Resonant formation of $dd\mu$ muonic molecules

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The rates $\lambda_{dd\mu}(T)$ of resonant formation of $dd\mu$ muonic molecules in collisions between $d\mu$ muonic atoms and D_2, HD, and DT molecules at temperatures $10 \lesssim T \lesssim 1000$ K are calculated. The calculated rates $\lambda_{dd\mu}(T)$ for the $d\mu + D_2 + [(dd\mu)dee]$ reaction agree well with the measured ones in the entire range of the deuterium temperature.

1. INTRODUCTION

Resonant production of $dd\mu$ muonic molecules was first observed in experiment by Dzhelepov and coworkers.[1] In contrast to ordinary nonresonant of $dd\mu$ molecule production, in which the binding energy of the produced muonic molecules is carried away by the conversion electron

$$d\mu + D_2 \rightarrow [(dd\mu)de]^+ + e, \tag{1}$$

the mechanism proposed in 1967 by Vesman[2] for resonant production of $dd\mu$ molecules is essentially the following. As the $d\mu$ muonic atom with kinetic energy ε_p approaches one of the nuclei of the D_2 molecule, it joins it to form the molecule $dd\mu$ (more accurately the muon-molecular ion $(dd\mu)^+$) in a weakly bound rotational-vibrational state $(J = v = 1)$; this molecule becomes then the "heavy nucleus" of the muonic-molecular complex $[(dd\mu)dee]$:

$$d\mu + D_2 \rightarrow [(dd\mu)dee]. \tag{2}$$

The released binding energy $|\varepsilon_{Jv}| = |\varepsilon_{11}|$ of the muonic molecule $dd\mu$ goes then to excitation of the rotational-vibrational states (vK) of the muon-molecular complex $[(dd\mu)dee]_{vK}$ (see Fig. 1).

If $|\varepsilon_{11}| \sim 2$ eV, the reaction (2) has at deuterium temperatures $T \gtrsim 100$ K a rate $\lambda_{dd\mu} \sim 10^6$ s^{-1}, which is several times ten larger than the rate of the nonresonant process (1).

The existence of a weakly bound state $(J = v = 1)$ of the $dd\mu$ molecule was reliably established in theoretical papers in 1973–1983 (Ref. 3). This made it possible to carry out in 1977 the first sufficiently corroborated calculation of the dependence $\lambda_{dd\mu}(T)$ of the rate of production of the $dd\mu$ molecule on the temperature T of the medium. On the whole, however, this calculation was only by way of a demonstration, since the value of ε_{11} was known at that time only accurate to ~ 0.1 eV, and was itself determined by comparing the theoretical $\lambda_{dd\mu}(T)$ curve with experiment[5] at $T \approx 300$ K. Moreover, the calculation of Ref. 4 did not take into account the spin structure of the $d\mu$ atoms in the $dd\mu$ molecules, of the rotational structures of the D_2 molecule and of the $[(dd\mu)dee]$ complexes, or of the features of the kinetic muonic-molecular processes in deuterium, viz., decay and stabilization of the muonic-molecular complexes $[(dd\mu)dee]$, the nuclear reaction and the cascade transition in the $dd\mu$ muonic molecule, the spin flip of the $d\mu$ atom in collisions, and others.

All these effects are taken into account in the present paper.[1)] The energy of the $(J = v = 1)$ state of the $dd\mu$ molecule was recently calculated with high accuracy ($\sim 10^{-3}$ eV),[7,8] and the rates λ_f^{Jv} of the nuclear reaction[9] and $\lambda_{\text{dex}}^{Jv}$ of the de-excitation[10] in the $dd\mu$ molecule were also found. In addition, new more accurate expressions were obtained in Refs. 11–13 for the rate $\lambda_{dd\mu}(T)$ (see also Refs. 14 and 15). On the basis of the foregoing results, we have theoretically calculated in the present paper the rates $\lambda_{dd\mu}(T)$ for the reactions

$$d\mu + DA \rightarrow [(dd\mu)aee], \tag{2a}$$

where $A = D, H, T$; $a = d, p, t$, without using any adjustment parameters or additional hypotheses. The calculated $\lambda_{dd\mu}(T)$ dependence describes well all the known experimental data on the reaction (2), which we use hereafter as the example to demonstrate the calculation procedure.

