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SEARCH FOR THE RADIATIVE CAPTURE $d+d \to {}^4\mathrm{He}+\gamma$ REACTION FROM THE $dd\mu$ MUONIC MOLECULE STATE

Submitted to «Ядерная физика»

L. N. Bogdanova[1], V. R. Bom[2], D. L. Demin, C. W. E. van Eijk[2], V. V. Filchenkov, N. N. Grafov, V. G. Grebinnik, K. I. Gritsaj, A. D. Konin, A. V. Kuryakin[3], V. A. Nazarov[3], V. V. Perevozchikov[3], A. I. Rudenko, S. M. Sadetsky[4], Yu. I. Vinogradov[3], A. A. Yukhimchuk[3], S. A. Yukhimchuk, V. G. Zinov, S. V. Zlatoustovskii[3]

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[1] State Scientific Center of Russian Federation, Institute of Theoretical and Experimental Physics, Moscow, Russia

[2] Delft University of Technology, 2629 JB Delft, the Netherlands

[3] Russian Federal Nuclear Center, All-Russian Research Institute of Experimental Physics, Sarov, Nizhny Novgorod Region, Russia

[4] St. Petersburg Nuclear Physics Institute, Gatchina, Russia

1 Introduction

It is understood that investigations of the fusion reactions between hydrogen isotope nuclei at low energies are of great importance for determining properties of the lightest nuclei and for astrophysics. In particular, there is a need for new or improved measurements of many radiation capture reactions included in various astrophysical scenarios. Due to the Coulomb repulsion fusion cross-sections $\sigma(E)$ drop rapidly at low ($E \leq 100keV$) collision energies (in an exponential scale for "bare" nuclei).

The reaction

$$d + d \longrightarrow^4 He + \gamma + 23.8\, MeV \tag{1}$$

is involved in both primordial and stellar nucleosynthesis. Its cross section is rather small (about 1 pb at 50 keV, compared to 1 mb for the main fusion channels $d(d,n)^3He$ and $d(d,p)^3H$) and its experimental investigations in dd collisions are rather difficult.

At energies $E > 400$ keV reaction (1) proceeds mainly by a d-wave $E2$ transition to the $^1S_0$-state of ^{4}He [1]. The reason is the identical boson character of the entrance channel requiring $L+S$ to be even (L and S are the orbital angular momentum and the total spin of the dd system). At lower energies, the centrifugal barrier suppresses the d-wave $E2$ capture, allowing an s-wave $E2$ transition to the D-state admixture of ^{4}He. Measurements extended to energies below 100 keV [2] have confirmed this picture. However, existence of multipoles other than $E2$ in reaction (1) was not excluded experimentally despite the belief that the dipole transitions $E1$ and $M1$ with $\Delta S = 0$ should be suppressed due to the standard isospin selection rule $\Delta T = 0$.

Measurements of the cross section angular distributions $\sigma(\theta)$, of the vector A_y and tensor A_{yy} analyzing powers performed with a polarized deuteron beam with energy $E_d(lab) = 80\ keV$ stopping in the target have yielded an unexpected observation of the p-wave strength in $^2H(\vec{d},\gamma)^4He$ reaction [3]. It was found that over 50% of the cross section strength at those low energies were due to $E1$ and $M2$ p-wave capture. This finding might affect the low-energy behavior of the S-function and be considered as an isospin selection rule violation.(Other evidence for non-$E2$ radiation can be found in [3].) It would be extremely interesting to observe manifestation of this p-wave in an independent measurement.

During the past decades experiments in which various fusion reactions between hydrogen isotopes are catalyzed by muons have provided supplementary information about these reactions at energies well below the lowest energies accessible by conventional beam-target experiments [4]. In the muon catalyzed (MC) process fusion takes place from the bound states of muonic molecules. Nuclei inside muonic molecules are practically at rest, being separated by average distances $a_\mu \sim \hbar^2/e^2 m_\mu^2 = 2.5 \cdot 10^{-11}\, cm$ (m_μ is the muon mass).

