Resonance in the rate of production of $dd\mu$ mesic molecules in gaseous deuterium

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(Submitted 6 September 1978)
Zh. Eksp. Teor. Fiz. **76**, 460–469 (February 1979)

A deuterium-gas target was used in the muon beam of the 680-MeV synchrocyclotron at the Laboratory for Nuclear Problems of the Joint Institute for Nuclear Research to determine the rate of production of $dd\mu$ mesic molecules ($\lambda_{dd\mu}$) as a function of the energy $\epsilon_{d\mu}$ of the $d\mu$ atom in the range 0.015–0.050 eV (deuterium temperature 120–380°K). Experimental results confirm the theoretically predicted resonance mechanism of production of $dd\mu$ mesic molecules. Analysis of the experimental data shows that the resonance occurs at $\epsilon^0_{d\mu} = 0.050 \pm 0.003$ eV and that the value of $\lambda_{dd\mu}$ at the peak of the curve is $\lambda_{dd\mu}(\epsilon^0_{dd\mu}) = (0.85 \pm 0.11) \times 10^6$ sec^{-1}. This has been used to determine, with high precision, the binding energy of the $dd\mu$ molecule in the state with quantum numbers $L = 1$, $v = 1$, which is responsible for the resonance: $\epsilon_{dd\mu}$ $(L = 1, v = 1) = -2.196 \pm 0.003$ eV.

PACS numbers: 36.10.Dr

The production of the dd mesic molecules has been investigated both experimentally[1-4] and theoretically.[5-10] The first measurements of the rates of production $\lambda_{dd\mu}$ of the $dd\mu$ mesic molecules were reported by Fetkovich *et al.*[1] and Doede,[2] who used liquid deuterium (bubble chambers) in which a determination was made of the yield of the fusion reaction in the $dd\mu$ system:

$$dd\mu \to t + p + \mu. \qquad (1a)$$

In our experiments with the diffusion chamber,[3] we recorded, for the first time, the reaction

$$dd\mu \to \mathrm{He}^3 + n + \mu \qquad (1b)$$

and determined the yields of both (1a) and (1b), which were found to be roughly equal. In the experiments reported in Ref. 4, which were carried out with a deuterium-gas target at 41 atm, we determined the yield and time distribution of neutrons from reaction (1b). Table I summarizes the results of measurements[1-4] and of calculations[5-8] of the quantity $\lambda_{dd\mu}$ up to 1977.

It is clear from Table I that the values of $\lambda_{dd\mu}$, obtained for liquid and gaseous deuterium, differ by roughly an order of magnitude. On the other hand, the values of $\lambda_{dd\mu}$ obtained for gaseous deuterium are in clear disagreement with calculations based on the usual mechanisms of production of muonic molecules through $E1$ and $E0$ transitions with the transfer of the binding energy to the conversion electron. To explain the discrepancy between experimental data for liquid and gaseous deuterium, and between experiments and theory, we have pointed out[3] the possibility of resonance in the dependence of the rate of production of the $dd\mu$ molecules on the deuterium temperature or the mean kinetic energy $\varepsilon_{d\mu}$ of the $d\mu$ atoms forming these molecules.

In 1967, Vesman[8] considered a specific mechanism for the resonance production of $dd\mu$ molecules. In his scheme, the energy released during the production of the $dd\mu$ molecules is expended in exciting vibrations in the compound system consisting of the molecular ion $(dd\mu)^+$, a deuteron, and electrons; but this mechanism operates only when the $dd\mu$ system has a state with a low binding energy ($\varepsilon_{dd\mu} \sim 1$ eV), and the position of the peak on the function $\lambda_{dd\mu}(\varepsilon_{d\mu})$ is very sensitive to this level energy.

