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# RECOMBINATION PROBLEMS BETWEEN 4 AND 100 K

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## ABSTRACT

Recombination of hydrogen atoms on surfaces of interest in low temperature atomic beam production is discussed. Solutions found at 6 K and 35 K are presented. Binding energies of $H^o$ on $H_2O$ and $N_2$ are given.

## 1. Introduction

Large improvements in the output of polarized hydrogen ion sources are expected if the atomic beam stage is designed for operation with low velocity atoms. The efficiency of the ionizer depends on the dwell time of the atoms in the ionizing region and the solid angle accepted by a separation magnet varies like $T^{-1}$. Thus a $T^{-3/2}$ dependence of the ionic output of the source is suggested. Compared with a room temperature source, the gains would be about 7.5, 55 and 600 at liquid nitrogen, liquid hydrogen and liquid helium temperatures respectively. While these numbers are very impressive, the question is whether they are realistic for a practical device. Indeed, different kind of limitations will influence the final performances. For example, if the cold atomic beam has to match an existing long ionizer, the latter sets a limit to the phase space accepted by the whole system. This and other technical constraints on the geometry determine the optics and, thus, the range of velocity in which an optimum transportation can be achieved [1].

A reduction of the expected gain also arises from the increased loss of beam (scattering) if the vacuum conditions cannot be improved. Further, the beam formation may suffer from the changed flow conditions in and in front of the nozzle. As a result, one may be forced to operate a cold source at a reduced gas input.

As will be discussed in this paper, the most important problem is the loss of atoms by surface recombination during the cooling process. In a typical cold source, the atoms are generated at room temperature and transported to an accommodator by a tubing of suitable low recombination material. Optionally, an intermediate cooling can be used, either by keeping the tip of the dissociator at LN2 temperature [ 2 ] or with a separately cooled section [3]. The surface and the geometry of the accommodator should be chosen such that an efficient cooling without excessive recombination loss is achieved. In addition, the exit aperture, which forms the nozzle, should allow a reasonable beam formation.

The above considerations show that many different temperature dependent effects set conflicting requirements on the choice of the optimum temperature. In the following it is shown that, in the temperature range to be discussed here, the nature of deposits on the accommodator surface is of primary importance for the recombination losses and for the solution of this problem.

## 2. Experimental Investigations

### 2.1. Previous work

Several investigations using a variety of techniques have already been performed in the past. The following informations are available in the temperature range of interest. Reflection of hydrogen atoms on a cold surface has been investigated by Brackmann and Fite [ 4 ] for $2.5<T<80$ K and by Marenco et al. [5] and Schutte et al. [6] at $T \sim 4$ K. Crampton has determined binding energies for $H^o$ on $H_2$ [7] and very recently on Ne [8] by means of magnetic resonance studies. Walvaren and Silvera have generated a low intensity beam at 8 K [9]. At liquid hydrogen temperature, Toennies et al. observed a very strong recombination [10]. At liquid nitrogen temperature, investigations of nozzle material have been performed by Toennies et al [11] and Mathews et al. [12]. For the recombination coefficient of $H^o$ on glass and quarz measurements have been done above 77 K. They are discussed by Wood and Wise [13, 14] and Gelb and Kim [15]. Intense sources are successfully operated in this temperature range [16, 17].

The investigations at the lower temperature show that the presence of frozen or adsorbed deposits on the cold surface can strongly affect the recombination behaviour at least at the lowest temperatures. This will be particularly true in a practical source where the vacuum conditions in the first stage are generally poor and where the practically achievable gas purity is not sufficient to avoid a contamination of the surface. Therefore, without a strong cleaning effect due to reactions with $H^o$ or to desorption induced by the large energy deposited by the recombining atoms, the properties of the cooling system will be rather characterized by the nature of the deposits than by the material used for the accommodator. In the following, results obtained on test benches for intense polarized source are presented.

