Search for the radiative capture $d + d \rightarrow ^4 He + \gamma$ reaction from the $dd\mu$ muonic molecule state.

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$^3$

Joint Institute for Nuclear Research (JINR), Dzhelepov Laboratory of Nuclear Problems, Dubna, 141980 Russia
1— State Scientific Center of Russian Federation, Institute of Theoretical and Experimental Physics, Moscow, 117218
2— Delft University of Technology, 2629 JB Delft, the Netherlands
3— Russian Federal Nuclear Center, All-Russian Research Institute of Experimental Physics (RFNC-VNIIEF), Sarov, Nizhny Novgorod reg., 607200
4— St. Petersburg Nuclear Physics Institute (PNPI), Gatchina, 188350

Abstract

A search for the muon catalyzed fusion reaction $dd \rightarrow ^4 He + \gamma$ in the $dd\mu$ muonic molecule was performed using the experimental $\mu CF$ installation TRITON and $NaI(Tl)$ detectors for $\gamma$-quanta. The high pressure target filled with deuterium at temperatures from 85 K to 800 K was exposed to the negative muon beam of the JINR phasotron to detect $\gamma$-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 $n_{\gamma} \leq 2 \times 10^{-5}$ per one fusion.

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 lightest nuclei and for the 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). Reaction

$$d + d \rightarrow ^4 He + \gamma + 23.8 \text{ MeV}$$ (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 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 orbital angular momentum and total spin of the \(dd\) system). At lower energies, the centrifugal barrier suppresses the d-wave E2 capture, allowing 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, an existence of multipoles other than E2 in reaction (1) was not excluded experimentally despite belief that dipole transitions \(E1\) and \(M1\) with \(\Delta S = 0\) should be suppressed due to standard isospin selection rule \(\Delta T = 0\).

Measurements of the cross section angular distributions \(\sigma(\theta)\), of vector \(A_y\) and tensor \(A_{yy}\) analyzing powers performed with a polarized deuteron beam with energy \(E_d(\text{lab}) = 80\) keV stopping in the target have yielded an unexpected observation of the p-wave strength in \(^2\)H\((d, \gamma)^4\)He reaction \[3\]. It has been found that over 50% of the cross section strength at these low energies is 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. (Some other evidences for non-E2 radiation are cited in \[3\]). It would be extremely interesting to observe this p-wave manifestation in an independent measurement.

During 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 \(a_\mu \sim \hbar^2/e^2m_\mu^2 = 2.5 \times 10^{-11}\) cm (\(m_\mu\) is the muon mass).

Muonic molecules can be formed in the states with 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 possible study fusion reactions at super-low energies from prepared s- and p- nuclear states with definite spins.

Study of the MC fusion process in \(dd\mu\) muonic molecule resulted in the complimentary and detailed information about charge-symmetric reactions \(d(d, n)^3\)He and \(d(d, p)^3\)H. A significant difference in the p-wave parts of the \(d(d, p)^3\)H and \(d(d, n)^3\)He reaction yields was observed in the experiments with low energy polarized deuteron beams \[4\]. 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 an evidence for charge symmetry breaking forces.)

Direct measurement of the yields ratio \(R_p(n/p)\) for reactions

\[
\begin{align*}
dd\mu \longrightarrow ^3He + n + \mu, \quad &\mu^3He + n, \\
dd\mu \longrightarrow t + p + \mu
\end{align*}
\]
proceeding from the \( J = 1 \) state of \( dd\mu \) molecules \(^3\), \(^6\) gave the value \( R_p(n/p) = 1.42 \pm 0.03 \). It agreed with the ratio from \(^4\) 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 corresponding nuclear reaction constants were extracted from MC data \(^9\).

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

\[
dd\mu \rightarrow ^4He\mu + \gamma + 23.8 \, MeV,
\]

has not previously been 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 \( T=85-800 \) 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 \( J=1 \) state and fusion reactions proceed from the p-wave of relative nuclear motion. Hence, being detected, 23.8 MeV \( \gamma \)-quanta would unambiguously indicate a finite p-wave contribution to process (4) rate.