Muonic molecules can be formed in the states with the total angular momenta $J = 0$ and $J = 1$ that correspond to the relative orbital angular momenta of nuclei $L = 0$ and $L = 1$. Depending on the hydrogen isotope mixture parameters various states of muonic molecules can be populated. This makes it possible to study fusion reactions at super-low energies from prepared $s-$ and $p-$ nuclear states with definite spins.

Study of the MC fusion process in a $dd\mu$ muonic molecule resulted in the complementary and detailed information about charge-symmetric reactions $d(d,n)^3He$ and $d(d,p)^3H$. A significant difference in the p-wave parts of the $d(d,p)^3H$ and $d(d,n)^3He$ reaction yields was observed in the experiments with low energy polarized deuteron beams [5]. Comparison of two reaction branches showed some s-wave enhancement together with essential p-wave suppression of the proton branch. (This result was then interpreted by some as evidence for charge symmetry breaking forces.)

Direct measurement of the yield ratio $R_p(n/p)$ for the reactions

$$dd\mu \longrightarrow {}^3He + n + \mu,\ \mu^3He + n, \tag{2}$$

$$dd\mu \longrightarrow t + p + \mu \tag{3}$$

proceeding from the $J = 1$ state of $dd\mu$ molecules [6, 7] gave the value $R_p(n/p) = 1.42 \pm 0.03$. It agreed with the ratio from [5] determined from the elaborate (and model dependent) analysis of the in-flight data. Rates of $dd\mu$ fusion reactions (2),(3) from the p-wave were experimentally measured [8] and the corresponding nuclear reaction constants were extracted from the MC data [9].

The deuteron radiative capture reaction in the $dd\mu$-molecule

$$dd\mu \longrightarrow {}^4He\mu + \gamma + 23.8\,MeV, \tag{4}$$

was not previously investigated because of the extreme smallness of its expected yield. In the systematic study of the MC process in deuterium we have recently performed [10] measurements in the temperature range $T = 85 - 790\ K$. As in our earlier experiments [11], neutrons from reaction (2) were detected. At temperatures $T > 150\ K$ $dd\mu$ molecules are mainly formed in the $J = 1$ state and fusion reactions proceed from the p-wave of relative nuclear motion. Hence, if detected, $23.8 - MeV$ γ-quanta would unambiguously indicate a finite p-wave contribution to the rate of process (4).

In view of this, we investigated the possibility of detecting of process (4) in our last measurements of the $dd\mu$ molecule formation rate [10]. For this aim one of two usually used neutron detectors [11] was removed and a gamma detector was installed instead. The level of the radiation background in our installation was measured. We present the first experimental estimation for the yield of the radiative deuteron capture in the p-wave from the $dd\mu$ molecule.

2 Experimental method

The experimental setup (its lay-out is shown in Fig. 1) is described in detail in [12]. A high pressure deuterium target (HPDT) [13] was exposed on the muon beam of the JINR phasotron.

Scintillation counters 1-3 in front of the HPDT detected the incoming muons. Cylinder-shaped multiwire proportional chambers $4+5$ served to identify the muon stop in the target and to detect electrons from muon decay. A coincidence between signals of the counters 5 and $1e, 2e$ served as the electron signal.

A full absorption neutron detector ND (volume of NE-213 $v = 12.5\,l$) [12, 14] was aimed to detect neutrons from reaction (2). To reduce the background, $n-\gamma$ separation was realized by comparing the signals for the total charge and the fast component charge of the ND pulse. The efficiency of the γ-quantum discrimination was better than 10^{-3} for energies $E_{\gamma,e} > 100\,keV$.

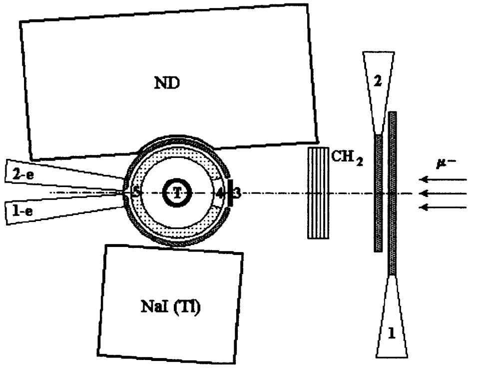

Figure 1. Experimental lay-out.