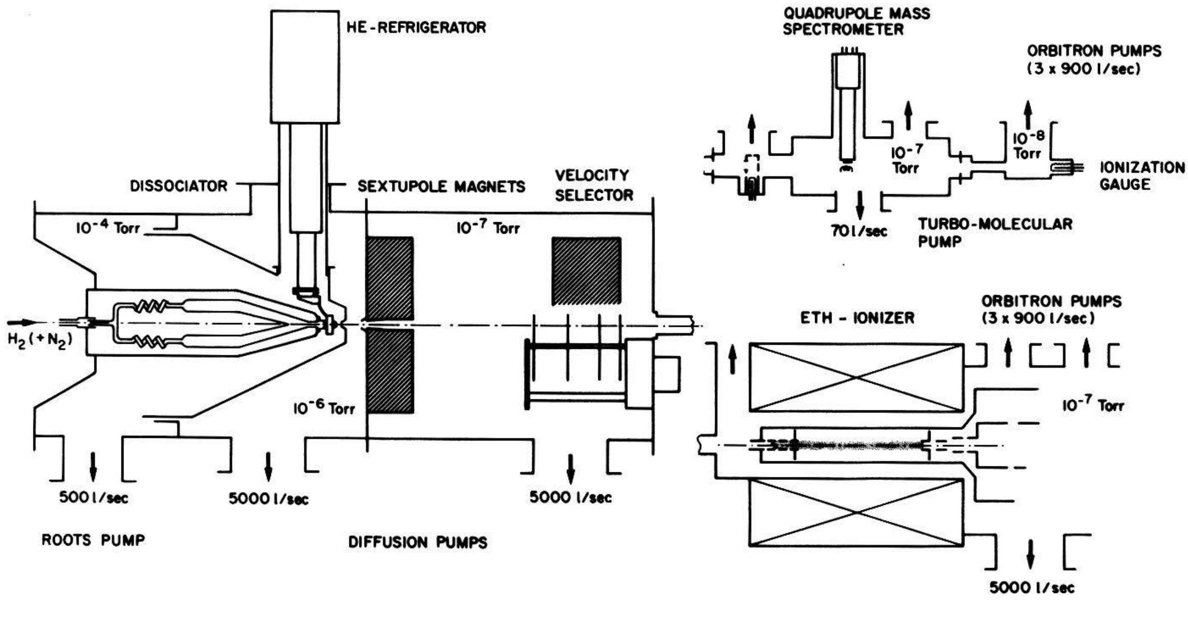


*Fig. 1* *Test bench for a cold atomic beam. The ETH ionizer is also shown.*

**2.2. Results above 15 K**

The recombination problems between room temperature and 15 K have been investigated at ETH, partly with the set-up of ref. 18, where some observations have already been presented, and partly on the improved version shown in fig. 1. Compression tubes allow beam intensity measurements and a quadrupole mass spectrometer is used for relative density determination. The details of atom generation, transport and cooling are shown in fig. 2 together with the beam forming elements. Cooling power is provided by a 10 W closed-cycle helium refrigerator.

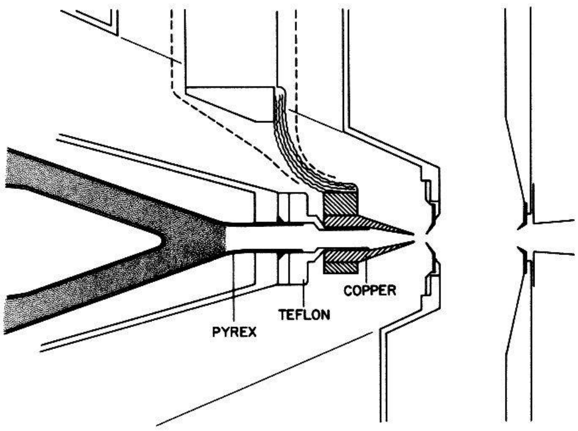


***Fig. 2*** *Details of the cold beam production.*

From a historical point of view the results on recombination are a byproduct of an intensive search for a suitable accommodator material. First Teflon coatings were used, then uncoated Al or Cu, freshly polished or oxydized, with, at first glance, a surprising result: the same behaviour is observed below about 200 K. A typical evolution of the beam density as a function of the temperature is shown by the thick curve in fig. 3. A copper accommodator is used in this experiment and the focusing magnets are switched off. The thin curve indicates the behaviour one expects if the recombination remains constant at the value at 150 K over the whole temperature range. It is obtained by correcting the $T^{-1/2}$ dependence (broken line) for the loss due to the temperature dependent beam attenuation (beam forming, scattering) measured for this experiment. The most striking feature is the rapid drop observed below 50 K, indicating a dramatic increase of the recombination below this temperature. The strong recombination at room temperature is typical for a copper accommodator and is absent if a Teflon coating is used.