In view of this, we performed a feasibility study of process (4) detection 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 has been 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 setup

The experimental setup (its layout is shown in Fig. 1) has been described in detail in \(^12\). The muon beam of the JINR phasotron was directed into a high pressure deuterium target (HPDT) \(^13\). Scintillation counters 1-4 detected incoming muons. Cylinder-shaped counter 5 served to identify the muon stop in the target and to detect electrons from muon decay. A coincidence between signals of counters 5 and 1e, 2e served as the electron signal.

A large neutron detector ND (volume of NE-213 \( v = 12.5 \, l \)) \(^14\), \(^12\) was aimed to detect neutrons from reaction (2). To reduce a background, the \( n - \gamma \) separation was realized by comparing the signals for the total charge and the fast component charge of the ND pulse. The efficiency of the \( \gamma \)-quanta discrimination was better than \( 10^{-3} \) for energies \( E_{\gamma,e} > 100 \, keV \).
The $\gamma$-quanta were detected with a NaI(Tl) crystal of a 150 mm diameter and a 100 mm height. It was calibrated with $\gamma$-sources $^{60}$Co (total energy of two $\gamma$-s 2.5 MeV), Pu-Be ($E_\gamma = 4.43$ MeV) and with 5.5 MeV $\gamma$-s from the reaction

$$ pd\mu \rightarrow ^3He\mu + \gamma. \quad (5) $$

Reaction (5) was observed in the test exposures when the target was filled with H/D mixture containing about 20% of protium. The energy resolution of the detector was 15% FWHM at 5.5 MeV.

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

We estimated the $\gamma$-quanta detection efficiency using values of $\gamma$-s cross sections in NaI and the known solid angle of the detector. Taking into account efficiency losses (30 – 40%) due to the bremsstrahlung in the target walls we determined the efficiency for 24 MeV $\gamma$-quanta detection

$$ \epsilon_\gamma = (5 \pm 1)\%. \quad (6) $$

Primary selection of the events detected by the neutron and $\gamma$-detectors was realized by the trigger. It allowed only those events for further time and amplitude analysis which were connected with electron registration, i.e., delayed coincidences $\mu - n$, $\gamma - e$ were used. Under this condition the timing sequence of the NaI and ND signals was registered by flashes ADC and recorded on the PC.

### 3 Measurements

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 $n_0 = 4.25 \cdot 10^{22} \text{ nucl/cm}^3$ is the liquid hydrogen density (LHD). For all exposures the intensity of muons detected by counters
1-4 was 2.5 · 10^3 s⁻¹. The electron counting rate was 15-30 per second depending on the deuterium density.

### Table 1. Experimental conditions.

| Exposure | T, K  | Content, % | φ, LHD | N_e     | N_n     | N_{ddµ} |
|----------|-------|------------|--------|---------|---------|---------|
|          |       | C_p(H)     | C_d(D) |         |         |         |
| 1        | 85 (5)| 20.7 (0.5) | 79.3 (0.5) | 0.84 (0.03) | 712 300 | 4 000 | 1.2 · 10^5 |
| 2        | 110 (5)| 20.7 (0.5) | 79.3 (0.5) | 0.84 (0.03) | 474 600 | 4 700 | 1.2 · 10^5 |
| 3        | 230 (5)| 20.7 (0.5) | 79.3 (0.5) | 0.83 (0.03) | 433 200 | 15 000 | 4.5 · 10^5 |
| 4        | 301 (4)| 20.7 (0.5) | 79.3 (0.5) | 0.83 (0.03) | 443 700 | 20 200 | 6.1 · 10^5 |
| 5        | 299 (4)| 20.7 (0.5) | 79.3 (0.5) | 0.47 (0.02) | 388 900 | 13 900 | 4.2 · 10^5 |
| 6        | 298 (4)| 0.1 (0.1)  | 99.9 (0.1) | 0.50 (0.02) | 232 500 | 18 100 | 5.7 · 10^4 |
| 7        | 548 (10)| 0.1 (0.1) | 99.9 (0.1) | 0.50 (0.02) | 240 000 | 19 500 | 5.1 · 10^4 |
| 8        | 791 (15)| 0.1 (0.1) | 99.9 (0.1) | 0.49 (0.02) | 315 000 | 20 500 | 6.1 · 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 one was found in the exposure with the empty target. Number of electrons detected per 10 hours of phasotron operation (one exposure) was ≃ (0.5 − 1.0) · 10^6. 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−γ plot [14]. The neutron background was measured in the special exposure with the target filled with helium. Thus determined numbers of N_e and N_n are presented in Table 1.

