объединенный
институт
ядерных
исследований
дубна

E15-92-82

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MEASUREMENTS OF THE $p\mu d$-MOLECULE FORMATION RATE AT DIFFERENT HYDROGEN TEMPERATURE

Submitted to "Muon Catalyzed Fusion"

1992

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Измерения скорости образования мюонных молекул $p\mu d$ в зависимости от температуры водорода

В опытах с криогенной мишенью высокого давления (0,6 кбар) на мюонном пучке фазотрона ОИЯИ измерены временные распределения гамма-квантов из реакции $p\mu d \rightarrow {}^3He\mu + \gamma + 5,5$ МэВ в смеси H_2 + 1,7%D_2 в диапазоне температур T = 21-300 К. Из их анализа найдены значения скорости $\lambda_{p\mu d}$ образования молекул $p\mu d$ и скорости ядерной реакции p + d в системе $p\mu d$. Полученные данные свидетельствуют об отсутствии зависимости величины $\lambda_{p\mu d}$ от температуры. Усредненное по всем температурам значение $\lambda_{p\mu d} = (5,49 \pm 0,30)$ мкс$^{-1}$ хорошо согласуется с теорией и другими измерениями, выполненными при T=20 K и T=300 K.

Работа выполнена в Лаборатории ядерных проблем ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1992

Dzhelepov V.P. et al. E15-92-82

Measurements of the $p\mu d$-Molecule Formation Rate at Different Hydrogen Temperature

In experiments with a cryogenic target of high pressure (0,6 kbar) on the muon beam of the JINR phasotron the time distributions of γ-quanta from the reaction $pd\mu \rightarrow {}^3He\mu + \gamma + 5.5$ MeV have been measured in the temperature region T = 21-300 K. From their analysis the values of the $p\mu d$-molecule formation rate $\lambda_{p\mu d}$ and the p + d nuclear fusion rate have been found. The data obtained show $\lambda_{p\mu d}$ to be independent of temperature. The value $\lambda_{p\mu d} = (5.49 \pm 0.30)$ μs^{-1} averaged on all temperatures is in good agreement with theory and previous measurements made at T=20 K and T=300 K.

The investigation has been performed at the Laboratory of Nuclear Problems, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1992

The study of the muon catalyzed pd-cycle in a protium-deuterium mixture is interesting, first of all, from the point of view of the existence of the spin dependence of the reactions,

$$p d\mu \longrightarrow {}^3He\mu + \gamma + 5.5 \text{ MeV}, \qquad (1a)$$

$$ {}^3He + \mu + 5.3 \text{ MeV}, \qquad (1b)$$

which can go from the states with two values of the total proton and deuteron spin: $S_{pd}=3/2$ and $S_{pd}=1/2$. Varying the experimental conditions one can change the populations of these states and hence change the yield of reaction (1) (the Gerstein- Wolfenstein effect) [1-3]. The results of recent investigations of this effect and their interpretation including new calculations [4,5] of p+d fusion rates are comprised in papers [5,6]. Note that there is a problem of understanding the results of the absolute γ- yield obtained in a series of previous experiments.

Another important feature of the problem is connected with the possible dependence of the pμd-molecule formation rate ($\lambda_{p\mu d}$) on the hydrogen temperature. In refs.[7] one pointed out the possible resonance (in dμ-atom kinetic energy) behavior of the pμd-system formation cross section caused by the existence of weakly bound levels with abnormal parity $P=1^I$ (I is the total orbital moment of the pμd-system) in the pμd-molecule. Such a state with an energy 3.5 eV has been established in calculations [8,9]. However the question on what formation rate of the pμd-molecule could be in this state has not been studied.

In refs.[10,11] the authors assumed, that the population of the pμd-molecule levels with the different values of the total nuclear spin can be changed in collisions of the molecular complex containing the ion $(p\mu d)^+$ with rather energetic hydrogen molecules and it induces the change of reaction (1) yield with a temperature.

