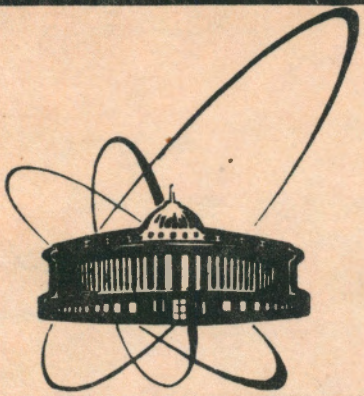

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CALORIMETRIC METHOD FOR THE MEASUREMENT OF THE DISSOCIATION EFFICIENCY

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I. INTRODUCTION

Due to the large energy release during recombination the flux of hydrogen isotopes from a dissociator can be determined calorimetrically. For example, hydrogen atoms recombine to molecules under an energy release of 4.48 eV/molecule. This energy is essentially higher than both their kinetic energy of about 25 meV/molecule (for 100 K) and their sublimation energy of about 10 meV/molecule.

In our apparatus [see: Fig.1] the hydrogen gas recombines at a liquid-helium cooled copper surface. After losing its recombination energy in multiple wall collisions molecular hydrogen (H_2) freezes out at the surface. Thus by cryopumping the hydrogen vapor pressure is kept at a level that will not hinder the flow of atoms to the cryogenic surface. The released recombination energy is determined by measuring the increase of the He evaporation rate from the helium bath during the operation of the dissociator. Without inserted cryostat the 'background' evaporation rate of the used He dewar is about 1 l/min corresponding to a heat input of about 62 mW. With installed cryostat this value increases to 73..92 mW depending on the He level in the dewar. We expect an additional heat input P_H caused by recombination of H atoms

$$P_H = E_r \cdot \varepsilon \cdot \Phi_{H2} , \qquad [1]$$

where

$$\varepsilon = \Phi_H / (2 \cdot \Phi_{H2}) . \qquad [2]$$

Here E_r = 4.48 eV/molecule is the specific recombination energy, ε is the efficiency of the dissociation and the transport of atoms to the cryosurface, Φ_{H2} is the H_2 flow into the dissociator and Φ_H is the atom flow to the cryosurface. E.g. for $\Phi_{H2} = 1 \cdot 10^{17}$ H_2/s Eq. [1] yields $P_H = \varepsilon \cdot 72$

mW. Thus we expect changes of the He evaporation from the dewar in the range of the 'background' evaporation rate for $\Phi_{H2} > 10^{17}$ H_2/s.

II. THE EXPERIMENTAL APPARATUS

The schematic diagram of the apparatus is given in Fig.1. It is especially designed to determine the efficiency of a dissociator separately, i.e. before its employment in other experiments. The dissociator is installed at the top of the cryostat. For our measurements we have used two different dissociators. The 2.45 GHz microwave dissociator and the molecular hydrogen feed system are described in more detail in /1/. Molecular hydrogen is supplied from a calibrated storage volume into the discharge tube by an electromagnetic valve. The flow into the dissociator is determined by measuring the decrease of the gas pressure p_{H2} in the storage volume (V ≈ 3 l). The second used, water cooled rf dissociator is shown in Fig.2. This dissociator was designed after the construction used by Luppov et al. /2/. It works at a frequency of 33 MHz. The rf coil is inductively coupled to the generator. The maximum rf power output of the used generator is about 300 W.

In each construction the dissociated gas flows through a orifice into a teflon tube (inner diameter $d = 2 \cdot r = 0.9$ cm, length $l = 80$ cm) served to guide the gas to the cryogenic surface. To stabilize the teflon tube mechanically and to prevent thermal contact with the cryostat at low temperatures the teflon tube was inserted into a glass tube. This tube has only one thermal contact to the cryostat at a point, where the wall temperature is about 100 K. In this way in all experiments the temperature of the teflon tube was kept at a temperature higher than 95 K as measured by means of a thermometer at the lower end of the teflon tube.