The γ-quanta were detected with a $NaI(Tl)$ crystal 150 mm in diameter and 100 mm in height. It was calibrated with the γ-sources of ^{60}Co (total energy of two γs 2.5 MeV), *Pu-Be* ($E_\gamma = 4.43$ MeV) and with $5.5-MeV$ γs from the reaction

$$pd\mu \to ^3He\mu + \gamma. \qquad (5)$$

Reaction (5) was observed in the test exposures when the target was filled with H/D mixture containing about 20% of protium. The amplitude distribution for $5.5-MeV$ γs is shown in Fig. 2. The obtained energy resolution of the detector is 15% FWHM.

The calibrating line for the γ-detector is presented in Fig. 3. Linearity of the energy scale was checked under different voltages supplied to the detector in the measurements with the available ^{60}Co and *Pu-Be* γ-sources. In the amplitude region used it proved to be linear at the level of 2-3%. The expected position for γs from (4) is then approximately the 200th channel. Stability of the spectrometric system was controlled with γ-sources during the run.

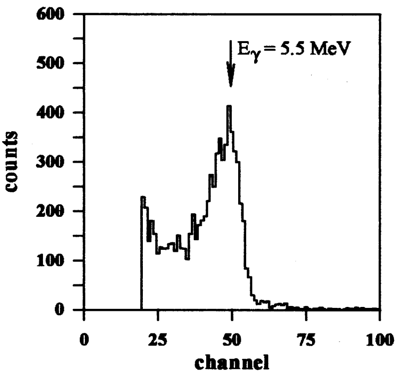

Figure 2. Amplitude distribution of the events detected by the $NaI(Tl)$ detector in the exposures with the H/D mixture.

The detection efficiency for γ-quanta was estimated from their cross sections in NaI and the known solid angle of the detector. With the efficiency losses (30–40 %) due to the bremsstrahlung in the target walls taken into account, the detection efficiency for $24 - MeV$ γ-quanta was found to be

$$\epsilon_\gamma = (5 \pm 1)\% \tag{6}$$

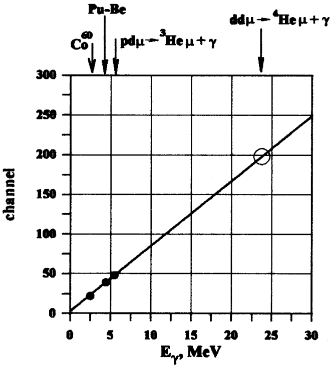

Figure 3. Calibrating line of the γ-spectrometer. Points correspond to the calibration with the ^{60}Co and $Pu - Be$ γ-sources and with γs from reaction (6).

Primary selection of the events detected by the neutron and γ-detectors was realized by the trigger. It allowed only those events for further time and amplitude analysis which were connected with electron registration, that is, delayed $\mu - n, \gamma - e$ coincidences were used. Under this condition the timing sequence of the NaI and ND signals was registered by flash ADC and recorded on the PC. An example of the "oscillogram" of thus stored events is shown in Fig. 4.

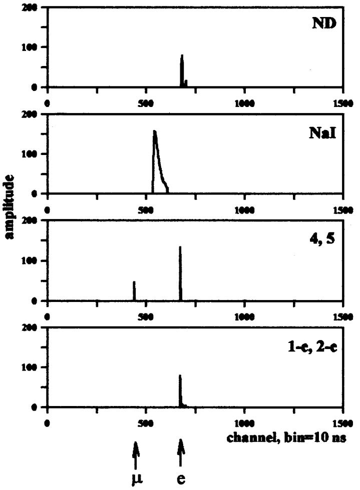

Figure 4. Signals on flash ADC. The muon is detected by the counter 4. The electron from μ-decay is detected by the counters 5, e and, for the presented event, by the *ND* too.