2. PLAN OF CALCULATING THE RATES $\lambda_{dd\mu}(T)$

When muonic atoms $(d\mu)_F$ in the spin state F collide with a molecule $(D_2)_{v_i}^{K_i}$ in vibrational-rotational state $(v_i K_i)$, excited muon-molecular complexes $[(dd\mu)dee]_{v_f K_f}$ are produced in a vibrational-rotational state $(v_f K_f)$ are produced in accordance with the reaction

$$(d\mu)_F + (D_2)_{v_i K_i} \rightarrow [(dd\mu)_S^{Jv} dee]_{v_f K_f} \tag{3}$$

at a rate $\lambda_{FK_i SK_f}$, where S is the total spin of the $dd\mu$ molecule.

The complex produced either decays with a rate $\Gamma_{SF'}$ into the initial fragments (in the general case $F' \neq F$, $v_i' \neq v_i$ and $K_i' \neq K_i$), or is stabilized at a rate λ_{dex} by the de-excitation $(Jv) \rightarrow (J'v')$ of the $dd\mu$ molecules:

$$[(dd\mu)_S^{Jv} dee] \xrightarrow{\Gamma_{SF'}} (d\mu)_{F'} + (D_2)_{v_i' K_i'} \tag{4a}$$

$$[(dd\mu)_S^{Jv} dee] \xrightarrow{\lambda_{\text{dex}}} [(dd\mu)_S^{J'v'} de]^+ + e \tag{4b}$$

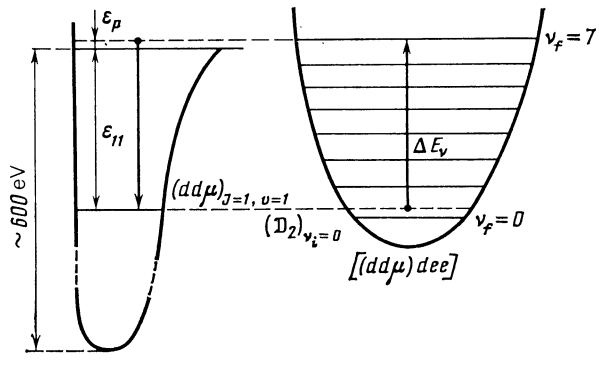

FIG. 1. Resonant formation of muonic molecules $dd\mu$: the reaction is possible if the resonance condition $\varepsilon_p + |\varepsilon_{11}| = \Delta E_v$ is met.

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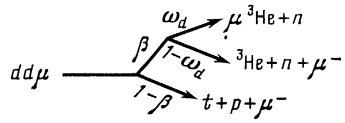
The decay of the complexes is hindered also by the nuclear fusion reactions



(5)

which proceed at a rate λ_f and with corresponding probabilities $\beta = 0.58$ and $1-\beta = 0.42$,[16,17] while μ^- sticks to the helium in the first channel with probability $\omega_d = 0.122$.[17,18]

The rate of the reaction (3) is[2)] (Refs. 11 and 13)

$$\Lambda_{FK_i,sK_f}=\lambda_{FK_i,sK_f}\varphi,$$

$$\lambda_{FK_i,sK_f}=2\pi N_0 W_{Fs}\int d^3p\, f(\varepsilon_p,T)\,|V_{fi}|^2\delta(E_i-E_f), \qquad (6)$$

where $N_0 = 4.25\cdot 10^{22}$ cm^{-3} is the liquid-hydrogen density, $\varphi = N/N_0$ is the relative density of the deuterium, $\mathbf{p}$ is the momentum of the relative motion of the $d\mu$ atom and the molecule D_2, $f(\varepsilon_p T) = 2(\varepsilon_p/\pi T^3)^{1/2}\exp(-\varepsilon_p/T)$ is the Maxwellian distribution in the relative collision energy ε_p at a given temperature[3)] T, V_{fi} is the coordinate matrix element of the transition from the initial state i to the final one f:

$$|V_{fi}|^2=\frac{1}{2K_i+1}\sum_{M_{K_i},M_{K_f},M_J}|\langle\Psi^{(f)}|\hat{V}|\Psi^{(i)}\rangle|^2, \qquad (7)$$

M_{K_i}, M_{K_f}, and M_J are the third projections of the orbital momenta K_i, K_f, and J.