For $\Phi_H = 1 \cdot 10^{18}$ H/s a maximum density of $2 \cdot 10^{15}$ H/cm^3 at the upper end of the teflon tube is estimated. At this density the mean free path of the H atoms is larger than the tube diameter. Under these conditions the transport of the gas can be described using a Knundsen flow model /3/. This model also enables to estimate the recombination losses in the teflon tube. H atoms may recombine in the transport tube due to volume recombination, first- and second-order surface recombination. At the above-mentioned density volume recombination and second-order surface recombination can be neglected. The first-order recombination probability per wall collision is $\gamma = 2 \cdot 10^{-5}$ for temperatures higher than liquid-nitrogen temperature. The ratio α of the hydrogen flow out of the tube to the hydrogen flow into the tube can be calculated by

$$\alpha = \Phi_{out} / \Phi_{in} = [\cosh (2 \cdot \gamma \cdot N_c)^{1/2}]^{-1} \qquad [3]$$

with

$$N_c = 3 \cdot l^2 / 8 \cdot r^2 . \qquad [4]$$

Here N_c is the average number of wall collisions of hydrogen atoms in the tube. The efficiency of the dissociator ε_d is given by

$$\varepsilon_d = \varepsilon / \alpha . \qquad [5]$$

In our case $N_c = 1.2 \cdot 10^4$ is calculated by Eq. [4]. Using Eq. [3] $\alpha = 0.8$ is estimated. Therefore, ε_d is about a factor of 1.25 higher than the efficiency ε determined by recombination heating. But it should be mentioned, that the real losses due to recombination of hydrogen in the teflon transport tube can be higher than estimated, because the teflon surface can be deteriorated e.g. by oil contaminations, what leads to a decrease of α. Due to these uncertainties in the estimation of α, we use in the following only ε to describe the efficiency of the dissociator. It represents a lower limit of the real efficiency. In particular, this means no restriction for relative

measurements, e.g. for optimization of the working parameters of the dissociator.

The copper pot at the bottom of the cryostat has a inner surface of A ≈ 100 cm^2 and is cooled by the He bath. The pressure of the hydrogen gas in this copper pot, p, is given by

$$p = \Delta p + p_{back} \qquad [6]$$

with

$$\Delta p = (T / 300\ K)^{1/2} * \Phi_H / (S_H \cdot A). \qquad [7]$$

Here, p_{back} is the background pressure corresponding to the equilibrium vapor pressure of frozen H_2, Δp is the pressure increase due to the H flow, T is the temperature of the cryosurface and S_H is the pumping speed for atomic hydrogen. For T = 4.2 K the background pressure is $1 \cdot 10^{-6}$ Torr /4/. For an incoming hydrogen flux $\Phi_H = 1 \cdot 10^{18}$ H/s a pressure increase of $\Delta p = 8.2 \cdot 10^{-6}$ Torr is calculated by Eq. [7] using $S_H = 4.2 \cdot 10^{20} * \exp [(3 - T/K) / 1.12]$ atoms/(Torr·s·cm^2) /2/. Thus, by Eq. [6] the total pressure in the copper pot is estimated to be $p \leq 9.2 \cdot 10^{-6}$ Torr. This value is at least a factor of thousand lower than the pressure at the upper end of the teflon tube. Consequently the pressure at the bottom of the cryostat does not hinder the H transport from the dissociator to the cryosurface.

In order to collect most of the recombination energy in the copper pot, it was closed at the top by an annular disc [see Fig.1]. The inner diameter of the opening of the disc was about 2 mm larger than the diameter of the teflon tube, so that the teflon tube fits in without thermal contact. For the above calculated pressure of $9.2 \cdot 10^{-6}$ Torr the leakage of H atoms from the copper pot into the cryostat through the gap between the teflon tube and the annular disc is estimated to be $5 \cdot 10^{16}$ H/s, i.e. 5% of the incoming flux.

Before setting the cryostat into the He bath the cryostat was pumped out. The time to cool

down our cryostat is about four hours. After this time the evaporation rate reaches equilibrium. Since the 'background' evaporation rate depends on the He level in the dewar it has to be determined between each measurement. After switching the dissociator on or off it was necessary to wait about 15 minutes for setting of equilibrium. The change of the evaporation rate during operating the dissociator was measured with an uncertainty of about 0.04 l/min corresponding to a H flow of $1 \cdot 10^{16}$ H/s. In order to measure the heat input caused by thermalization and sublimation of the H_2 gas we also condensed H_2 into the copper pot without operating the dissociator. We found, that this heat input is less than the above-mentioned uncertainty of our measurement up to a flux of $1 \cdot 10^{18}$ H_2/s.