Previously the values of $\lambda_{p\mu d}$ were measured in liquid hydrogen at T=20 K [3,6,12] and in gaseous hydrogen at T=300 K [13]. The results of these experiments are presented in table 1.They coincide with each other. However, the authors of ref. [14] observed a noticeable change in the reaction (1a) yield with a temperature in experiment at TRIUMF.

Taking into consideration these circumstances we believed it to be important to measure the temperature dependence of of the pμd- molecule formation rate in a H+D mixture.

The scheme of the processes caused by negative muons in a $C_pH_2 + C_dD_2$ mixture, where C_p and C_d are the partial concentrations of protium and deuterium molecules $(C_p+C_d=1)$ is presented in fig.1, and the corresponding values of kinetics parameters are given in table 1.

As usually, the values of the spin-flip rate $F_{d\mu}=3/2 \dashrightarrow F_{d\mu}=1/2$ (λ_d) and the pdμ-molecule formation rate $(\lambda_{p\mu d})$, are normalized to the liquid hydrogen density $n_0=4.25\text{x}10^{22}$ atoms per cm^3. Their "current" values for real experimental values of hydrogen density and deuterium concentration are expressed

as $\Lambda_d = \lambda_d C_d \phi$ and $\Lambda_{p\mu d}=\lambda_{p\mu d}(1-C_d)\phi$, where $\phi=n/n_0$ is the relative hydrogen density.The values of the fusion rate $\lambda_{f,\gamma}^{1/2}$ was found in refs.[3,12,17] from the analysis of experimental data on the yield of reaction (1a) on the assumption that $\lambda_{f,\gamma}^{3/2} = 0$.

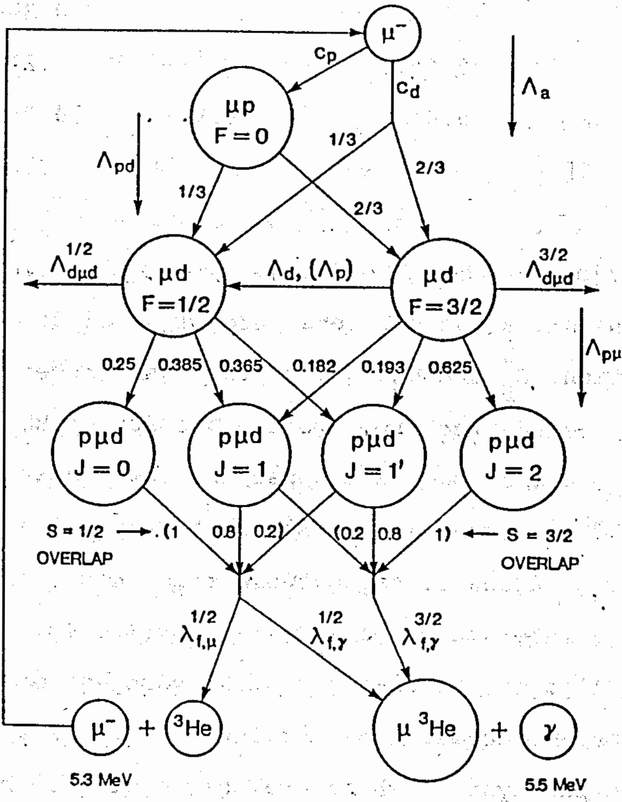

Fig.1. Scheme of the processes caused by negative muons in H_2+D_2 mixture (taken from ref.[6]).

Table 1

Kinetics parameters of the processes involved in muon catalyzed pd-cycle (in μs^{-1}).