3 Measurements and analysis of *ND* data

During the run eight exposures were performed at different deuterium temperatures and densities. Experimental conditions for them are presented in Table 1. Deuterium density is given in relative units: $\phi = n/n_0$, where $n_0 = 4.25 \cdot 10^{22}\, nucl/cm^3$ is the liquid hydrogen density (LHD). For all exposures the intensity of muons detected by the counters 1-4 was $2.5 \cdot 10^3\, s^{-1}$. The electron counting rate was 15-30 per second depending on the deuterium density.

Table 1. Experimental conditions.

Run	T, K	Content, %		ϕ,	N_e	N_n	$N_{dd\mu}$
		H	D	LHD			
1	85 (5)	20.7 (0.5)	79.3 (0.5)	0.84 (0.03)	712 300	4 000	$1.2 \cdot 10^5$
2	110 (5)	20.7 (0.5)	79.3 (0.5)	0.84 (0.03)	474 600	4 700	$1.2 \cdot 10^5$
3	230 (5)	20.7 (0.5)	79.3 (0.5)	0.83 (0.03)	433 200	15 000	$4.5 \cdot 10^5$
4	301 (4)	20.7 (0.5)	79.3 (0.5)	0.83 (0.03)	443 700	20 200	$6.1 \cdot 10^5$
5	299 (4)	20.7 (0.5)	79.3 (0.5)	0.47 (0.02)	388 900	13 900	$4.2 \cdot 10^5$
6	298 (4)	0.1 (0.1)	99.9 (0.1)	0.50 (0.02)	232 500	18 100	$5.7 \cdot 10^5$
7	548 (10)	0.1 (0.1)	99.9 (0.1)	0.50 (0.02)	240 000	19 500	$5.1 \cdot 10^5$
8	791 (15)	0.1 (0.1)	99.9 (0.1)	0.49 (0.02)	315 000	20 500	$6.1 \cdot 10^5$

The number of detected electrons from muon decay N_e was determined from the fit of the electron time spectra taking into account the background from muon stops in the target walls. The latter was found in the exposure with the empty target. The number of electrons detected for 10 hours of phasotron operation (one exposure) was $\simeq (0.5 - 1.0) \cdot 10^6$ depending of gas density. Details of the analysis can be found in [11].

The number of neutrons from reaction (2) N_n was obtained from the analysis of the time spectra of the events detected by ND and belonging to the neutron region in the $n - \gamma$ plot [14]. The neutron background was measured in a special exposure with the empty target. The determined numbers N_e and N_n are presented in Table 1.

As is seen from Table 1, part of the exposures were made with H/D mixture. It allows detection of reaction (5) which was used both for energy calibration and for checking the γ-quantum detection efficiency.

The kinetics scheme of the main processes occurring after the muon stop in hydrogen-deuterium (H/D) mixture is shown in Fig. 5. As expected, exposures 1 and 2 are characterized by a low neutron/electron ratio. In other words, only a small fraction of the muon stops in the target lead to formation of $dd\mu$-molecules and subsequent reaction (2), detected in our experiment. It is due to the fact [6, 7, 8, 11] that at low temperatures $T < 150\ K$ the $dd\mu$ formation rate $\lambda_{dd\mu} \cdot \phi < 0.1 \cdot 10^6\, s^{-1}$ is small compared to the $d\mu$ atom disappearance rate $\lambda_{d\mu} = \lambda_0 + \lambda_{dd\mu} \cdot \phi \cdot (1 - C_p) + \lambda_{pd\mu} \cdot \phi \cdot C_p$, where $\lambda_0 = 4.55 \cdot 10^5\, s^{-1}$ is the free muon disappearance rate and $\lambda_{pd\mu} = (5.53 \pm 0.16) \cdot 10^6\, s^{-1}$ [15] is the $pd\mu$ formation rate. This allowed the use of these exposures for estimating of the accidental background. Exposures 3-8 were accepted for the search of γs from reaction (4) and estimation of its relative yield.