The amount of evaporated helium was measured by a calibrated gas flow meter. Furthermore, we measured the dependence of the He evaporation rate on the amount of electrical heating power supplied to a heater mounted at the bottom of the copper recombination pot. The result of this measurement is shown in Fig.3. We observed, that the gas meter measures only about half of the evaporation rate one would expect by calculation from the known electrical heat input [long-dashed line in Fig.3]. Probably, this effect is caused by a decrease of the 'background' evaporation rate, since forced evaporation of helium increases the gas cooling of the dewar and the cryostat tube. Thus, with increasing electrical heat input the 'background' evaporation rate decreases till it is completely compensated by gas cooling. Then, for a heat input higher than 200 mW, the evaporation rate will follow the short-dashed line in Fig.3. This line shows the calculated result for a cryostat with zero 'background' evaporation. The solid line in Fig.3 represents a fit of the measured values and was used as a calibration curve for the amount of recombination energy released at the cryosurface.

III. RESULTS

Fig.4 shows the results for the 2.45 GHz microwave dissociator with an 1 mm dia. orifice at the exit /1/. This dissociator is working in a pulsed regime. Synchronously with the start of the gas pulse (typical duration of the valve pulse: 0.5..3 ms) into the dissociator the microwave power with a 40 W peak power is switched on. The microwave pulse duration is 100 ms, matched to the characteristic time of the hydrogen outflow from the dissociator /1/. The repetition rate is 7 Hz. The measured H_2 flow into the dissociator, the H flow into the copper pot and the efficiency ε are given in dependence on the pressure p_{H2} in the storage volume. As can be seen in Fig.4, the flux of hydrogen molecules into the dissociator and, consequently, the gas density inside the dissociator can be controlled over a wide range by the pressure p_{H2}. The measured dissociation efficiency ε shows a maximum of 40% at the pressure $p_{H2} \approx 7$ Torr. This result is understandable if we consider dissociation as chain reaction similar impact ionization. At low densities the probability, that an accelerated ion will hit a hydrogen molecule, is too small to achieve an essential dissociation effect. At densities higher than the optimal one the mean free path becomes to small to accelerate ions to energies higher than the dissociation energy of hydrogen molecules and the dissociation process will be chocked. First, the obtained H flow from the dissociator increases with increasing pressure. Than it saturates at a value of about $2 \cdot 10^{17}$ H/s. The saturation occurs at higher pressures (in our case at about 15 Torr), when a further increase of the H_2 flow is compensated by the decrease in dissociation efficiency. Since for a given feedrate of molecular hydrogen the gas density in the dissociator is determined by the flow impedance of the orifice, it should be possible to move the maximum in ε to a higher feed rate of molecular hydrogen by increasing the diameter of the orifice. This would cause an increase

of the saturation value of the achieved H flux from the dissociator.

Results for the rf dissociator are shown in Fig.5 and Fig.6. The molecular hydrogen is supplied into the discharge tube in the same way as described for the microwave dissociator. The repetition rate of the electromagnetic inlet valve was varied between 7 and 10 Hz. The rf field is supplied continuously with an outcoming electrical power of about 200 W from the generator. Fig.5 shows the results for a 2 mm dia. orifice. In this case we observed a maximum H flow of $4 \cdot 10^{17}$ H/s and a maximum efficiency of 54% at a pressure of about 3 Torr. In Fig.6 the results for a larger, 3 mm dia. orifice are shown. Moreover, in this experiment the H_2 feedrate into the dissociator for a certain pressure in the H_2 storage volume was reduced approximately by a factor of two. Both effects cause a shift of the maximum in the dissociation efficiency to higher pressures as shown in Fig.6. Under these conditions a maximum dissociation efficiency of 75% has been achieved. Furthermore, a maximum H flow of $5 \cdot 10^{17}$ H/s has been measured; 20% higher than for the dissociator with the 2 mm dia. orifice.

IV. SUMMARY

An apparatus to measure the dissociation efficiency of a hydrogen dissociator calorimetrically has been built and tested. Due to the uncertainties in the determination of: (1) recombination losses during transport of the dissociated gas to the cryosurface and (2) the fraction of recombination energy collected in the recombination pot, the method only allows to measure a lower limit of the efficiency. Nevertheless, the described method is especially suitable for relative measurements, e.g. for optimizing the working regime of a dissociator separately. Two dissociators were tested with measured maximum dissociation efficiencies from 40% to 75%. Maximum atom fluxes between $2 \cdot 10^{17}$ H/s and $5 \cdot 10^{17}$ H/s have been achieved.