Exposures made with H/D mixture allowed detection of reaction (5), which was used both for energy calibrating and checking the γ-quanta detection efficiency.

As expected, exposures 1 and 2 are characterized by a low neutron/electron ratio. In other words, only small fraction of the muon stops in the target lead to formation of ddµ-molecules and subsequent reaction (2), detected in our experiment. It is due to the fact [4], [5], [8], [11] that, at low temperatures T < 150 K, the ddµ formation rate λ_{ddµ} · φ < 0.1 · 10^9 s⁻¹ is small compared to the dµ-atom disappearance rate λ_{dµ} = λ_0 + λ_{ddµ} · φ · (1 − C_p) + λ_{pdm} · φ · C_p, where λ_0 = 4.55 · 10^8 s⁻¹ is the free muon disappearance rate and λ_{pdm} = (5.53±0.16)·10^6 s⁻¹ [15] is the pdµ formation rate. This allowed use these exposures for the estimate of the accidental background. Exposures 3-8 were accepted for the search of γ-s from reaction (4) and estimation of its relative yield.

From the measured numbers of neutrons N_n, known efficiency of neutron detection ε_n = 13% [10] and partial probability of reaction (2) β ≈ 0.58 [3], [4] ddµ formation rates λ_{ddµ} were determined for each exposure. With thus obtained values λ_{ddµ} and data from Table 1 we calculated average number of catalysis cycles, n_c, using a simplified
A formula describing $\mu$CF kinetics in H/D mixture (see, e.g. [17])

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

Here $\varpi_{dd} = 0.07$ and $\varpi_{pd} = 0.85$ are branching ratios of fusion reactions with muon sticking to helium with respect to all fusion channels in muonic molecules $dd\mu$ and $pd\mu$. These reactions lead to the muon loss from the catalysis cycle. 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 reaction (4) yield.

### 4 Analysis of $\gamma$-events

Of all events detected by the $\gamma$-detector those with $\gamma$ energy

$$E_\gamma > 17 \text{ MeV} \quad (8)$$

were selected for the further analysis. These events accumulated in exposures 3-8 were displayed in a two-dimensional plot $\gamma$ time ($t_\gamma$) - electron time ($t_e$), shown in Fig. 2.

![Figure 2. Two-dimensional $t_e - t_\gamma$ plot for the events with $E_\gamma > 17$ MeV detected by NaI detector in exposures 3-8.](image)

Fusion events from reaction (4) should arrive after the muon entrance ($t_\mu$) and before the muon decay ($t_e$), so for the primary selection the following time sequence $t_e, t_\gamma > t_\mu, t_e > t_\gamma$ was required, corresponding to the dashed area in Fig. 2.

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) 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 $\gamma$- and electron emission should be excluded from the consideration by introducing a time delay with respect to $t_{\mu}$.

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

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

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

To provide the suppression of the accidental background and simultaneously avoid the efficiency losses the following time intervals were chosen:

$$1 \mu s < t_\gamma - t_{\mu} < 3 \mu s; \quad 0.5 \mu s < t_\gamma - t_e < 4.5 \mu s.$$  \hspace{1cm} (9)

The corresponding region is indicated by a BOX in Fig. 2 with 3 $N^t_\gamma$ events inside.

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) $N^b_\gamma = 2 \pm 1$.

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

The background is found to exceed the measured intrinsic background of the installation, corresponding to the cosmic ray intensity at sea level, 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. 3. The dashed line is the spectrum for the normalized background.