Value	λ_d	$\lambda_{p\mu d}$	$\lambda_{f,\gamma}^{3/2}$	$\lambda_{f,\gamma}^{1/2}$	
	30-40				[15,16]
		5.8(3) [3]	-----	0.305(10)	[3]
		5.53(16)			[13]
Experiment			-----	0.287(22)	[17]
		5.9(9)	-----	0.289(27)	[12]
		5.6(2)	0.11(1)	0.35(2)	[6]
Theory	40-50[18]	5.6 [19]	0.11 [4]	0.39 [4]	

Stopping in the H+D mixture, muons form $p\mu$- and $d\mu$-atoms with initial populations proportional to the partial concentrations C_p and C_d. Due to the fast process $p\mu+d$-- $d\mu+p$ muons are transferred from protium to deuterium in time $t \cong 10^{-10}/(\phi C_d)$ s and populate upper and lower states of the $d\mu$-atom hyperfine structure (HFS) with statistical weights 2/3 and 1/3. The transitions between them can occur in further spin-exange interactions $d\mu(3/2)+d \rightarrow d\mu(1/2)+d$.

In collisions of $d\mu$-atoms with hydrogen molecules they can form $p\mu d$-molecules. The populations of the four states of HFS of the $p\mu d$-system, which are characterized by different effective fusion rates, depend on the $d\mu$-atom spin state. That is why the yield of the p+d reaction and the character of the time distribution of its products are determined not

only by the values of $\lambda_{p\mu d}$, $\lambda_{f,\gamma}^{3/2}$ and $\lambda_{f,\gamma}^{1/2}$, but also by the spin-flip transition rate λ_d.

In the present experiment the characterictics of γ-radiation from the p+d fusion reaction in the $p\mu d$-molecule have been studied. The measurements were made on the muon beam of the JINR phasotron using the same experimental technique as one employed in our previous works [15,20,21], but one of two neutron detectors was replaced by a γ-spectrometer (NaJ(Tl), ϕ 150 mm x100 mm). The registration efficiency of γ-quanta from reaction (1a) was equal to $\varepsilon_\gamma \cong 3\%$.

A cryogenic target of high pressure [22] (0.6 kbar) was used in the experiment. It was placed in a cryostat which allowed the target temperature to be kept in the range 20-300 K with an accuracy 1.5 K. Before the run the target was completely filled with a liquid 98.3% H_2+ 1.7% D_2 mixture at T=21 K (the first exposure). With this amount of hydrogen (ϕ=1,0) the measurement were made at T=21-81 K. Then a part of gas was let out from the target, and measurements were performed at T=170 K. After this the next part of gas was let out and other exposures were performed. The experimental conditions (hydrogen temperature and density) are shown in table 2. Numbers of exposures are given according to the measurement order.

The γ-quanta from reaction (1a), the neutrons from reaction $d+d \rightarrow {}^3He+n$ in the $dd\mu$-molecule and the electrons from decay of muons stopping in the target were detected in the runs. For further computer analysis the events were selected for which the neutron or γ-quanta signal and sequent electron signal are required in the time interval T=10μs after a muon stop in the target. For discrimination of the background caused by muon stops in the target walls an additional necessary condition t_e-t_o>0.5 μs was included in the electronic trigger (t_e is the time of electron detection and t_o is the moment of muon stop).

Table 2

The parameters of the main exposures with H_2+D_2 mixture.

Temperature,K	21	48	81	170	94	165	220	302
Density, ϕ	1.0	0.99	0.99	0.67	0.41	0.41	0.41	0.41
Number of γ-quanta, N_γ	4356	2959	3303	3197	1825	2020	1987	1872
Number of electrons, N_ex10^{-3}	309	211	247	235	146	153	159	156

In the present experiment we used the following method of the determination of hydrogen density. The "current" values of density were delivered by using the values of a normalized electron yield (corresponding to exponent with a factor τ=2.2 μs) measured in different exposures. The value of this yield for the first exposure (T=21 K, ϕ=1), where the density was well known, was used as a reference. The yield of electrons from μ-decay in the target walls (τ=0.2 μs) was used for normalization. Note that for measurements with the same amount of hydrogen in the target the values of electron yield did not differ from one exposure to another with an accuracy of 1%.