From the measured numbers of neutrons N_n, the known neutron detection efficiency $\epsilon_n = 13\,\%$ [16] and the partial probability of reaction (2) $\beta \cong 0.7$ [7] the $dd\mu$ formation rates $\lambda_{dd\mu}$ were obtained for each exposure.

The average number of catalysis cycles n_c per muon corresponding to the kinetics scheme in Fig. 5 was calculated with the experimentally determined values $\lambda_{dd\mu}$:

$$n_c = \lambda_{dd\mu} \cdot \phi \cdot (1 - C_p)/[\lambda_0 + \lambda_{dd\mu} \cdot \phi \cdot (1 - C_p) \cdot \omega_{dd} + \lambda_{pd\mu} \cdot \phi \cdot (1 - C_p) \cdot \omega_{pd}], \quad (7)$$

where $\omega_{dd} = 0.07$ [7] and $\omega_{pd} = 0.75$ [17] are the probabilities of muon sticking to helium in reactions (2) and (5) respectively. Using thus determined n_c and measured numbers of electrons N_e we could calculate numbers of $dd\mu$ molecules $N_{dd\mu}$, formed in each exposure, as $N_{dd\mu} = N_e \cdot n_c$. The results are presented in Table 1 and the total number of $dd\mu$ molecules for exposures 3-8 was used for the estimation of the reaction (4) yield.

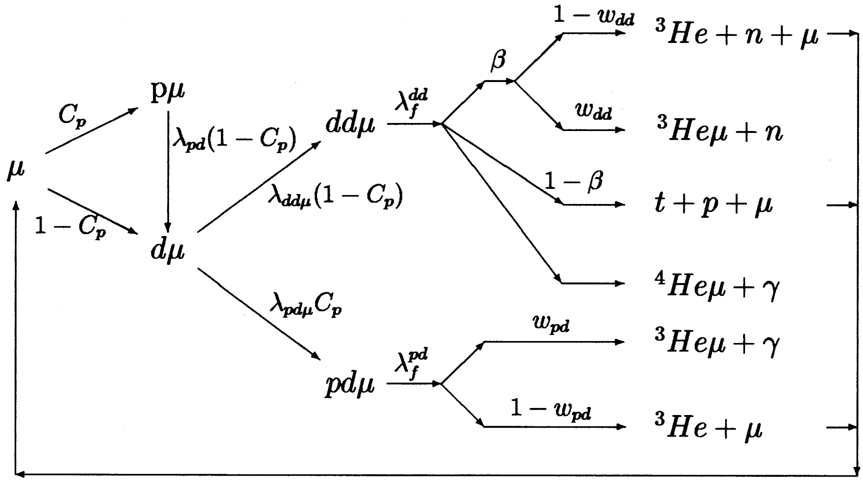

Figure 5. Scheme of the MC fusion cycle in H/D mixture.

4 Analysis of γ-events

Of all events detected by the γ-detector those with the γ energy

$$E_\gamma > 17\,MeV \tag{8}$$

were selected for the further analysis. These events accumulated in exposures 3-8 were displayed in a two-dimensional γ time (t_γ) - electron time (t_e) plot shown in Fig. 6.

Fusion events from reaction (4) should arrive after the muon entrance (t_μ) and before the muon decay (t_e), so for the primary selection the required time sequence is t_e, $t_\gamma > t_\mu$, $t_e > t_\gamma$ (dashed area in Fig. 6).

It is seen that a noticeable fraction of events in this plot is concentrated at small $t_e - t_\mu$, $t_\gamma - t_\mu$. These events might be a manifestation of the muon stops in the target walls. In their material (Ni, Fe) the muon disappears after the average time $\tau_\mu = 0.2\,\mu s$, either due to its decay starting the false trigger, or due to the nuclear capture with emission of capture products. To reduce the background originating from such processes, events corresponding to fast γ and electron emission should be excluded from consideration by introducing a time delay with respect to t_μ.

