

# Measurement of the rate of charge exchange of muonic atoms of deuterium in tritium

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An ionization chamber recording charged products of  $dt$  and  $dd$  fusion has been used to measure the rate of charge exchange of muonic atoms of deuterium in tritium from the ground state of the  $d\mu$  atoms. This rate, referred to the density of liquid hydrogen, is  $\lambda_{dt}^0 = (2.8 \pm 0.2) \cdot 10^8 \text{ sec}^{-1}$ . In addition an upper limit  $Y \leq 3\%$  has been obtained for the relative probability of charge exchange from excited states of the  $d\mu$  atom under the following conditions of the experiment: pressure of gas mixture 84.3 atm, tritium content 1.24%, and temperature 296 K.

In this article we investigate the muon catalysis of the  $dt$  fusion reaction. We report measurements of the rate of charge exchange  $\lambda_{dt}^0$  from the ground state of the  $d\mu$  atom to the ground state of the  $t\mu$  atom:



In addition we give an experimental estimate of the relative contribution of charge exchange from excited states of  $d\mu$  atoms to the corresponding states of  $t\mu$  atoms:

$$Y = \sum_{n \geq 2} Y_n,$$

where  $Y_n$  is the relative yield of the reaction



The quantity  $Y$  is related in an obvious way to the population  $q_{1s}$  of the ground state of the  $d\mu$  atoms:  $q_{1s} = 1 - Y$ .

A theoretical treatment of the process (1) was carried out in Ref. 1. According to these calculations,  $\lambda_{dt}^0 = 2 \cdot 10^8 \text{ sec}^{-1}$ . Here  $\lambda_{dt}^0$  is the rate of charge exchange normalized to a tritium density equal to the atomic density of liquid hydrogen  $\rho_0 = 4.25 \cdot 10^{22} \text{ atoms/cm}^3$ . A possible role of reaction (2) was first pointed out in the theoretical study by Men'shikov and Ponomarev.<sup>2</sup> In a later publication by the same authors it is mentioned in addition that inclusion of ternary collisions can lead to a further increase of the rate of charge exchange from excited states of the  $d\mu$  atom.

At the present time two experiments have been carried out on measurement of the rate of charge exchange  $\lambda_{dt}^0$ . In the work of the JINR group<sup>4</sup> measurements were made for a density of the deuterium-tritium mixture  $\varphi = 0.01-0.08$  and for a concentration  $c_t = (0.8-7.8)\%$ ; here  $\varphi = \rho/\rho_0$ , where  $\rho$  is the atomic density of the gas mixture. In the papers of the Los Alamos group<sup>5,6</sup> the density of the gas mixture was  $\varphi = 0.1-1.2$ , and the tritium concentration was  $c_t = (4-90)\%$ . It should be mentioned that in actuality in these experiments the ratio  $\lambda_{dt}^0/q_{1s}$  was measured. However, for a small value of  $c_t$ , the quantity  $q_{1s} \rightarrow 1$ . On this assumption the result of the Dubna experiment<sup>4</sup> is  $\lambda_{dt}^0 = (2.9 \pm 0.4) \cdot 10^8 \text{ sec}^{-1}$ . Jones *et al.*,<sup>6</sup> in order to find the quantity  $\lambda_{dt}^0/q_{1s}$ ,

selected data at small  $c_t$ . Then for description of the data they used the dependence  $q_{1s}(c_t)$  proposed by them. As a result they found

$$\lambda_{dt}^0 = [1 + (6 \pm 1) \cdot 10^{-4} T] (2.8 \pm 0.4) \cdot 10^8 \text{ sec}^{-1};$$

here  $T$  is the temperature in kelvins. This result practically coincides with those of Ref. 4.

The previous measurements were carried out by detection of the neutrons from  $dt$  fusion. The efficiency for detecting neutrons in these experiments was only about 1–3%, and therefore what was measured was not  $\lambda_{dt}^0$ , but the ratio  $\lambda_{dt}^0/q_{1s}$ . In our work we used the method of detecting  ${}^4\text{He}$  nuclei by means of a pulsed ionization chamber at high pressure. This method provides a sufficiently high efficiency for detection of  $dt$  fusion events and for this reason permits independent measurement of the two quantities:  $\lambda_{dt}^0$  and  $q_{1s}$ . The method has previously been used successfully for study of  $dd$  fusion.<sup>7</sup> However, work with tritium required a substantial modification of the method. The main problem is due to the fact that the  $\beta$  decay of tritium in the sensitive volume of the ionization chamber leads to the appearance of a noise current at the chamber electrodes. As a result the energy and time resolution is much poorer. In addition, the threshold for detection of signals turns out of necessity to be so high that this leads to a reduction of the efficiency for detection of  $dt$  fusion events. To improve the conditions for observation of signals, the anode was divided into 19 independent electrodes. The noise current in each electrode was reduced, while the useful signals remained at the previous level, since the range of  ${}^4\text{He}$  nuclei under our conditions is less than the size of an individual electrode. As a result the signal-to-noise ratio rose by  $(19)^{1/2}$  times. The efficiency for detection of  ${}^4\text{He}$  nuclei turned out to be close to 90% and, in addition, it can be rather reliably calculated by the Monte Carlo method.