To determine the background of gamma-quanta, neutrons and electrons the expousures with helium and with an empty target were performed. The numbers of γ-quanta (N_γ) pointed out in table 2 correspond to the criterion t_e-t_γ =(0.5÷2.5) μs used in the final analysis. The background was subtracted from these values. The relative part of the background did not exceed 5%. The value of N_e means the number of electrons detected by the gamma - detector. Fig.2 and 3 show the amplitude distribution and time spectrum of γ- quanta respectively. They are recorded in the exposure at T=21 K. The character of each of these distributions is in good agreement with the expected one.

The abundance of admixture with Z>1 (C_Z) in hydrogen was determined from the analysis of electron time spectra. They are fitted by an expression dN_e/dt $\exp(-\lambda_e t)$. It turned out that for the first five exposures the value of λ_e coincides with the free muon dissapearance rate λ_o=0.455 μs^{-1} with an accuracy better than 0.4%. It means that a probability of muon transfer from dμ-atom to admixtures did not exceed 2-3%. However for other exposures this probability was found to be ≅ 7-8%. The corresponding corrections were made in the final results.

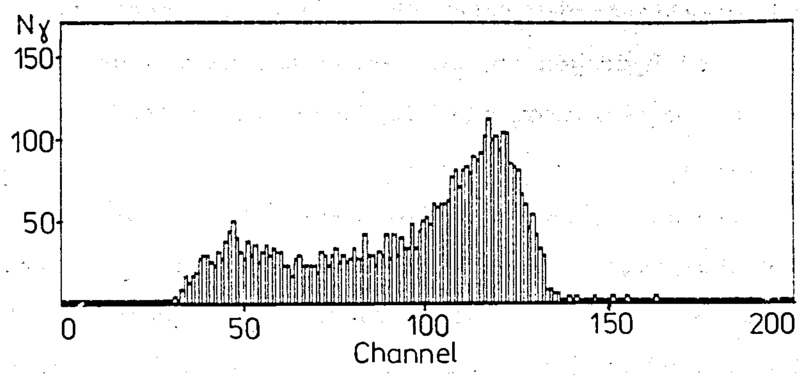

Fig.2. Amplitude distribution of 5.5 MeV γ-quanta from reaction (1a) measured by a NaJ(Tl) detector in the exposure at T=21 K.

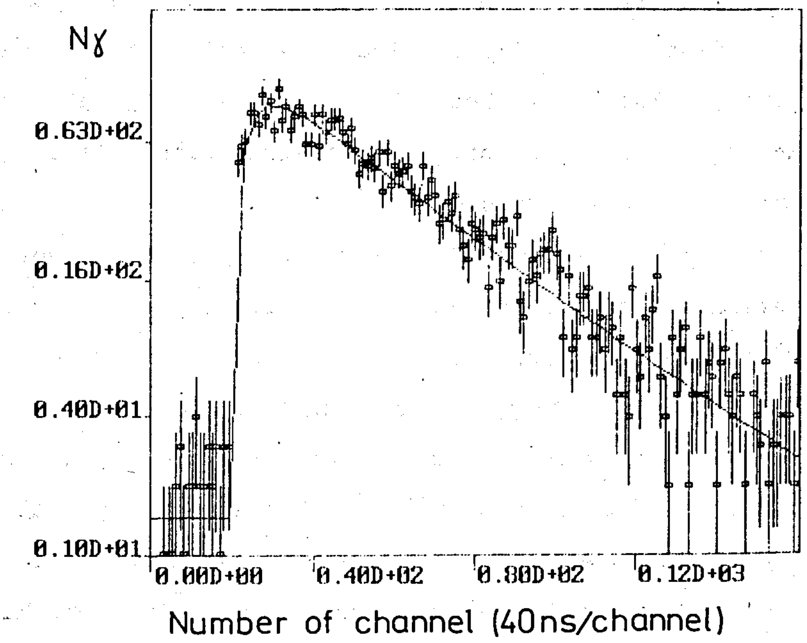

Fig.3. Time distribution of γ-quanta from reaction (1a) for exposure at T=21 K. The curve represents function (3) with optimal parameters found from a fit.

