

**DIRECT DETERMINATION OF THE TEMPERATURE AND DENSITY
OF GASEOUS ATOMIC HYDROGEN AT LOW TEMPERATURE
BY ATOMIC BEAM TECHNIQUES**

Isaac F. SILVERA and J.T.M. WALRAVEN

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

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We have measured the translational temperature of atomic hydrogen gas (H) in a cryogenic environment at $T \approx 8$ K. An atomic beam is formed; time of flight measurements are consistent with a maxwellian velocity distribution of 8.5 K. The flux of low velocity atoms is increased by ≈ 20 over that of the same source operated at room temperature. The low temperature density is estimated to be 3.4×10^{13} H/cm³. General considerations for transporting and cooling of a gas of H are given.

We have determined the translational temperature of atomic hydrogen (H) gas in a cryogenic environment by forming an atomic beam and measuring the velocity distribution. H₂ gas is dissociated in a room temperature source and interacted with cold walls ($T = 7.7$ K). Our results are the first demonstration that the atoms approach equilibrium with the walls; the velocity distribution can be fit with the maxwellian distribution with $T = 8.5$ K. In cooling from room temperature to helium temperatures, the total H flux decreased by a factor of ≈ 3 due to recombination. Absolute measurements indicate a total flux of 2.4×10^{16} H/s, to our knowledge, the most intense source of low velocity H that exists. This leads to a low temperature steady state average density of 3.4×10^{13} H/cm³. Recently, Crampton et al. [1] and Hardy et al. [2] have studied H (created in rf discharges) by flowing it into H₂ and He covered enclosures at helium temperatures. In these experiments a steady state H density of order 5×10^{13} at/cm³, as measured by resonance techniques, builds up. The density could possibly have been an order of magnitude higher, with uncertainty arising from the unknown spin temperature [1]. Although limits could be suggested for the translational temperature of H from indirect observations [1], it was not at all clear that a maxwellian temperature existed as the rapid re-

combination can supply up to $E_t/k \approx 52000$ K of kinetic energy per event.

Our motivation in developing this source is to use it as an injector in our experimental effort to stabilize atomic hydrogen and to observe its possible Bose condensation at very low temperatures in the presence of intense magnetic fields [3-7]. The field serves to place the atoms in a spin polarized state so that they interact via the triplet potential and could be stabilized at high densities [6]. In addition such a source will be useful in the study of the interaction of H with surfaces [7,8] and the study of interatomic potentials by atomic beam scattering [9].

Toennies et al. [9] have successfully produced atomic H beams at temperatures of $T \approx 90$ K (for use in beam scattering experiments) using a technique first described by Wilsch [10]. H, created in a hot rf discharge, is cooled by passing through a narrow liquid nitrogen temperature aluminium channel. In this case, evidently the H is accommodated by an aluminium oxide layer and does not recombine, as would be expected if the surface were metallic. However when they tried to further cool the beam by accommodating with a liquid H₂ (20.4 K) cooled aluminium channel, substantial recombination took place and the remaining atoms were not effectively accommodated [11].

In order to understand the basis of the cold source,

let us consider the limitations on a high flux beam. It is rather straightforward to build a microwave or rf discharge [12,7] source operating at room temperature or higher. In our design, operating pressures for which H_2 is efficiently dissociated (better than 90%) extend up to a few Torr; typically fluxes $\phi = \frac{1}{4}Kn\bar{v}A \approx 5 \times 10^{17}$ at/s (n is the density, \bar{v} the average velocity and K the Clausing factor) expand out of an $A = 0.1 \text{ mm}^2$ orifice of length 1 mm into a 5 mm id teflon tube which guides the H to a liquid helium cooled accommodator. The flux of atoms drifting down the teflon tube will be attenuated due to the finite surface (ϕ_σ) and volume (ϕ_v) recombination rates, the density being determined by the following rate equation:

$$dn/dt = (1/V)(\phi_{in} - \phi_{out} - \phi_v - \phi_\sigma). \quad (1)$$

Here ϕ_{in} and ϕ_{out} represent the atomic fluxes in and out of the tube with volume V and surface area σ ; $\phi_v = K_v V n^3$ and $\phi_\sigma = K_\sigma \sigma n^\delta$ ($\delta = 1$ or 2) [13]. Eq. (1) can be solved for the steady state condition and the ratio ϕ_{out}/ϕ_{in} can be optimized in terms of the tube dimensions. The problem of the H density in the tube has been studied under conditions in which ϕ_v is negligible and ϕ_σ is due to first order processes ($\delta = 1$) [14]. However, it is more illuminating to discuss the individual terms in eq. (1).

Since there is considerable knowledge of the volume recombination rate, conditions can be chosen so that this is not limiting ϕ_{out} . The measured rate constant is $K_v \approx 2.7 \times 10^{-32} \text{ cm}^6/\text{at s}$ [15] and appears to have only a weak temperature dependence. The Clausing formula [16], $\phi_{out} = \frac{1}{4}Kn\bar{v}A_{out}$ ($K \approx \frac{8}{3}R/L$ where R is the radius and L the tube length), may be used to estimate the density at the beginning of the teflon tube, i.e. immediately behind the source orifice. For our system this yields an inlet density $n_p = 2.4 \times 10^{15} \text{ H/cm}^3$, using $\phi_{in} = \phi_{out} = 5 \times 10^{17} \text{ H/s}$, with $L = 25 \text{ cm}$. This density leads to an initial decay rate, $(1/n_p) dn_p/dt$, of 16%/s due to volume recombination. Comparing this figure with the average time the atoms spend in the tube ($\approx 7 \text{ ms}$) shows that one can use fluxes up to $\approx 10^{19} \text{ H/s}$ in such a tube before volume recombination losses become significant.

The surface loss term is more difficult to handle as K_σ depends strongly on the surface composition, and ϕ_σ depends strongly on temperature. For $kT \gg |E_{ads}|$ (the adsorption energy of a particle bound

to the surface), a particle has a mean surface residence time (τ) of the order of one vibrational period ($\approx 10^{-13} - 10^{-15} \text{ s}$). The surface coverage is $N/\sigma \approx \frac{1}{4}n\bar{v}\tau$. The best room temperature wall material that we have tested is teflon. For example, we have passed a flux $\phi_{in} \approx 2 \times 10^{17} \text{ H/s}$ through a teflon tube with $L = 50 \text{ cm}$ and $R = 0.23 \text{ cm}$ finding a measured $\phi_{out} = 7 \times 10^{16} \text{ H/s}$. More active surfaces can also be effectively coated with teflon [17].

For $kT \ll |E_{ads}|$, $\tau \rightarrow \infty$ and all particles condense on the surface. This is one of the severe problems in cooling H by interaction with walls. The best conceivable surface to inhibit condensation is that of liquid helium for which the adsorption energy of H is calculated to be -0.1 K [18]. However, use of this surface requires very low temperatures to maintain a low density of the saturated helium vapor which can also catalyze recombination. Therefore, for $T \gtrsim 0.5 \text{ K}$ one must rely on other surfaces with a weak physisorption potential for H, such as H_2 , Ne, etc. Since all of these surfaces will adsorb H at helium temperatures, one must attempt to limit the mean number of wall collisions⁺¹, $\eta = \frac{1}{8}(L/R)^2$, to a minimum number necessary for cooling of the gas to the temperature of the accommodating wall. For our helium temperature accommodator we use a teflon coated fused quartz tube 2.0 cm long with a 0.4 cm id. The tube is glued into a copper cylinder which is in thermal contact with a cold finger from a cryostat.

⁺¹ Our situation is slightly different than that of Clausing [16] and the number of collisions can be about a factor of three larger than η .