At the lowest temperature several interesting observations can be made. The minimum reachable temperature depends on the thermal load from the recombination of the atoms. With very light beams, the density shows a tendency to increase again between 20 and 15 K. If the dissociator is turned off for several minutes at the lowest temperature, i.e. the system is flushed with undissociated gas, a new behaviour is seen when the atomic

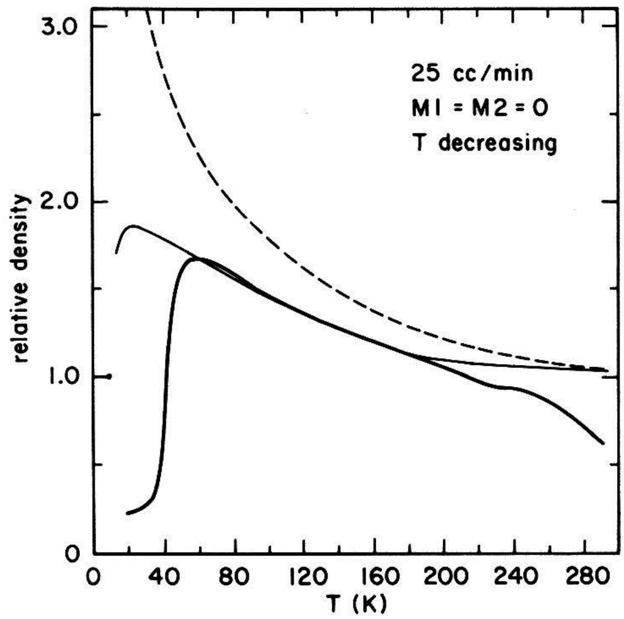


*Fig. 3* *Beam density as a function of the accommodator temperature. The curves are discussed in the text.*

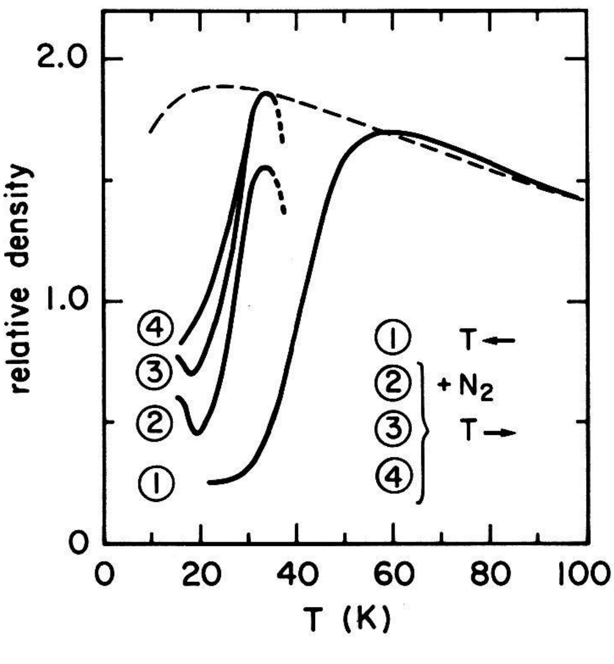


*Fig. 4* *Behaviour of the beam density for different amounts of $N_2$ added to the gas at 15 K.*

beam density is measured with increasing temperature. A maximum developes now around 35 K, the height of which depends on the flushing time. For long expositions to the undissociated gas (> 30 minutes) an increase in the density of the atomic beam is already observed at the lowest temperature. This behaviour suggests that a recombination inhibiting surface is generated during the flushing and that this surface survives a heating up to about 40 K, where it starts to evaporate. The significance of such an effect is immediately seen. In order to understand it investigations with different deposits or with contaminants added to the gas ($N_2$, $H_2O$, $O_2$, Ar, Ne, $CO_2$, ...) were performed.