Vesman assumed that $dd\mu$ did have a state with low binding energy and, by including only the resonance mechanism, found that the maximum value $\lambda_{dd\mu} \approx 1.5 \times 10^6$ sec^{-1} was reached only for energies $\varepsilon_{d\mu} \approx 0.02$ eV. It is important to note that previous theoretical work[5] provided some evidence for the existence of a weakly bound state of the $dd\mu$ molecule with orbital angular momentum $L = 1$ and vibrational quantum number $v = 1$. This was confirmed by subsequent calculations[9,10] which yielded $\varepsilon_{dd\mu} = -1.96$ eV. More careful calculations[10] of $\lambda_{dd\mu}(\varepsilon_{d\mu})$ took into account both the resonance mechanism and the usual mechanisms of $dd\mu$ production through $E1$ and $E0$ transitions with conversion of the electron in the D_2 molecule. It was shown[10] that for $d\mu$ atom energies in the range $\varepsilon_{d\mu} \gtrsim 0.02$ eV, the resonance mechanism of $dd\mu$ production should dominate in com-

TABLE I. Experimental and calculated rates of production of mesic molecules of deuterium, obtained up to 1977.

$\lambda_{dd\mu} \cdot 10^{-6}$, sec^{-1}	$\varepsilon_{d\mu}$, eV	Source of data
	Experiment	
0.076±0.015	0.0039	[1] liquid D_2
0.103±0.004	0.0039	[2] »
0.75±0.11	0.032	[3] gaseous D_2
0.73±0.07	0.039	[4] »
	Theory	
0.04	He depends on $\varepsilon_{d\mu}$	[5]
0.036	»	[6]
0.042	»	[7]
≈1.5*	≈0.02	[8]

*Maximum value reported by Vesman.[8]

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parison with the nonresonance mechanism, and that a change in the energy of the $d\mu$ atom by a few hundredths of eV should lead to a substantial change in $\lambda_{dd\mu}$.

Experimental verification of the predicted resonance form of $\lambda_{dd\mu}(\varepsilon_{d\mu})$ is of considerable interest both for the elucidation of the mechanisms responsible for mesomolecular processes and for the verification of precision calculations of the binding energy of the three-body system. If one starts with the expected form of this dependence, one can hope to determine the position of its peak and hence the binding energy of the *dd* molecule to within about 10^{-3} eV. We recall that the level energy scale of mu-atomic systems is determined by the quantity $1/2m_\mu\alpha^2 \approx 2500$ eV, whereas the precision of modern calculations[9] of the energies of such levels is about 0.1 eV.

The aim of the present work was to determine $\lambda_{dd\mu}$ for the $d\mu$-atom energy range $\varepsilon_{d\mu} = 0.015$–$0.050$ eV, which corresponded to the temperature range 120–400°K. The experiment was performed in the muon beam of the 680-MeV beam of the synchrocyclotron at the Joint Institute for Nuclear Research. Preliminary results were published in Ref. 11.

The method used to determine $\lambda_{dd\mu}$ was analogous to that employed previously and involved the measurement of the yield and time distribution of neutrons from reaction (1b). The experimental arrangement is illustrated schematically in Fig. 1. The beam of muons with momentum $p = 130$ MeV/sec was extracted through the meson channel into the low-background laboratory containing the experimental setup. The muons were recorded by the scintillation counters 1–3, were slowed down in the copper filter 6, and were eventually intercepted by the deuterium-gas target. The target design is described elsewhere.[12,13] Muons coming to rest in the gas were recorded by scintillators located in the interior of the target. One was in the form of a disk, 110 mm in diameter and 0.3 mm thick (counter 4), and the other was in the form of a cup of diameter 120 mm, length 205 mm, and wall thickness 5 mm (counter 5).