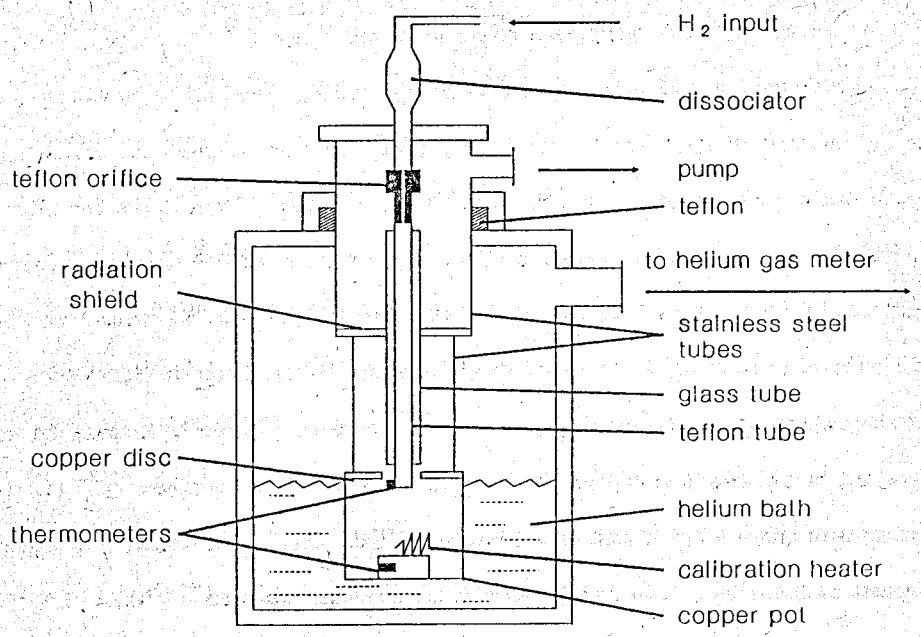

Fig.1. Schematic diagram of the cryostat and the He dewar

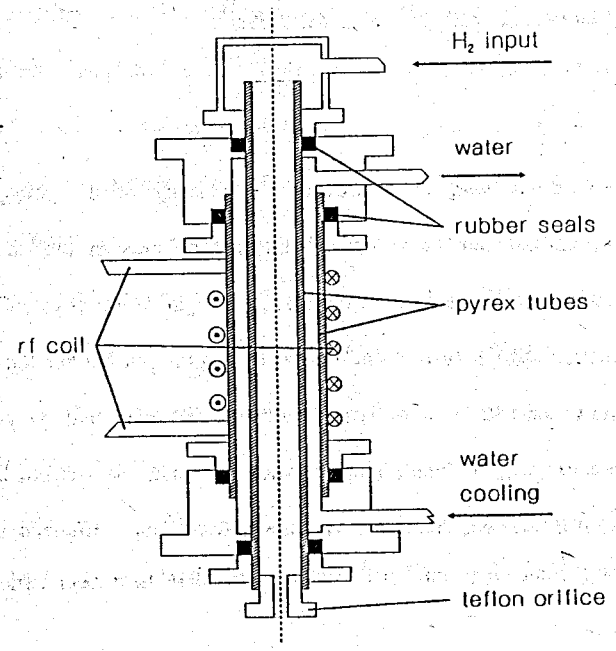

Fig.2. Schematic diagram of the rf dissociator

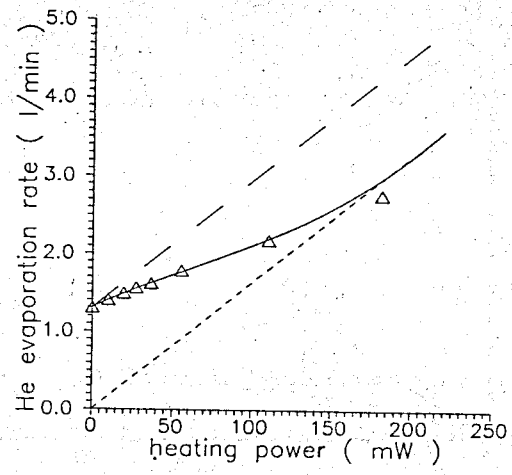

Fig.3. Dependence of the helium evaporation rate from the dewar on the electrical heating power supplied to a heater mounted at the recombination pot: solid line - fit of the measured values, long-dashed line - calculated for a 'background' evaporation rate of 1.3 l/min, short-dashed line - calculated for zero 'background' evaporation.