![Figure 3. Amplitude $\gamma$-quanta spectra for the events selected with criteria (8) for exposures 3-8 (solid line) and the normalized "background" ones (dashed line). Response function of the NaI detector is represented by the gaussian.](image-url)
It is seen from the figure that these spectra practically coincide for energies $E > 17$ MeV. Some excess of events for lower energies can be ascribed to the background induced by neutrons from reaction (2).

From considerations above the number of candidate events can be obtained

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

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

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

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

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

Using the estimation (6) for the efficiency of $\gamma$-quanta registration and taking into account the selection efficiency due to the accepted criteria (9) one obtains the detection efficiency of $\gamma$-s from reaction (4)

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

Substituting values (10), (13) and (14) into Eq. (11) we obtain for the absolute $\gamma$ yield per one $dd\mu$-molecule

$$\eta_{\gamma}^{(1)} = (1 \pm 2) \cdot 10^{-5}.$$ (14)

In the second run new measurements with deuterium target and NaI(Tl) detector of larger size have been conducted and result of the similar analysis of the new data set is in agreement with (14)

$$\eta_{\gamma}^{(2)} = (0.8 \pm 1.5) \cdot 10^{-5}.$$ (15)

Combining (14) and (15) one obtains (at 90% C.L.)

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

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

$$\lambda_{\gamma}^1 < 8 \cdot 10^3$ s$^{-1}$.

5 Conclusion

The first attempt has been undertaken to estimate the yield of radiative capture reaction (4) from the $J=1$ state of $dd\mu$ muonic molecule. The background conditions were evaluated and 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. (With the data from [3] we would expect $\eta_{\gamma} \sim 10^{-6}$..) We plan to achieve this level of sensitivity using new $\gamma$-detectors of larger efficiency. Of crucial importance is the understanding of the background structure and elaboration of background suppression methods.
6 Acknowledgements

This work has been supported by the Department of Atomic Science and Technology of Minatom of Russia under the Contract 6.25.19.19.00.969. The authors thank L.I. Ponomarev for support and V.B. Belyaev, V.E. Markushin and L.N. Strunov for the stimulating discussions.

References

[1] F.J. Wilkinson and F.E. Cecil, Phys.Rev. C 31 (1985) 2036.
[2] C.A. Barnes et al., Phys.Lett. B 197 (1985 )315.
[3] L.H. Kramer et al., Phys.Lett. B 304 (1993 )208.
[4] L.N. Bogdanova, Surveys in High Energy Physics 6 (1992) 177.
[5] B.P. Ad’yasevich, V.G. Antonenko, V.E. Bragin, Yad.Fiz. 33 (1981) 1167.
[6] D.V.Balin et al., Phys.Lett. 141B (1984) 173, JETP Lett. 40 (1984) 112.
[7] D.V. Balin et al., Muon Cat. Fusion 1 (1987) 127.
[8] C. Petitjean et al., Hyp. Int. 118 (1999) 127.
[9] G.M.Hale, Muon Catalyzed Fusion 5/6 (1990) 227.
[10] V.R. Bom et al, Proceedings of $\mu$CF-01, Shimoda, Japan, April 2001.
[11] V.P. Dzhelepov et al., Sov. Phys. JETP 74 (1992) 589.
[12] Yu.P. Averin et al., Hyp. Int. 118 (1999) 111.
[13] V.V. Perevozchikov et al., Proceedings of $\mu$CF-01, Shimoda, Japan, April 2001.
[14] V.P. Dzhelepov et al., Nucl. Instr.& Meth. A 269 (1988) 634.
[15] V.M. Bystritsky et al., Sov. Phys. JETP 43 (1976) 606.
[16] V.R. Bom and V.V. Filchenkov, Hyp. Int. 119 (1999) 365.
    V.V. Filchenkov, L. Marczi, Communications JINR, E13-88-566, Dubna, 1988.
[17] C. Petitjean, K. Lou, P. Ackerbauer, et al., Muon Cat. Fusion, 5/6 (1990/1991) 199.