On the other side, the time distribution of events resulting from the $dd\mu$ molecule fusion (4) should obey the exponential law

$$f_\gamma(t) = Const \cdot exp([-\lambda_0 + \phi\lambda_{dd\mu}\omega_{dd}]t),$$

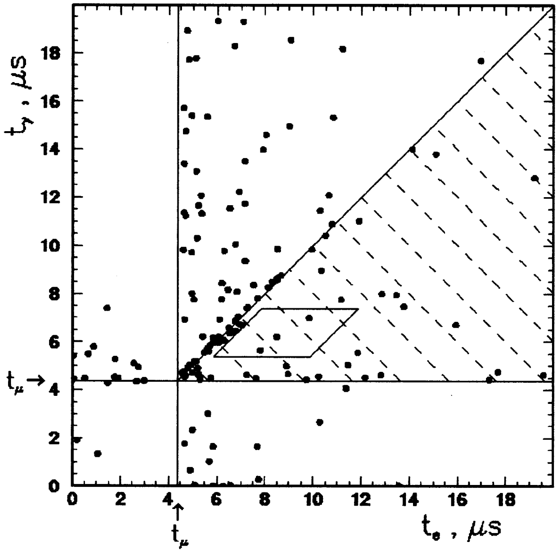

Figure 6. Two-dimensional $t_e - t_\gamma$ plot for the events with $E_\gamma > 17$ MeV detected by the NaI detector in exposures 3-8.

so allowing a large time delay would lead to the loss of the efficiency.

To suppress the accidental background and simultaneously avoid the efficiency losses, the following time intervals were chosen:

$$1\,\mu s < t_\gamma - t_\mu < 3\,\mu s; \qquad 0.5\mu s < t_\gamma - t_e < 4.5\,\mu s. \tag{9}$$

The corresponding region is indicated by the BOX in Fig. 6 with 3 N_γ^t events belonging to it.

To estimate the background, we selected the area $t_\gamma > 1\,\mu s$, $0.5\,\mu s < t_e < 4.5\,\mu s$ and found 7 events there. This corresponds to the number of background events in the region (9) $N_\gamma^{b1} = 2 \pm 1$.

In addition, for independent estimation of the accidental background, events from exposures 1,2 satisfying (8) and (9) were selected. The number of such events normalized to the number of electrons in exposures 3-8 was found to be $N_\gamma^{b2} = 2$. It proved to be at the level of the previous estimate obtained from exposures 3-8.

The background level is found to exceed the measured intrinsic background of the installation, corresponding to the cosmic ray intensity $(0.05/(MeV \cdot s))$ by a factor of 2. We conclude that additional background is correlated with phasotron operation.

The energy distribution of the events detected by the NaI detector and selected with criteria (9) for exposures 3-8 (solid line) is shown in Fig. 7. The dashed line is the spectrum for the normalized background.

It is seen from the figure that these spectra practically coincide for energies $E_\gamma > 17$ MeV. Some excess of events for lower energies can be ascribed to the background induced by neutrons from reaction (2).

From the above considerations the number of candidate events can be obtained

$$N_\gamma = N_\gamma^t - N_\gamma^b = 1 \pm 2 \tag{10}$$

The measured yield of reaction (4) per $dd\mu$-molecule is evaluated as

$$\eta_\gamma = \frac{N_\gamma}{\epsilon_\gamma \cdot N_{dd\mu}^{tot}}, \tag{11}$$

where $N_{dd\mu}^{tot}$ is the total number of $N_{dd\mu}$ molecules accumulated in exposures 3-8:

$$N_{dd\mu}^{tot} = 3.4 \cdot 10^6 \tag{12}$$

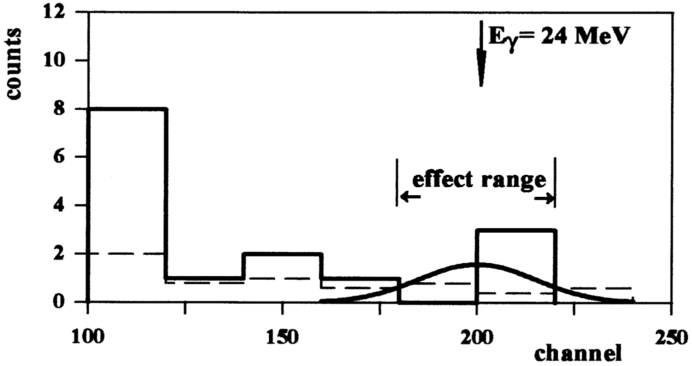

Figure 7. Amplitude γ-quantum spectra for the events selected with criteria (8) for exposures 3-8 (solid line) and the normalized "background" spectra (dashed line). The response function of the NaI detector is represented by the gaussian.