The use of CsI(Tl) scintillators for the detection of muons stopping in the gas target has certain definite advantages. Thus, firstly, muons have a short life-

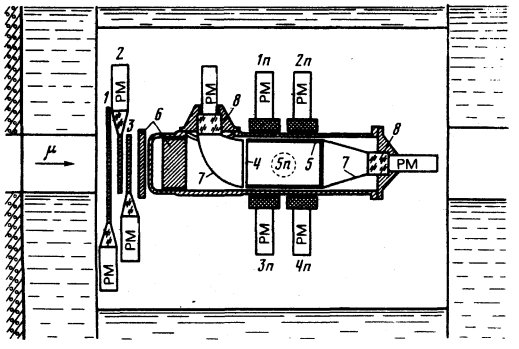

FIG. 1. Schematic illustration of the experimental arrangement: 1–5, $1n$–$5n$—scintillation counters; 6—slowing down medium; 7—hollow light guides; 8—light guides.

time in cesium iodide ($\tau_\mu = 0.09$ μsec), which means that the background due to muons stopping in the scintillator itself can be suppressed. Secondly, cesium iodide has a low saturation vapor pressure, so that a high degree of purity of the deuterium gas can be maintained. However, the properties of this scintillator (conversion efficiency and decay time) are functions of temperature. According to our data, the light yield of the CsI(Tl) scintillator decreases by roughly an order of magnitude when the temperature is varied by ±120° from $T_0 = 20$°C. In our experiments, a large change in the light yield of the scintillators in counters 4 and 5 at low temperatures would produce a substantial reduction in the efficiency of detection of stopping muons and an increase in the number of background stopping events.

The scintillating properties of a plastic phosphor do not change when its temperature is reduced, but the phosphor can only be used at temperatures below −20°C because of the rapid increase in its saturation vapor pressure with increasing temperature. We therefore divided our temperature range into two overlapping intervals: we used a plastic phosphor (experiment I) between −160 and −20°C and the CsI(Tl) phosphor (experiment II) between −60 and +110°C.

The relative atomic amount of impurities with $Z > 1$ in the deuterium used in these experiments was 10^{-7}. The purification and target filling systems were described elsewhere.[12,14] The target was cooled by liquid nitrogen vapor flowing through a coil attached to the body of the target. The degree of cooling could be controlled by varying the rate of flow of the cold nitrogen gas. The target was heated by a nichrome coil. Its temperature was measured by two thermocouples and was also monitored through measurements of the gas pressure in the target.

The target was surrounded with five scintillation counters ($1n$–$5n$) incorporating stilbene crystals, 70 mm in diameter and 30 mm long. These detectors recorded the 2.5-MeV neutrons produced in reaction (1b) and the muon-decay electrons stopping in the target. The neutron detection system is described elsewhere.[13,15]

The logic employed in the experiment is illustrated by the block diagram of Fig. 2. The muon-stopping signal ($234\bar{5}$) triggered the 10-μsec time gate during which signals from counters 1, 5, $1n$–$5n$ were analyzed. The time-to-code converters (t-C) were used to determine the time of appearance of signals from the detectors relative to the muon-stopping time. To separate neutrons from electrons, we used the component separation unit (CSU) and the amplitude encoders (A-C); the former produces two pulses, one of which is proportional to the total area Σ under the light signal from the stilbene phosphor (or the energy of the electron E), whereas the amplitude of the other was proportional to the "fast" component of the light signal.

The amplitude-time data converted into digital code, and the content of the register of the identifying logic, were transmitted to a computer which periodically

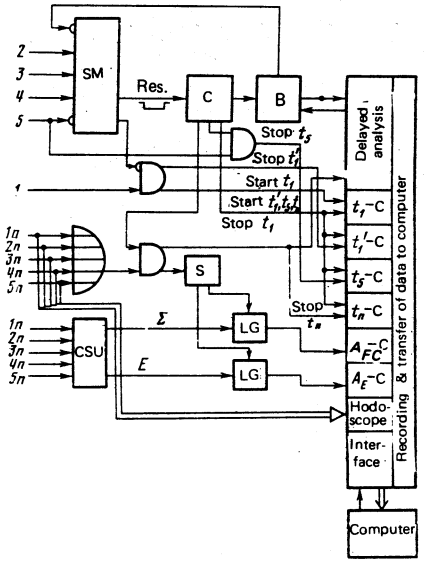

FIG. 2. Block diagram of the logic: SM—stopping muon identification; C—coupler; S—shaper; B—trigger blocking; LG—linear gat; CSU—component separation unit; $t-C$—time-to-code converter; $A-C$—amplitude-to-code converter.