The values of the relative gamma - yield $\eta_\gamma = N_\gamma/(N_e\varepsilon_\gamma)$ obtained from the data of table 2 are presented in fig.4. The mean values of the yield averaged over all exposures with a density ϕ=0.41 (solid line) and with a density ϕ=1 (dashed line) are shown in the figure. The difference between them is in agreement with the one obtained from calculations taking into account full kinetics of the considered processes. Approximately (with an accuracy of 1- 2%) this difference factor can be estimated from the following expression for the pμd-molecule formation probability :

$$W_{p\mu d} = \lambda_{p\mu d}(1-C_d)\phi/[\lambda_0+\lambda_{p\mu d}(1-C_d)\phi+\Lambda_Z] . \qquad (2)$$

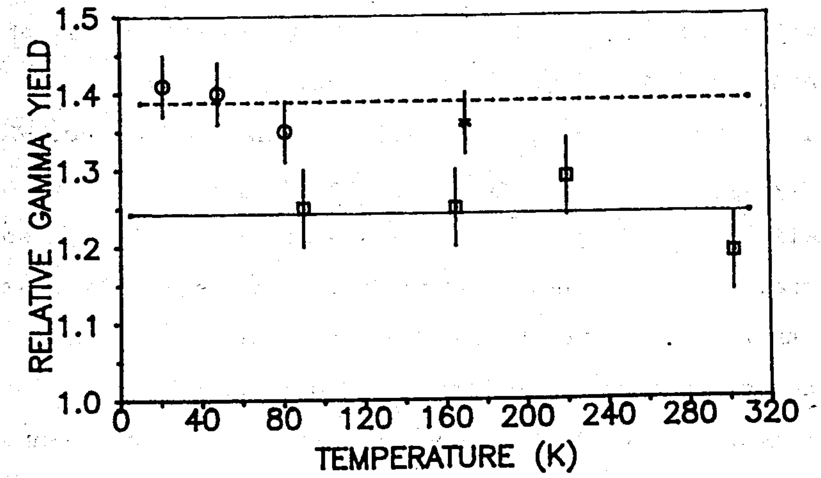

Fig.4. Relative γ-yield as a function of H_2+D_2 mixture temperature. Symbols : ○- data for ϕ=1; X - ϕ=0.67; □ - ϕ=0.41. The dashed line is the value of η_γ averaged over the exposures at ϕ=1, the solid one is the same for ϕ=0.41.

It follows from eq.2 that under the conditions of our experiment (high hydrogen density) the value of η_γ is slightly sensitive to the pμd-molecule formation rate, while it is very sensitive to $\Lambda_Z=\lambda_Z C_Z \phi$. This circumstance allows the reliable determination of the corrections caused by the muon transfer to impurities from the common fit of the electron and γ-quanta time distributions and of the data on γ-yield.

A rather complicated system of differential equations corresponds to the scheme of the processes displayed in fig.1. It allowed only numerical solutions for which the computer code developed in paper [23] was used in ref. [6]. In our analysis of γ- quanta time spectra we used the analytical solution $y_\gamma(t)$ obtained in approximation of the absence of muon recycling in reaction (1). We took into account the fact that the yield of the p+d reaction is relatively small (25%) and the probability of muon sticking to helium-3 fusion products is large ($\omega_{pd} \cong 0.85$ [5,6]), so the integral effect of recycling is estimated to be only 3-4%.

As seen from fig.1, dμ-atoms can form not only pμd-molecules but also ddμ-molecules. Under the conditions of our experiment ($C_d \ll 1$) the contribution of this channel is suppressed by a factor of $\cong 10^2$ relative to the studied process (1). With allowance for almost 100% recycling of muon in the d+d reaction, there was no influence of the ddμ-molecule formation on the main parameters of process (1). As to the registration of neutrons from reaction $d+d \rightarrow {}^3He+n$ we could not obtain the reliable data on the parameters of their time spectra due to smallness of the effect.