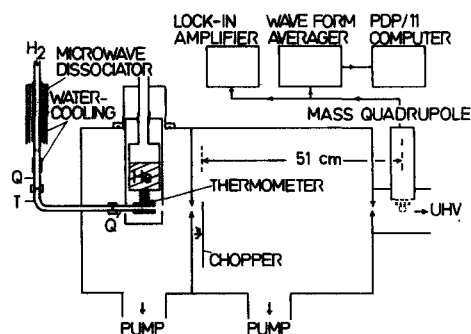


Fig. 1. Schematic diagram of the atomic H source and detection system. All tubing outside of the discharge is teflon (T) or teflon coated quartz (Q).

Our beam apparatus is shown in fig. 1. The beam is chopped with an equal segment on-off chopper and travels 51 cm to the mass quadrupole detector. The resulting signal is averaged on a waveform analyzer. This data is transmitted to a PDP/11 computer where further smoothing and processing of data is done. The derivative of the waveform gives the time of flight distribution (TOF). This technique has the advantage over the use of a narrow slit chopper in that the average beam flux can be continuously monitored on a lock-in amplifier enabling simple alignment and a rapid indication that atoms are coming out of the source. The measurements correspond to the actual TOF distribution weighted by the density $n(v)$ in the beam, since the detector sensitivity is proportional to n . Because of the finite turn-on time of the chopped beam the distribution is distorted. In fig. 2a we show maxwellian distributions and the convolutions with our turn-on function which was determined by a rectangular slit. The zero-time for the TOF's was determined by fitting of TOF distributions of a number of gases to the suitably weighted maxwellian distributions.

The TOF's were transformed to velocity distributions as shown in fig. 2b. The data are scaled so that the areas under the curves give the total measured

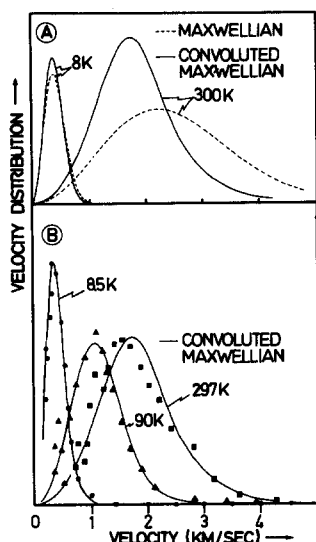


Fig. 2. Velocity distributions of H. (a) Theoretical maxwellian distribution and the convoluted maxwellian. (b) Experimental velocity distribution compared to convoluted maxwellian at various temperatures. The data has been smoothed.

atomic flux *leaving* the source. We show data for which the accommodator was at 297 ± 3 m 78 ± 2 and 7.7 ± 0.2 K. The distributions can best be fit with temperatures 297 ± 5 , 90 ± 5 , and 8.5 ± 1 K, respectively. (The same temperatures were found for H_2 with the discharge on; these data are not shown). A dramatic shift of the distribution is observed with cooling, indicating a large increase in the flux of low velocity atoms. We estimate that the average number of wall collisions is 13 and gas collisions is 4 for atoms entering the accommodator. Evidently this is insufficient to accommodate at 78 K, but about optimum at 8 K. The difference can be attributed to a different surface adsorption time at the different temperatures. At 8 K the teflon surface will be covered with a few monolayers of H_2 . Crampton et al. [1] determined the adsorption time on H_2 at 4.2 K to be 3×10^{-8} s, which is very long compared to residence times for physisorption at higher temperatures. Radiation and conduction heat leaks limited our present experiment to accommodator temperatures above 7.7 K.