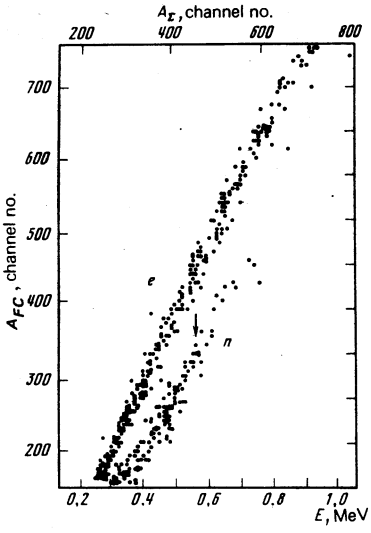

FIG. 3. Distributions on the A_{FC}, A_Σ plane illustrating the separation of neutrons from γ rays and electrons, $T = 180°K$. The channel number for the Σ signal and the light-yield equivalent energy E is plotted along the horizontal axis and the channel number for the FC signal along the vertical axis. The arrow indicates the limiting energy in the spectrum of recoil protons for neutrons from reaction (1b).

received the counting rate from the monitor (23), the stopping counting rate ($234\bar{5}$), and the counting rate from the neutron detectors.

Only those events were selected for analysis that did not involve the detection of a second muon by counter 1 for 5 μsec before and 10 μsec after a stopping muon. The separation of neutrons and electrons was performed by the simultaneous analysis of the amplitudes A_Σ and A_{FC}.[15] The electron and neutron events lie within two nonoverlapping regions on the (A_{FC}, A_Σ) plane. Events due to the detection of monoenergetic neutrons from reaction (1b) should appear on the E axis in the form of an edge at about 0.6 MeV (on the scale equivalent to the light yield corresponding to the electron energy). Figure 3 shows a typical distribution of events on the (A_{FC}, A_Σ) plane, recorded by one of the neutron detectors at $T = 180°K$.

Table II summarizes the data characterizing the exposure of deuterium in experiments I and II. The same amount of deuterium was used in each experiment with the exception of the last exposure in experiment II ($T = 213°K$) in which the gas pressure was increased from 33.5 to 48.5 atm in order to increase the rate of accumulation of the events. The target temperature was held constant to better than 5° within each exposure.

The data on the number of electrons (N_e) and neutrons (N_n) obtained by analyzing the (A_{FC}, A_Σ) distributions are shown in Table II. To avoid electrons in neutron detection, we used the condition that there was no coincidence between the pulse from counter 5 and the pulse from the neutron detector for 0.1 μsec in experiment I and 0.5 μsec in experiment II. The use of this criterion resulted in a certain loss (not more than 25%) in the efficiency with which neutrons from the reaction under investigation were detected, but this was allowed for in subsequent analysis. The neutrons were recorded in the energy range between 0.3 and 1.0 MeV (equivalent electron energy) and the electrons were recorded for energies $E \geq 0.3$ MeV.

TABLE II. Summary of Data Characterizing Exposures of the Deuterium Target.

Exposure number	Target temperature, °K	Deuterium pressure, atm	Monitor counting rate, $N(23) \times 10^{-6}$	Counting rate $N(234\bar{5}) \times 10^{-3}$	Number of electrons $N_e \times 10^{-3}$	Number of neutrons N_n
			Experiment I			
1	250	69.0	235	992	53.5±0.8	1061±52
2	213	59.3	229	951	54.9±0.8	915±47
3	183	51.5	220	932	48.2±0.7	692±38
4	155	43.8	371	1569	83,5±1.2	1007±53
5	117	34.4	204	860	46.7±0.7	267±24
			Experiment II			
1	298	46.8	217	841	22.8±0.2	261±20
2	336	52.9	321	930	26.3±0.2	315±20
3	380	60.0	239	652	16.8±0.2	205±17
4	263	41.3	353	1295	32.0±0.3	329±25
5	213	48,5	190	835	19.5±0.2	282±24

The listed values of N_e and N_n were corrected for background which was determined in special experiments. In experiment I, the electron background was mainly due to the decay of muons stopping in the plastic phosphor of counter 4 (the muon lifetime in this material is 2 μsec). The relative contribution of this source of background, found in an experiment with an evacuated target, was 0.23 ± 0.01.