The expression for the γ-quanta time distribution can be presented in the following form

$$y_\gamma(t) = \Lambda_{p\mu d}\,(y_{\gamma,F=3/2}(t) + y_{\gamma,F=1/2}(t)),$$

$$y_{\gamma,F} = \sum C_{F,J}\, Z_J(t)\, [a_{1,F}\, V_J(t) + a_{2,F}\, U_J(t)], \qquad (3a)$$

$$Z_J = \lambda_{f,\gamma}^{J} \exp[-(\lambda_0 + \lambda_{f,\gamma}^{J} + \lambda_{f,\mu}^{J})\, t],$$

$$V_J = [1 - \exp(-\beta_J t)]/\beta_J\,, \qquad \beta_J \equiv \Lambda_{p\mu d} - (\lambda_{f,\gamma}^{J} + \lambda_{f,\mu}^{J}),$$

$$U_J = [1 - \exp(-\alpha_J t)]/\alpha_J\,, \qquad \alpha_J \equiv \Lambda_{p\mu d}+\Lambda_d + \Lambda_d' - (\lambda_{f,\gamma}^{J} + \lambda_{f,\mu}^{J}).$$

Here $C_{F,J}$ are populations of the HPF states of the pμd - molecule [24];

λ_F^J are partial fusion rates in these states;

$a_{i,F}(i=1,2)$ are partial coefficients in functions of populations of dμ-atom spin states:

$$n_F(t)=a_{1,F}\exp[-(\lambda_0+\Lambda_{p\mu d})t] + a_{2,F}\exp[-(\lambda_0+\Lambda_{p\mu d}+\Lambda_d+\Lambda_d')t],$$

$$a_{1,1/2} = \Lambda_d/(\Lambda_d+\Lambda_d'), \qquad a_{2,1/2} = -(2\Lambda_d-\Lambda_d')/3(\Lambda_d+\Lambda_d'), \qquad (3b)$$

$$a_{1,3/2} = \Lambda_d'/(\Lambda_d+\Lambda_d'), \qquad a_{2,3/2} = - a_{2,1/2};$$

$$\Lambda_d' = 2\,\Lambda_d \exp(-560/kT) \qquad (3c)$$

- the rate of inverse transitions F=3/2→F=1/2.

To estimate the distortion caused by neglect of recycling we have considered:

- the exact solution for the case which is characterized only by one ("effective") fusion rate ;
- the γ-quanta time distributions obtained by the Monte-Carlo code with taking into account the full kinetics of the considered processes.

It follows from both cases that neglect of the muon recycling does not practically influence the value of $\lambda_{p\mu d}$ (with an accuracy of 1-2%) but leads to a decrease in the fusion rate by 7-8%. Corresponding corrections were included into the final results.

In the analysis of time spectra of γ-quanta using the formulae (3) the values of density (ϕ) and temperature (T) were taken according to the experimental conditions, λ_d - according to the data obtained in measurements [15,16] and $\lambda_{f,\mu}=0.056\ \mu s^{-1}$- according to ref.[5]. The values of $\lambda_{p\mu d}$ and $\lambda_{f,\gamma}^{1/2}$ were determined from the fit.

The analysis of the data of papers [3,12,17] was made on the assumption $\lambda_{f,\gamma}^{3/2}=0$, moreover in ref.[17] they used the value of $\lambda_{p\mu d}$ found independently by the "direct" method in ref. [13]). That is why we analysed our data (for the first four exposures) for the case $\lambda_{f,\gamma}^{3/2}=0$ too. The values of $\lambda_{p\mu d}=(5.78\pm0.29)\ \mu s^{-1}$ and $\lambda_{f,\gamma}^{1/2}=(0.319\pm0.13)\ \mu s^{-1}$ (statistical errors) were obtained which are close to the results of the indicated ref. [3,12,13,17].