Using the estimate (6) for the efficiency of γ-quantum registration and taking into account the selection efficiency due to the accepted criteria (9) one obtains the detection efficiency of γs from reaction (4)

$$\epsilon_\gamma = (3 \pm 0.5)\,\% \tag{13}$$

Substituting (10), (12) and (13) into Eq. (11) we obtain

$$\eta 1_\gamma = (1 \pm 2) \cdot 10^{-5}, \tag{14}$$

for the absolute γ yield per $dd\mu$-molecule.

After this work was reported and the article was prepared the new measurements with a deuterium target and a $NaI(Tl)$ detector of larger size were conducted and a similar analysis was performed. The result is

$$\eta 2_\gamma = (0.8 \pm 1.5) \cdot 10^{-5} \tag{15}$$

Combining (14) and (15) one obtains

$$\eta_\gamma < 2 \cdot 10^{-5}. \tag{16}$$

at the 90% C.L.

From here an upper limit for the radiative fusion rate λ^1_γ from the $J = 1$ state of the $dd\mu$ molecule can be deduced using the experimental value of the total fusion rate $\lambda^1_f = 4 \cdot 10^8\, s^{-1}$ [9]

$$\lambda^1_\gamma < 8 \cdot 10^3\, s^{-1}.$$

5 Conclusion

The first attempt has been made to estimate the yield of radiative capture reaction (4) from the $J = 1$ state of the $dd\mu$ muonic molecule. The background conditions were evaluated and the appropriate methods of data analysis were elaborated. The sensitivity of the present experiment is not enough to make a decisive conclusion about the p-wave contribution to the process of radiative dd capture. (The expected level estimated from the data [3] is $\eta_\gamma \sim 10^{-6}$.) The improvement of our result by one-two orders of magnitude will be possible with a new γ-detectors of larger efficiency and more intense muon beams. Of crucial importance is the understanding of the background structure and elaboration of background suppression methods.

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Богданова Л. Н. и др. E15-2001-264

Поиск реакции радиационного захвата $d+d \to {}^4\mathrm{He}+\gamma$ из состояния $dd\mu$ мюонной молекулы

Проведен эксперимент по поиску реакции мюонного катализа $dd \to {}^4\mathrm{He}+\gamma$ из состояния $dd\mu$ мюонной молекулы на установке ТРИТОН с использованием детекторов γ-квантов NaI(Tl). На пучке отрицательных мюонов фазотрона ОИЯИ экспонировалась мишень высокого давления, заполненная дейтерием, с целью регистрации γ-квантов с энергией 23,8 МэВ. Получена первая экспериментальная оценка выхода реакции радиационного захвата дейтрона из $J=1$ $dd\mu$-состояния на уровне $n_\gamma \le 2\cdot10^{-5}$ на одно событие синтеза.

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Search for the Radiative Capture $d+d \to {}^4\mathrm{He}+\gamma$ Reaction from the $dd\mu$ Muonic Molecule State

A search for the muon catalyzed fusion (MCF) reaction $dd \to {}^4\mathrm{He}+\gamma$ in the $dd\mu$ muonic molecule was performed using the experimental MCF installation TRITON and NaI(Tl) detectors for γ-quanta. The high pressure target filled with deuterium was exposed to the negative muon beam of the JINR phasotron to detect γ-quanta with energy 23.8 MeV. The first experimental estimation for the yield of the radiative deuteron capture from the $dd\mu$ state $J=1$ was obtained at the level $n_\gamma \le 2\cdot10^{-5}$ per fusion.

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

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