In experiment II, the background due to stopping muons in the phosphor of counter 4 (cesium iodide) could be almost entirely suppressed by using the condition $t \geq 0.5$ μsec, where t is the time of detection of the event relative to the muon-stopping time. When this criterion was used, the background in the number of electrons was entirely due to random coincidences and its contribution amounted to about 5%.

The background for neutrons was determined by using helium in the target. The amount of helium was equivalent in relation to the number of stopping muons to the amount of deuterium in the main exposures. In experiment I, measurements with the helium filling were performed at 250, 190, and 120°K, whereas, in experiment II, they performed at 210, 300, and 380°K. The background for neutrons was about 10% at 250°K and about 30% at 120°K.

The uncertainties in N_e and N_n indicated in Table II include both statistical errors and errors introduced during the separation of neutrons and electrons. The latter amounted to less than 3% for neutrons and were negligible for electrons.

The time distributions of neutrons were analyzed for each exposure of the deuterium target with the aid of the expression

$$\frac{dN_n}{dt} = N_\mu \eta_n \frac{\lambda_{dd\mu}\varphi\lambda_f}{2(\lambda_{dd\mu}\varphi+\lambda_f)}[\exp\{-\lambda_0 t\} - \exp\{-(\lambda_0+\lambda_{dd\mu}\varphi+\lambda_f)t\}], \quad (2)$$

which was obtained by taking into account the regeneration of muons in reactions (1a) and (1b). In this expression, N_μ is the number of stopping muons in deuterium, η_n is the efficiency with which neutrons from reaction (1b) were detected, $\varphi = \rho_{gas}/\rho_{liquid}$ are the densities of gaseous and liquid deuterium, λ_f is the rate of the fusion reaction (1) in the $dd\mu$ molecule, and $\lambda_0 = 4.55 \times 10^5$ sec^{-1} is the decay constant of the free muon. Analysis of the neutron time distributions yields (to a confidence level of 90%)

$$\lambda_f > 9\lambda_0, \quad (3)$$

which is in agreement with theoretical calculations[16] and our previous measurements.[4] The neutron time spectrum, summed over all exposures, is shown in Fig. 4.

If we integrate (2) between t_1 and t_2 for $\lambda_f \gg \lambda_0$, we obtain

$$N_n = N_\mu \eta_n \frac{\lambda_{dd\mu}\varphi}{2\lambda_0}(e^{-\lambda_0 t_1} - e^{-\lambda_0 t_2}). \quad (4)$$

We note that, if we replace (3) with $\lambda_f = \infty$, the accompanying change in N_n, given by (4), is very slight (less than 1%).

The time distribution of electrons from the decay of muons stopping in the deuterium gas is

$$dN_e/dt = N_\mu \eta_e \lambda_0 e^{-\lambda_0 t},$$

so that the number of electrons recorded by counters $1n-5n$ in the time interval (t_1, t_2) is

$$N_e = N_\mu \eta_e (e^{-\lambda_0 t_1} - e^{-\lambda_0 t_2}) \quad (5)$$

(where η_e is the efficiency of detection of electrons by counters $1n-5n$). It can be shown from (4) and (5) that the rate $\lambda_{dd\mu}$ can be expressed in terms of the ratio N_n/N_e, measured at different temperatures, as follows:

$$\lambda_{dd\mu}(T) = \frac{2\lambda_0}{\varphi}\frac{N_n}{N_e}(T)\frac{\eta_e}{\eta_n} = \text{const} \cdot \frac{N_n}{N_e}(T). \quad (6)$$

We note that normalization to the number of electrons recorded by the same detectors as those used to record neutrons from reaction (1b) results in certain definite advantages. In particular, a change in the characteristics of counters 4 and 5 with temperature will then have the same effect on the observed neutron and electron yields and will not change their ratio.