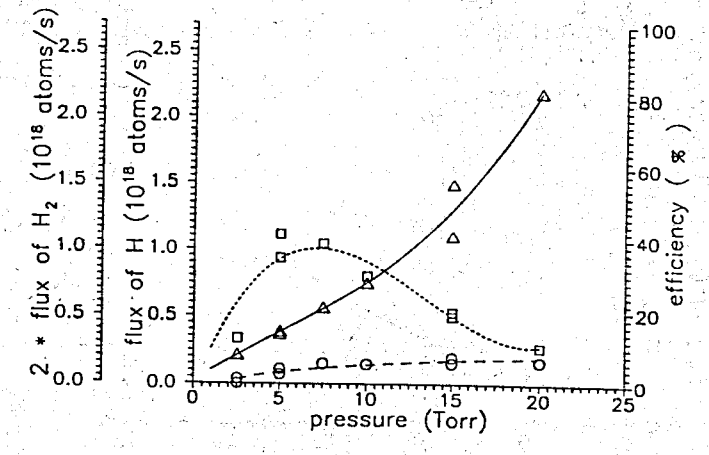

Fig.4. Results for the microwave dissociator: The feedrate Φ_{H2} of molecular hydrogen into the dissociator (△, solid line), the flux Φ_H of dissociated hydrogen atoms into the cryostat (○, long-dashed line) and the dissociation efficiency ε (□, short-dashed line) in dependence on the pressure p_{H2} in the storage volume.

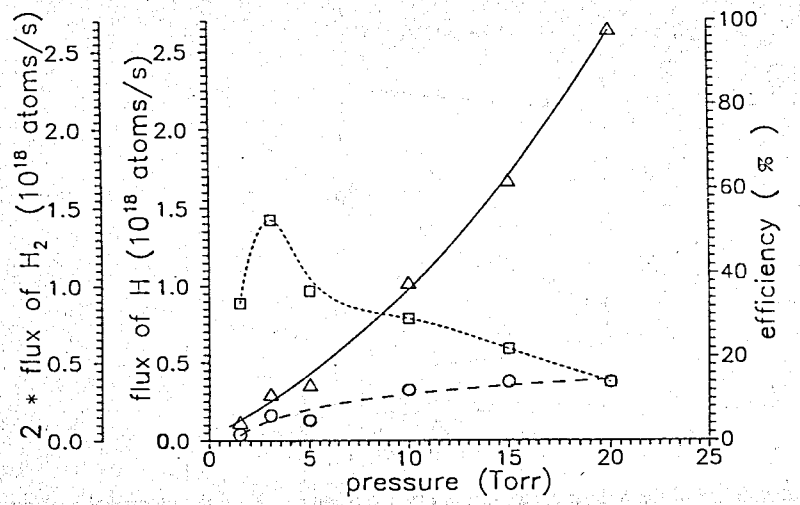

Fig.5. Results for the rf dissociator with a 2 mm dia. orifice: same symbols as in Fig.4.

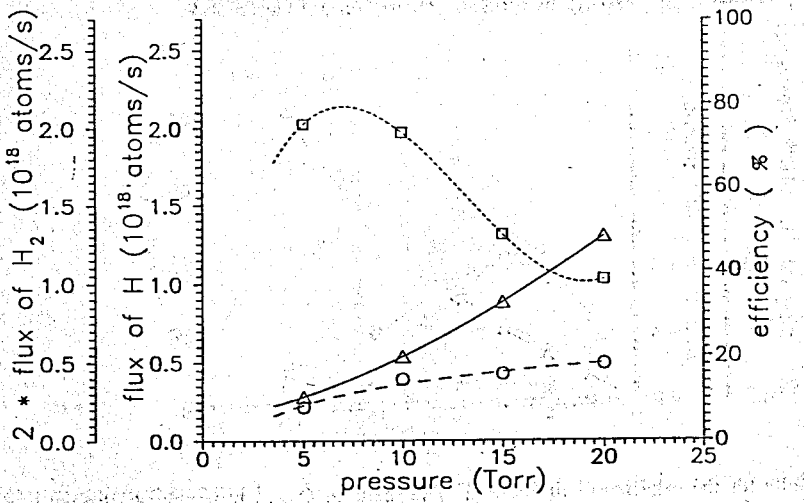

Fig.6. Results for the rf dissociator with a 3 mm dia. orifice: same symbols as in Fig.4.

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