The results obtained by adding different amounts of $N_2$ (estimated to be at most a few percents) to the gas and measuring the density of the atomic beam with increasing temperature is shown in fig. 4. The behaviour is the same as observed previously after long flushing times with undissociated $H_2$. Fig. 5a shows a measurement performed with the focusing magnets turned on, illustrating the better matching of the 35 K beam with the optics of a system designed for $v \lesssim 1000$ m/s. In fig. 5b the effect of the addition of $N_2$ at ~60 K by decreasing temperature is shown.

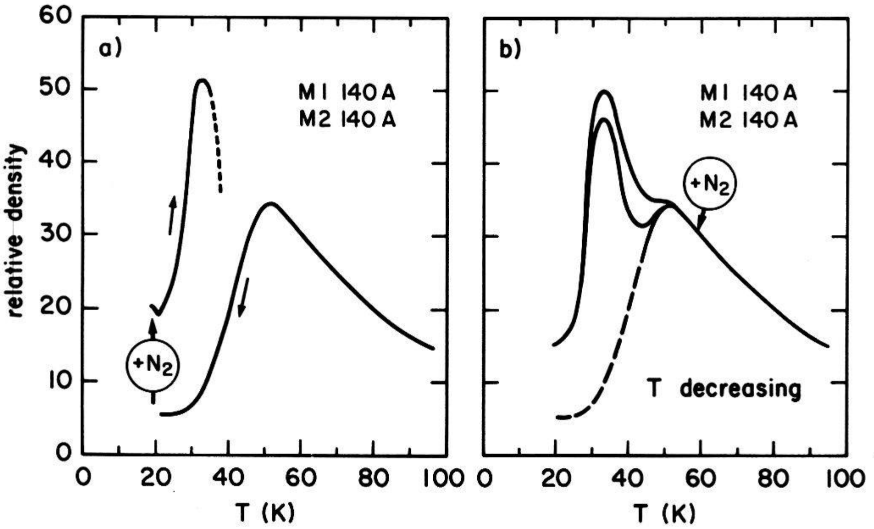


*Fig. 5 Focussed beam density as a function of the temperature*
*a) $N_2$ added at ~ 15 K*
*b) $N_2$ added at ~ 60 K*

**2.3. Investigations at liquid helium temperature**

At BNL Hershcovitch et al. are currently developping an intense pulsed atomic beam to be operated at liquid helium temperature [ 2]. Here, the $H_2$ layer covering the accommodator is expected to be a good recombination inhibiting surface, because of the low (~36 K) binding energy of $H^o$ on $H_2$ [7,8].

The results of beam density measurements at BNL are shown in fig. 6. Clearly, the observed maximum at 6 K demonstrates the good properties of the $H_2$ surface. It is interesting to notice the difference between measurements taken with decreasing and with increasing temperatures above 15 K, and the survival of a recombination inhibiting effect up to 27 K. In this

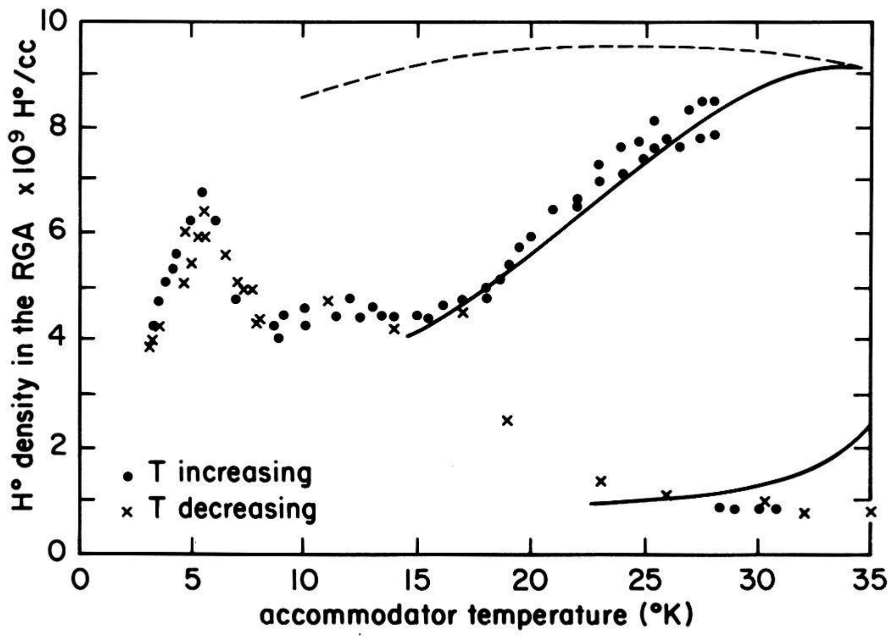


***Fig. 6*** *BNL measurements of the beam density as a function of the temperature. The curves (ETH results) are discussed in the text.*