Figure 5 shows the values of $\lambda_{dd\mu}(T)$ obtained in our experiments. To obtain these values, we corrected for certain differences (<5%) in the efficiency of detection of neutrons from reaction (1b) at different temperatures, which appeared through the use of anticoincidences with counter 5 and were connected with a variation in its characteristics with temperature. The

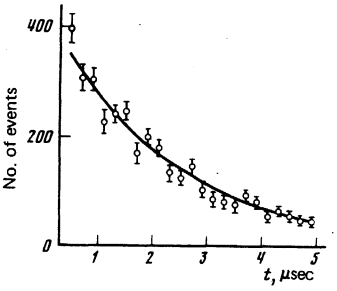

FIG. 4. Time separation of neutrons from reaction (1b), summed over all the exposures of the deuterium target. Points—experimental, curve—calculated from (4).

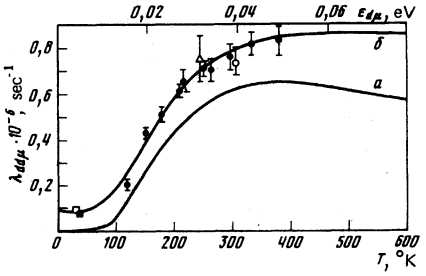

FIG. 5. Rate constant $\lambda_{dd\mu}$ as a function of the energy of $d\mu$ atoms (deuterium temperature). Experimental data: ■—Fetkovich *et al.*[1]; □—experimental[2]; △—Dzhelepov[3]; ○—experimental[4]; ●—present work. Solid curves—calculated in accordance with Ref. 10, using optimum values of the parameters listed in Table III. Curve a—resonance mechanism of production of $dd\mu$ molecules; curve b—sum of the resonance mechanism and nonresonance $E1$, $E0$ transitions [Eq. (7)].

figure also shows the experimental values of $\lambda_{dd\mu}$ reported in the literature.[1-4]

Our measured values of N_n/N_e, obtained at different temperatures, were approximately represented by

$$(N_n/N_e)_k = C\lambda_{dd\mu}(T_k),$$

where[10]

$$\lambda_{dd\mu}(T) = \lambda_1 + \lambda_2 T + \lambda_3 T^{-3/2} \exp(-\varepsilon_{d\mu}^0/kT). \quad (7)$$

Here, T is the absolute temperature, $k = 8.6167 \times 10^{-5}$ eV·deg^{-1} is Boltzmann's constant, and the terms λ_1 and $\lambda_2 T$ represent the contribution of nonresonance $E1$, $E0$ transitions to the rate of production of the $dd\mu$ system. The last term represents the resonance mechanism, where $\varepsilon_{d\mu}^0$ is the kinetic energy of the $d\mu$ atom at which $\lambda_{dd\mu}$ is a maximum. In the course of the analysis, we varied the parameters C, λ_1, λ_2, λ_3 and the position of the maximum $\varepsilon_{d\mu}^0$ on the $\lambda_{dd\mu}(T)$ curve. The normalizing factor

$$C = \frac{\varphi}{2\lambda_0}\frac{\eta_n}{\eta_e}$$

was varied separately for each experiment because the neutron and electron detection efficiencies in experiments I and II were different. The values of $\lambda_{dd\mu}$ reported in the literature[1-4] were included in the analysis for absolute normalization.