In the final analysis the value of $\lambda_{f,\gamma}^{3/2}=0.11\ \mu s^{-1}$ was fixed according to theory [4,5] and experiment [6]. The value of the partial fusion rate $\lambda_{f,\gamma}^{1/2}$ was varied together with the value of $\lambda_{p\mu d}$ in the analysis of the data obtained in the runs with high mixture density $\phi=1$ and $\phi=0.67$. The averaged value $\lambda_{f,\gamma}^{1/2}=(0.397\pm0.022)\ \mu s^{-1}$ was used in the analysis of the data obtained at lower density $\phi=0.41$. After the necessary corrections this value was estimated to be

$$\lambda_{f,\gamma}^{1/2}=(0.426\pm0.024)\ \mu s^{-1},$$

which is in rather good agreement with the calculated value [4]

$\lambda_{f,\gamma}^{1/2}=0.390\ \mu s^{-1}$, but it is larger than the measured value $\lambda_{f,\gamma}^{1/2}=(0.350\pm0.020)\ \mu s^{-1}$ [6].

The values of $\lambda_{p\mu d}$ obtained by us for different H_2+D_2 mixture temperatures are given in fig.5. The solid line in it corresponds to the value

$$\lambda_{p\mu d} = (5.49 \pm 0.19 \pm 0.23)\ \mu s^{-1}, \qquad (4)$$

averaged over all exposures. Here the first uncertainty is a statistical error and the second one is due to inexact knowledge of the deuterium concentration and of admixture abundance, and to some other factors. As seen from fig.5, the experimental results do not show any temperature dependence of the value of $\lambda_{p\mu d}$.

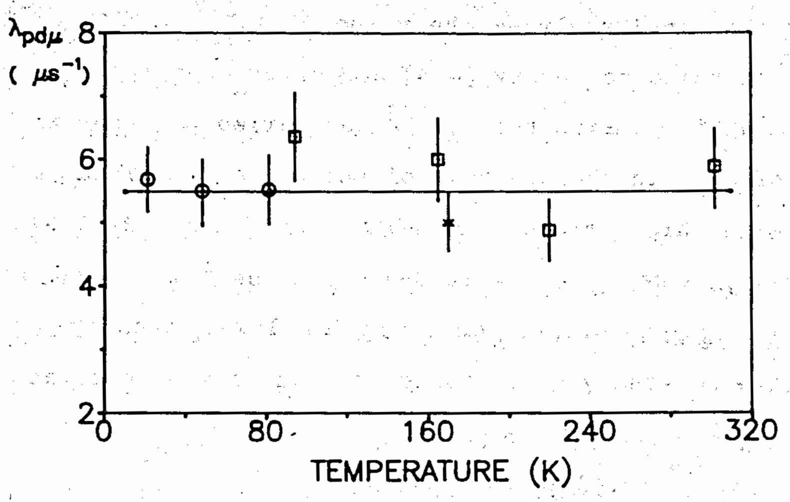

Fig.5. Values of $\lambda_{p\mu d}$ measured at different temperatures of H_2+D_2 mixture. Symbols are the same as in fig.4. The line corresponds to the averaged value (4).

The possible influece of the effect of dμ-atom thermalization was estimated from the fit of γ-quanta time distributions in which the inverse transition rate Λ_d' was taken for T=2000 K (see eq.3c). It turned out that in this case the value of $\lambda_{p\mu d}$ is decreased by 3%.

Combining the stastical and systematic uncertainties in (4) we get

$$\lambda_{p\mu d} = (5.49 \pm 0.30)\ \mu s^{-1},$$

which is in very good agreement both with theory and with previous experiments [3,6,12,13].

The authors are indebted to V.B. Belyaev, M.P. Faifman, V.E. Markushin and A.V. Matveenko for useful discussions.

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Received by Publishing Department

on March 2, 1992.