figure are also shown ETH results taken from fig. 4. The curves are normalized to the BNL value for increasing temperature at 27 K. The only aim of this comparison is to show that despite the very different experimental arrangements, an astonishing qualitative consistency exists between the observations made by the two groups.

**3. Interpretation of the measurements**

The observed behaviour of the recombination as a function of the temperature supports the conjecture that the properties of the accommodator surface are, under practical conditions of vacuum and gas purity, dominated by the presence of frozen or adsorbed species.

The measurement of the beam loss due to recombination allows an approximate determination of the recombination coefficient and the extraction of the corresponding values for the binding energy of $H^o$ on the involved surfaces.

The rapid decrease of the recombination observed on a copper accommodator between 300 and 200 K is not compatible with the extrapolation of the values observed at higher temperature [14] and suggests a change in the nature of the surface as the temperature is decreased. Between 200 and 60 K the recombination is negligible. Below 50 K the temperature dependence of the recombination coefficient extracted from curve 1 in fig. 4 can be fitted by the Gelb and Kim relation for second order recombination [15] and yields ~500 K for the binding energy of $H^o$ on the surface. This value is in the range one might expect if a layer of frozen $H_2O$ is present. This result is not completely unexpected and explains the identical low temperature behaviour for different accommodator material.

The observations made with increasing temperature between 15 K and 40 K after cooling to 4 K or flushing with molecular $H_2$ at 15 K may suggest the survival of a strongly adsorbed layer of $H_2$. Govers et al. [19] have shown that, up to a coverage of .25 monolayer, $H_2$ is bound to $H_2O$ with an energy of about 500 K. This process is however unlikely to play a role above 20 K and could not effectively protect the atoms against the $H_2O$ underlying surface. The behaviour is so similar to the one observed after addition of $N_2$ that one can attribute it to the presence of this gas as an impurity.

From the experiments performed with an $N_2$ layer one can again extract the temperature dependence of the recombination coefficient and, using the Gelb and Kim relation [15] calculate the binding energy of $H^o$ on a $N_2$ layer, which is found to be approximately 150 K.

**4. Practical consequences**

The use of $H_2$ or $N_2$ surfaces allows an efficient inhibition of surface recombination at low temperature and makes it possible to generate atomic beams of high intensity and density around 6 and 35 K. The lower temperature is probably convenient for pulsed operation only and requires a redesign of the complete source. The higher temperature, although practicable, corresponds to the upper limit of the velocity range suitable for an optic designed for optimum improvement of an existing source with a long ionizer and several rf-transitions. Therefore it will certainly be useful to pay attention to the solution of the recombination problem between these two temperatures, i.e. around 15 K. A neon layer adsorbed on a suitable substrate may be a possible solution. For the understanding of the properties of such recombination inhibiting layers it may be interesting to look at some other characteristics of the deposits. Is it just an accident that the lowest recombination on $N_2$ is observed in the vicinity of a change in its cristalline structure?

The observations discussed in this paper have already contributed to the improvement of operating polarized hydrogen ion sources. With the ETH source operated at 35 K a significant increase in ionic output could be demonstrated [20]. Routine production of polarized beams at SIN with a source based on the same techniques has prooved the reliability and the long term stability of the method [21]. At Saturne, investigations of its application on the pulsed source is in progress [17].

These investigations have only been possible thanks to the patient and tenacious work of my colleagues at ETH. I am also very grateful to all those who have provided me with the newest informations on their work.

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Secretary's report, Session (C), T.B. Clegg:

**W. Kubischta** : After preparing your surface, how long does it last?

Answer: The $H_2$ surface at low temperature lasts one hour or so. For $N_2$, once the temperature is adjusted and proper amount of $N_2$ is found in the feed gas, the surface is stable for weeks .... as shown by S. Jaccard at SIN.