The total flux was determined as follows. First the axial intensity is measured using the absolute calibration of our mass spectrometer; the total flux follows from kinetic theory taking into account the effect of beam peaking [19]. For H at room temperature we found $\phi_{297} = 7.2 \times 10^{16}$ H/s. In cooling from 297 to 90 K the H flux decreased by 0.65 ($0.65 \phi_{297}$); further cooling to 8.5 K resulted in $0.32 \phi_{297}$. Comparing this low-temperature flux of atoms (for $v < 2\alpha_b$, $= 918$ m/s; where α_b is the most probable velocity in the beam) with the room temperature flux in the same velocity interval, we find an intensity increase of approximately 20, the theoretical maximum being 71. We believe that fluxes of H a factor 5–10 larger can be achieved with minor modifications.

Assuming a linear increase in density of the stagnant H-gas, starting from zero at the end of the accommodator [19], we calculate the density gradient to be 3.4×10^{13} H/cm³ cm ($\nabla n = 3\phi_{out}/2R\bar{v}A_{out}$); this figure is also the average stagnant density in the accommodator which corresponds to the density of atoms which survive the passage through the accommodator without recombining and is thus a lower bound as we do not include the atoms which exist at the inlet of the accommodator, but recombine. We note that if the walls of the accommodator were covered with a thick film of solid H_2 , the H_2 density due to the satu-

rated vapor pressure would result in an H_2 beam three orders of magnitude more intense than was observed; from this, we conclude that an H_2 coverage of the teflon is at most a few monolayers.

In summary we have shown that by rapidly expanding atomic H to low density from a normal 300 K source, it can be transported without serious recombination. Rapid cooling to liquid helium temperatures can be achieved by interacting with cold walls made of materials which present a weak adsorption potential to the H atoms. The gas is characterized by the thermodynamic temperature of the environment. The high flux source described here should be more than adequate as an injector of H in high density stabilization cells.

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References

- [1] S.B. Crampton et al., Phys. Rev. Lett. 42 (1979) 1039.
- [2] W.N. Hardy, A.J. Berlinsky and L.A. Whitehead, Phys. Rev. Lett. 42 (1979) 1042.
- [3] C.E. Hecht, Physics 25 (1959) 1159.
- [4] R.D. Etters, J.V. Dugan and R.W. Palmer, J. Chem Phys. 62 (1975) 313.
- [5] W.C. Stwalley and L.H. Nosanow, Phys. Rev. Lett. 36 (1976) 910.
- [6] A.J. Berlinsky, R.D. Etters, V.V. Goldman and I.F. Silvera, Phys. Rev. Lett. 38 (1977) 356.
- [7] J.T.M. Walraven, E.R. Eliel and I.F. Silvera, Phys. Lett. 66A (1978) 247.
- [8] A. Schutte, F. Tommasini, A. Torelli, G. Scoles and L.F.J. Hermans, J. Chem. Phys. 64 (1976) 4135.
- [9] J.P. Toennies, W. Welz and G. Wolf, preprint, submitted to J. Chem. Phys.
- [10] H. Wilsch, J. Chem. Phys. 56 (1972) 1412.
- [11] W. Welz, Ph.D. Thesis, Max Planck Inst. für Stromungsforschung, Göttingen, Bericht 107 (1976).
- [12] G.O. Brink, R.E. Fluegge and R.J. Hull, Rev. Sci. Instrum. 39 (1968) 1171.
- [13] H. Wise and B.J. Wood, Adv. At. Mol. Phys. 3 (1967) 291.
- [14] A.I. Livshits, I.M. Metter and L.É. Rikenglaz, Sov. Phys. Tech. Phys. 16 (1971) 276.
- [15] D.N. Mitchell and R.J. Le Roy, J. Chem. Phys. 67 (1977) 1042.
- [16] P. Clausing, Ann. d. Phys. 7 (1930) 489.
- [17] P.W. Zitzewitz, Ph.D. Thesis, Harvard 1970, unpublished.
- [18] R.A. Guyer and M.D. Miller, to be published.
- [19] J.A. Giordamine and T.C. Wang, J. Appl. Phys. 31 (1960) 463.