The overlap of the spin functions of the initial (F) and final (S) states, with allowance for the identity of the nuclei in the $dd\mu$ molecule, is taken into account by the factor[11]

$$W_{Fs}=2(2S+1)\begin{Bmatrix} 1/2 & 1 & F \\ 1 & S & 1 \end{Bmatrix}^2, \qquad (8)$$

where

$$\begin{Bmatrix} S_\mu & S_d & F \\ S_d & S & I \end{Bmatrix}$$

is the Wigner 6j symbol, and $I = 1$ is the total spin of the two deuterium nuclei in the $dd\mu$ molecule.

The coordinate parts of the wave functions and the transition operator are given by

$$\Psi^{(i)}=\psi_{d\mu}(\mathbf{r}_1)\,\psi_{D_2}^{v_iK_i}(\rho_1)\,e^{i\mathbf{p}\rho_2},$$
$$\Psi^{(f)}=\psi_{dd\mu}^{Jv}(\mathbf{r},\mathbf{R})\,\psi_{MD}^{v_fK_f}(\rho),$$
$$\hat{V}=(\mathbf{d}\rho/\rho)\,\partial W(\rho)/\partial\rho. \qquad (9)$$

Here $\mathbf{r}_1$ and $\boldsymbol{\rho}_1$ are the internal coordinates of the $d\mu$ atom and of the D_2 molecule, $\boldsymbol{\rho}_2$ is the relative coordinate, $\mathbf{r}$ and $\mathbf{R}$ are the Jacobi coordinates of the $dd\mu$ molecule, $\boldsymbol{\rho}$ is the internuclear coordinates of the muonic-molecule complex MD = [$(dd\mu)dee$], $\mathbf{d}$ the dipole moment of the $dd\mu$ molecule, and $W(\rho)$ the $^1\Sigma_g$ term of the H_2 molecule. (The grounds for choosing expressions (6)–(9) are given in Refs. 11–15.)

The initial and final energies E_i and E_f for the reaction (3) are respectively

$$E_i=E_F(d\mu)+E_{v_iK_i}(D_2)+\varepsilon_p,$$
$$E_f=E_S(dd\mu)+E_{v_fK_f}(MD),$$
$$E_F(d\mu)=E_{1s}(d\mu)+\Delta E_F,$$
$$E_S(dd\mu)=E_{Jv}(dd\mu)+\Delta\varepsilon_S, \qquad (10)$$

where $E_{1s}(d\mu)$ and ΔE_F are the nonrelativistic energy and the relativistic hyperfine splitting of the $1s$ state of the $d\mu$ atom, corresponding to the total spin F, $E_{Jv}(dd\mu)$ and $\Delta\varepsilon_S$ are the corresponding values for the (Jv) state of the $dd\mu$ molecule with total spin S.[21,22] The resonance condition $E_f = E_i$ can be represented in the form $\varepsilon_p - \varepsilon_{if}$, where

$$\varepsilon_{if}=\varepsilon_{11}+\Delta\varepsilon_{FS}+\Delta E_{v_iK_i,v_fK_f}=\varepsilon_0+\Delta\varepsilon_{FS}+\Delta E_{if}. \qquad (11)$$