The optimum values of C, $\lambda_1 - \lambda_3$, $\varepsilon_{d\mu}^0$ are listed in Table III. Since λ_2 was determined with considerable uncertainty, we carried out calculations with the fixed value $\lambda_2 = 0.30 \times 10^{-3}$ deg^{-1}·sec^{-1}, predicted theoretically by Vinitskiĭ *et al.*[10] The data listed in Table III indicate that the position of the peak on the resonance curve then changes only slightly.

We assumed in our analysis that the muon attachment coefficient w_s of the charged products from reactions (1a) and (1b) was zero, i.e., that the muons were always liberated in these reactions and again formed $d\mu$ atoms. It is readily shown that, if the value $w_s = 0$ is replaced by the maximum possible value[3] $w_s = 0.13$, this has practically no effect on our values for $\lambda_{dd\mu}(T)$.

Figure 5 shows a plot of (7) for the parameter values listed in Table III (variant 2). It is clear from this figure that our results are in good agreement with the predictions based on the resonance mechanism for the production of $dd\mu$ molecules, and confirms its existence. Comparison of our values $\varepsilon_{d\mu}^0 = 0.050 \pm 0.003$ eV and $\lambda_{dd\mu}^0 \equiv \lambda_{dd\mu}(\varepsilon_{d\mu}^0) = (0.85 \pm 0.11)$ sec^{-1} with the calculations reported by Vinitskiĭ *et al.*[10] enables us to determine the binding energy of the $dd\mu$ molecule in the $L = 1$, $v = 1$ state with a high precision. The result is $\varepsilon_{dd\mu} = -2.196 \pm 0.003$ eV, which differs from the value reported by Vinitskiĭ *et al.*[10] by 0.24 eV. This is within the range of precision of the calculations. More precise calculations of the above level energy are highly desirable because they could then be used to determine the contribution of vacuum polarization to this energy level. Published data[17] indicate that this contribution should be +0.008 eV in the case of the $L = 1$, $v = 1$ state of the $dd\mu$ system.

In conclusion, we note one further important point. The calculations reported by Vinitskiĭ *et al.*[10] indicate

TABLE III. Optimum values of the parameters in the function $(\lambda_{dd\mu}(\varepsilon_{d\mu}))$, deduced from an analysis of experimental data.

Quantity		Value	
		variant 1	variant 2
C, 10^8 sec	expt. I	2.82±0.26	2.80±0.26
	expt. II	1.54±0.13	1.47±0.12
λ_1, 10^{-6} sec^{-1}		0.122±0.016	0.0880±0.0011
λ_2, 10^{-3} deg^{-1} sec^{-1}		−0.739±0.474	0.30
λ_3, 10^{-10} deg$^{3/2}$ sec^{-1}		2.954±0.447	2.200±0.279
$\varepsilon_{d\mu}^0$, eV		0.0489±0.0019	0.050±0.003

that, just as in the case of the $dd\mu$ molecule, the $dt\mu$ system should have an energy state with $L = 1$, $v = 1$. The calculated level energy is $\varepsilon_{dt\mu} = -1.1$ eV. The existence of this level should lead to the resonance production of $dt\mu$ molecules.[10,18] The agreement between our experimental data on the resonant nature of $dd\mu$ production and the theoretical conclusions may be looked upon as a serious experimental argument in favor of the theoretically predicted existence of the same mechanism in the production of the $dt\mu$ system in a mixture of deuterium and tritium. The essential point is that calculations[10] indicate that the rate of production of the $dt\mu$ molecules should exceed the rate of production of $dd\mu$ by a factor of roughly 100, so that one muon can produce up to 100 fusion reactions of the form $d + t \to He^4 + n$ during its lifetime (assuming that the attachment coefficient of the μ^- meson to the resulting He^4 is small).

Thus, μ-catalysis of the $d + t$ reaction can serve as a source of a large number of energetic neutrons ($E_n \sim 14.1$ MeV).

The authors are indebted to S. S. Gershteĭn and L. I. Ponomarev for fruitful discussions, to V. V. Antipov for assistance in the preparation of the experiments, and to Z. Seres for assistance in the analysis of the experimental data.

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Translated by S. Chomet