The experiment was carried out under the following conditions: the pressure of the ( $D-T$ ) mixture was 84.3 atm ( $\varphi = 0.0931$ ), the tritium content was  $c_t = (1.24 \pm 0.02)\%$ , and the temperature was  $T = 296 \text{ K}$ .

Figure 1 shows a diagram of the experimental apparatus, which was placed in the meson beam of the synchro-

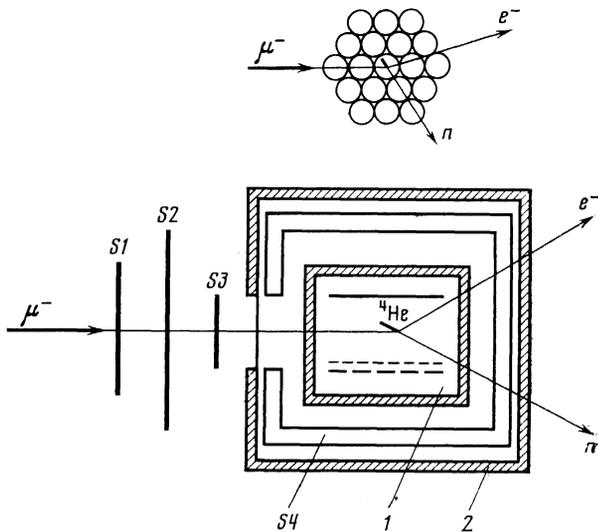


FIG. 1. Diagram of the experimental apparatus: S1–S4 are scintillation counters, 1 is an ionization chamber, 2 is the outer shield. In the upper part of the figure we have shown the geometry of the anode.

clotron at the Leningrad Institute of Nuclear Physics. Muons with momentum  $70 \pm 7$  MeV/c passed through thin scintillation counters S1–S3 and through the window of an outer shield and the wall of the ionization chamber. The thickness of material in the muon path corresponded to the maximum of muon stoppings in the sensitive volume of the ionization chamber. The trigger of stoppings was the signal  $TR = S1S3\bar{S4}$ . Counter S2 was used to monitor the absence of a second muon during the time of detection of events. The diameter of the cathode of the ionization chamber was 30 mm, the distance from the anode to the grid was 10 mm, and from the anode to the grid 0.5 mm; the diameter of each of the 19 anodes was 3.1 mm. The grid was wound of wire 0.025 mm in diameter with a 0.2 mm pitch. The potentials on the anode, grid, and cathode were chosen to be 0 kV,  $-4$  kV, and  $-30$  kV. The chamber was filled initially with tritium previously purified and adsorbed in a special container. The quantity of tritium was determined from the pressure in the chamber. Then deuterium of special purity was added; the purity was monitored also on the basis of the position of the  $\alpha$  line from a source deposited on the cathode. The electric field strength in the cathode-grid volume corresponded to 40% recombination of the number of ion pairs produced in the (D–T) mixture by a  ${}^4\text{He}$  particle with energy 3.53 MeV.<sup>7</sup> Signals from the anode were fed to 19 independent channels. In each of them it was possible to record up to four consecutive signals with measurement of the height of each signal, its duration, and the time of its appearance. The duration of the signals was 0.1–0.3  $\mu\text{sec}$ , depending on the orientation of the  ${}^4\text{He}$  track (the length of the track was approximately 1 mm). We took as the start signal the coincidence of the trigger with any of the anode signals:  $ST = TR \cdot OR$  (anodes). All signals which appeared in an interval of 10  $\mu\text{sec}$  after ST were digitized and recorded on magnetic tape. The ST signal could be produced by a fusion event or by noise operation of the anode-signal shapers (the noise rate in the shapers was  $\approx 100$  pulses/sec). However, events in which at least one additional signal was recorded after the ST signal on the same anode corresponded mainly to fusion events.

