, 1159 (1959).

[2]J. V. Dugan and R. D. Etters, J. Chem. Phys. 59, 6171 (1973); R. D. Etters, J. V. Dugan, and R. W. Palmer, J. Chem. Phys. 62, 313 (1975); R. D. Etters, R. L. Danilowitz, and R. W. Palmer, J. Low Temp. Phys. 33, 305 (1978).

[3]W. C. Stwalley and L. H. Nosanow, Phys. Rev. Lett. 36, 910 (1976).

[4]J. T. Jones Jr., M. H. Johnson, H. L. Mayer, S. Katz,

and R. S. Wright, Aeronutronic Systems Inc. Publication No. 6-216, 1958 (unpublished).

[5]W. C. Stwalley, Phys. Rev. Lett. 37, 1628 (1976).

[6]A. J. Berlinsky, R. D. Etters, V. V. Goldman, and I. F. Silvera, Phys. Rev. Lett. 38, 356 (1977).

[7]J. T. M. Walraven, E. R. Eliel, and I. F. Silvera, Phys. Lett. 66A, 247 (1978).

[8]I. F. Silvera and J. T. M. Walraven, Phys. Lett. 74A, 193 (1979).

[9]Others have evidently also had this idea. W. C. Stwalley, in *Quantum Fluids and Solids*, edited by S. B. Trickey, E. D. Adams, and J. W. Dufty (Plenum, New York, 1977, p. 293.

[10]J. T. M. Walraven and I. F. Silvera, following Letter [Phys. Rev. Lett. 44, 168 (1980)].

[11]The necessary field for stability considered in Ref. 6 will increase for $T \neq 0$ as the pair distribution function will allow for more penetrating pair collisions as $T$ increases.

[12]R. A. Guyer and M. D. Miller, Phys. Rev. Lett. 42, 1754 (1979).

[13]I. B. Mantz and D. O. Edwards, to be published.

[14]J. P. Toennies, W. Welz, and G. Wolf, Chem. Phys. Lett. 44, 5 (1976).

[15]For properties of these films, see, for example, J. Wilks, *The Properties of Liquid and Solid Helium* (Clarendon, Oxford 1967), Chap. 14.

[16]See Ref. 7 and references therein.

[17]S. B. Crampton, W. D. Phillips, and D. Kleppner, Bull. Am. Phys. Soc. 23, 86 (1978).

# Density, Magnetization, Compression, and Thermal Leakage of Low-Temperature Atomic Hydrogen

J. T. M. Walraven and Isaac F. Silvera

*Natuurkundig Laboratorium der Universiteit van Amsterdam, 1018 XE Amsterdam, The Netherlands*

(Received 19 November 1979)

The density distribution of low-temperature spin-polarized atomic hydrogen in a realistic magnetic field profile is calculated for densities below and above the critical value for Bose-Einstein condensation. The distribution is an identifying characteristic. Magnetic compression and instability due to thermal leakage of the atoms is treated.

Atomic hydrogen (H) which has been shown in the accompanying Letter[1] to exist in a long-time stable state provides us with a new Bose fluid. Since in general it is necessary to prepare this gas by injecting atoms into a magnetic field, a treatment of the effect of magnetic field gradients is required. In this Letter we calculate the density distribution for spin-polarized H (H↑) in a realistic magnetic field profile. Results are obtained below and above $n_c$, the critical density for Bose-Einstein (BE) condensation; the effects of interactions in the high-density BE condensed state are shown to be non-negligible. The density profile is a characteristic identifying feature of the gas. This calculation immediately provides the local static magnetization, which is related to the density by a proportionality constant: the Bohr magneton. The important concept of a magnetic compression is introduced. Finally, we consider the instability of a magnetically confined gas in an open-ended container, due to thermal leakage. Comparisons are made with experiment where possible.

At low temperatures H↑ is considered to be an extremely weakly interacting Bose gas.[2] To illustrate this point we have plotted in Fig. 1 the $p$-$V$ curves for H↑ at $T = 0$ K as calculated by Etters, Danilowicz, and Palmer[3] and for an ideal Bose gas at $T = 0.1$ and 0.3 K. As the 0-K curve can be looked upon as a measure of the interactions

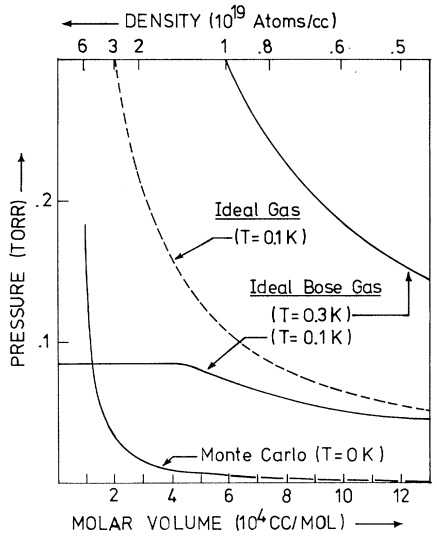


FIG. 1. $p$-$V$ relation for H↑ in various approximations and at various temperatures.



© 1980 The American Physical Society