In Fig. 2 we have shown the pulse-height spectrum of

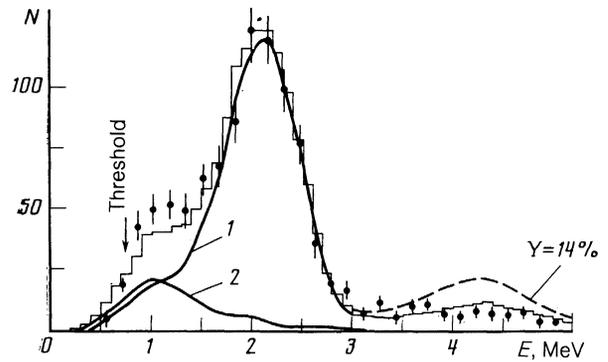


FIG. 2. Pulse-height spectrum of the third events of  $dt$  and  $dt$  fusion. The points are the data of the experiment, and the histogram is the result of a Monte Carlo calculation. The solid lines separate the contributions of the  $dt$  and  $dd$  fusion channels (respectively 1 and 2). The dashed line shows the expected yield of events with pileup as a result of the fast channel  $d\mu(n) + t \rightarrow t\mu(n) + d$  with  $Y = 14\%$ .

events in the case in which three signals were recorded successively on a single anode. These events were minimally distorted by noise. The histogram shows the spectrum obtained by the Monte Carlo method. As can be seen, the description of the pulse-height distributions is quite satisfactory. To check the calculations we used the spectra of  $dd\mu$  catalysis events obtained in a special experiment with pure deuterium. In this case, using the known value of the  $dd\mu$  catalysis rate,<sup>7</sup> we could directly measure the efficiency for detection of  $dd$  fusion events. The result of the measurements agreed satisfactorily with the calculation. In the further analysis we used the following values of the detection efficiency for  $dd$  and  $dt$  fusion events:  $\varepsilon_{dd} = 0.14 \pm 0.03$ ,  $\varepsilon_{dt} = 0.86 \pm 0.03$ .

To find the quantities  $\lambda_{dt}^0$  we used the "preserved-muon method" proposed by us previously.<sup>7</sup> We selected events in which on a single anode in addition to the ST signal there was recorded as a minimum one additional signal which appeared at a time  $T_0 \gg 2\mu\text{sec}$  after appearance of the ST signal. It is clear that in this case the muon had survived during the entire time interval  $[0, T_0]$ , i.e., it did not decay, was not captured by atoms of impurities or by products of nuclear fusion, and also did not leave the zone of this anode. For events selected in this way the probability of observing a signal in an interval  $[T_1, T_2]$  located inside the interval  $[0, T_0]$  is related in the following way to the quantity  $\lambda_{dt}$ :

$$P = \lambda^* \int_{T_1}^{T_2} \exp(-\lambda^* t) dt = 1 - \exp(-\lambda^* T), \quad (3)$$

where

$$\lambda^* = [(1 - \varepsilon_{dt})(Y + A_t)\varepsilon_{dt}^* + \varepsilon_{dt}](1 - \omega_{dt})\lambda_{dt} + [(1 - \varepsilon_{dd})(Y + A_t)\varepsilon_{dd}^* + \varepsilon_{dd}](1 - \omega_{dd})\lambda_{dd\mu}. \quad (4)$$

Here

$$\lambda_{dt} = \lambda_{dt}^0 \varphi c_t, \quad \lambda_{dd\mu} = \lambda_{dd\mu}^0 \varphi c_d, \quad T = T_2 - T_1,$$

$c_t$  and  $c_d$  are the relative concentrations of tritium and deuterium,  $\omega_{dt}$  and  $\omega_{dd}$  are the known coefficients of attachment of the muon respectively to  ${}^4\text{He}$  and  ${}^3\text{He}$ ,<sup>7,8</sup>  $A_t = 1.24\%$  is the relative probability of direct landing of the muon on tritium,  $\lambda_{dd\mu}$  is the rate of production of a  $dd\mu$  molecule, and  $\varepsilon_{dt}^*$  is the probability of recording  $dt$  fusion catalysed by a

TABLE I. Experimental and theoretical numbers of pileups of signals with  $E > 3.1$  MeV.

Number of fusion*	Number of pileups	Relative number of pileups, %
1	99	10.7±1.0
2	97	10.5±1.0
3	94	10.1±1.0
1+2+3	290	10.4±0.6
Theory**	—	10.5±1.0

\*Events of the first and second fusions have been selected with the condition of appearance of a third fusion.

\*\*In the calculation we have also taken into account direct sitting down of a muon on tritium. Its contribution to the number of pileups is about 1%.

muon which had produced a  $t\mu$  atom by the channel (2) or in the process of direct landing ( $\varepsilon_{dt}^* \gg \varepsilon_{dt}$  as a consequence of the effect of signal pileup). Equations (3) and (4) are valid in the case  $\lambda_{dt} \ll \lambda_{dt\mu}$ . In our experiment this condition is satisfied:

$$\lambda_{dt}/\lambda_{dt\mu} = \lambda_{dt}^0 c_t / \lambda_{dt\mu}^0 c_d \approx 0.01.$$

In this system we have used the value  $\lambda_{dt\mu}^0 = 3.3 \cdot 10^8 \text{ sec}^{-1}$  from Ref. 6. This large value of  $\lambda_{dt\mu}$  leads to the result that production of a  $dt\mu$  molecule (and consequently  $dt$  fusion) occurs in a very short time interval  $\approx 30$  nsec after formation of the  $t\mu$  atom. It is important that this time is significantly less than the duration of the signal which appears in the ionization chamber ( $t_{\text{sig}} = 100\text{--}300$  nsec). Therefore, if after a recorded event of  $dt$  or  $dd$  fusion the liberated muon rapidly forms a  $t\mu$  atom by the channel (2) or as the result of direct deposition, then as a consequence of the pileup of signals a signal of doubled height will be recorded. Therefore the number of observed pileups can be used to find the value of  $Y$  and consequently of  $q_{1s}$ . However, it is necessary here to take into account trivial pileups of signals as the result of subsequent  $dt$  fusion by channel (1) or  $dd$  fusion. Such pileups have been incorporated into the Monte Carlo calculation whose results are shown in Fig. 2. It is evident that the calculation satisfactorily describes the experimental data, and no additional pileups are observed which could be assigned to the manifestation of channel (2). The stability of the data obtained is demonstrated by the table, in which we have shown the results of analysis of the number of pileups specifically in the first, second, and third fusion signals arising in events in which at least three fusion signals are recorded. A quantitative estimate gives  $Y \leq 3\%$  ( $q_{1s} \geq 97\%$ ). This value of  $Y$  is substantially less than the theoretical value predicted for the conditions of our experiment,  $Y_{\text{theor}} = 14\%$  (Refs. 2 and 3) ( $q_{1s} = 86\%$ ). The dashed line in Fig. 2 corresponds to the theoretical value.

To check the correctness of calculation of the number of pileups we made control measurements. We determined the number of pileups of signals of a test generator on signals of  $\alpha$  particles from an  $\text{Am}^{241}$  source deposited on the cathode of the ionization chamber. The measured number of pileups agreed with high accuracy with the calculated value.

The foregoing estimate permits us to set  $Y = 0$  in Eq. (4), which leads to an error in determination of  $\lambda_{dt}$  of no more than 2%. The remaining quantities entering into Eq. (4) are known with high accuracy. To find  $\lambda^*$  we transform Eq. (3):

$$\exp(-\lambda^* T) = 1 - P = N_- / N_{\Sigma},$$

from which

$$\lambda^* = T^{-1} \ln(N_{\Sigma} / N_-). \quad (5)$$

Here  $N_{\Sigma} = 530$  is the total number of events selected in the preserved-muon method;  $N_- = 352$  is the number of events of the total number  $N_{\Sigma}$  in which in the interval  $[T_1, T_2] = 0.3\text{--}1.6 \mu\text{sec}$  no signals were recorded.

Using the experimentally determined numbers  $N_{\Sigma}$  and  $N_-$ , we were able by means of Eqs. (4) and (5) to obtain the rate of charge exchange  $(2.79 \pm 0.25) \cdot 10^8 \text{ sec}^{-1}$ . The charge exchange rate  $\lambda_{dt}^0$  was determined also by another means from the ratio of the yields of successive fusion events:

$$N_{Fi} / N_{Fi-1} = \lambda^* k_M / \Sigma \lambda, \quad \Sigma \lambda = \lambda_0 + \lambda_{dz} + \omega_{dd} \lambda_{d\mu} + \omega_{dt} \lambda_{dt} + \lambda^*,$$

where  $\lambda_0$  is the muon decay rate,  $\lambda_{dz}$  is the rate of recapture of the muon by an impurity with  $z > 1$ , and  $k_M$  is a correction taking into account the loss of successive fusion events as the result of the finite time of their detection. The quantity  $\Sigma \lambda$  was determined from analysis of the time spectra. The correction  $k_M$  was determined on the basis of experimentally measured distributions in the duration of the signals. By this method we obtained a value  $\lambda_{dt}^0 = (2.81 \pm 0.21) \cdot 10^8 \text{ sec}^{-1}$ . The final averaged result, taking into account systematic errors, was taken as  $\lambda_{dt}^0 = (2.8 \pm 0.2) \cdot 10^8 \text{ sec}^{-1}$ .

This result agrees satisfactorily with the results of the two preceding experiments—Ref. 4 and Refs. 5 and 6. Here we can mention that the experimental value of  $\lambda_{dt}^0$  exceeds the theoretical value by 1.4 times. On the other hand, the contribution of charge exchange from excited states of  $d\mu$  atoms turned out to be substantially smaller than the calculated value. This problem requires further study. It should be mentioned that under the conditions of the present experiment the main contribution to the calculated value of  $Y$  is from charge exchange from the  $2S$  state of the  $d\mu$  atom. It is possible that the calculation of this transition is not sufficiently accurate.

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