Here $\varepsilon_0 = \varepsilon_{11} + \Delta E_v$ is the resonance defect,

$$\Delta E_v=\Delta E_{v_i0,v_f0}=E_{v_f0}(MD)-E_{v_i0}(D_2),$$

$\varepsilon_{11} = E_{11}(dd\mu) - E_{1s}(d\mu)$ is the energy of the $(J = v = 1)$ of the $dd\mu$ molecule without allowance for the spin splitting, $\Delta\varepsilon_{FS} = \Delta\varepsilon_S - \Delta\varepsilon_F$ is the spin splitting of the energy level ε_{11},

$$\Delta E_{v_iK_i,v_fK_f}=E_{v_fK_f}(MD)-E_{v_iK_i}(D_2)=\Delta E_v+\Delta E_{if},$$
$$\Delta E_{if}=\{E_{v_fK_f}(MD)-E_{v_f0}(MD)\}$$
$$-\{E_{v_iK_i}(D_2)-E_{v_i0}(D_2)\}=\varepsilon_f-\varepsilon_i. \qquad (11a)$$

The energies $E_{v_iK_i}(D_2)$ and $E_{v_fK_f}(MD)$ of the rotational-vibrational states of the molecule D_2 and of the complexes MD were calculated in Ref. 23 accurate to $\sim 10^{-4}$ eV.

The energy-level splitting schemes of the initial and final states are shown in Figs. 2 and 3. We used in the calculation the values[4)] $\varepsilon_{11} = -1.964$ eV and $\Delta\varepsilon_{FS}$ and ΔE_{if} obtained on the basis of the results of Refs. 8 and 21–25. For the reaction (3) at $v_i = 0$ and $v_f = 7$ the resonance defect is $\varepsilon_0 = 0.0337$ eV, while the values of $\Delta\varepsilon_{FS}$ are

$$\Delta\varepsilon_{1/2\,1/2}=0.0163\text{ eV},\ \Delta\varepsilon_{1/2\,3/2}=0.0403\text{ eV},$$
$$\Delta\varepsilon_{3/2\,1/2}=-0.0322\text{ eV},\ \Delta\varepsilon_{3/2\,3/2}=-0.0082\text{ eV}. \qquad (12)$$

The rate λ_{FS} of formation of the muonic molecules $(dd\mu)_S$ from the initial state the muonic atoms $(d\mu)_F$, averaged over the rotational states of the molecule $(D_2)_{Ki}$ and summed over the final states of the complex $(MD)_K$, is equal to

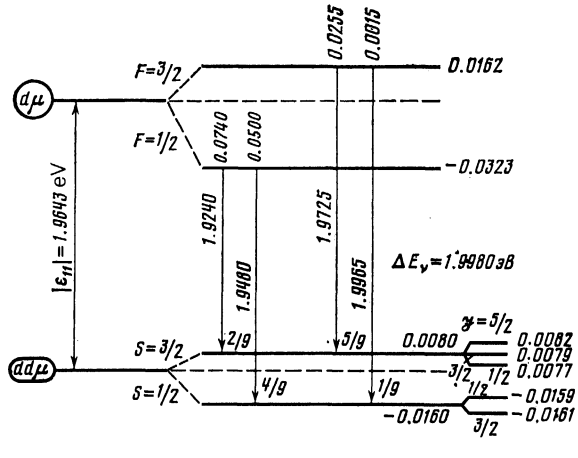


FIG. 2. Hyperfine and fine splitting of the energy level of the atoms $(d\mu)_F$ and of the muonic molecules $(dd\mu)_{S,J}$, $\mathbf{F} = \mathbf{S}_\mu + \mathbf{S}_d$ is the total spin of the $d\mu$ atom, $\mathbf{S} = \mathbf{S}_\mu + \mathbf{S}_d + \mathbf{S}_d$ and $\mathbf{J} = \mathbf{S} + \mathbf{J}$ are respectively the total spin and the total angular momentum of the $dd\mu$ molecule. The transition energies for the reaction (3) are marked on the vertical arrows of the $(F\to S)$ transitions, their resonant energies $\varepsilon_0 + \Delta F_{FS}$ at the start of the arrows, and the corresponding weights W_{FS} at their ends.

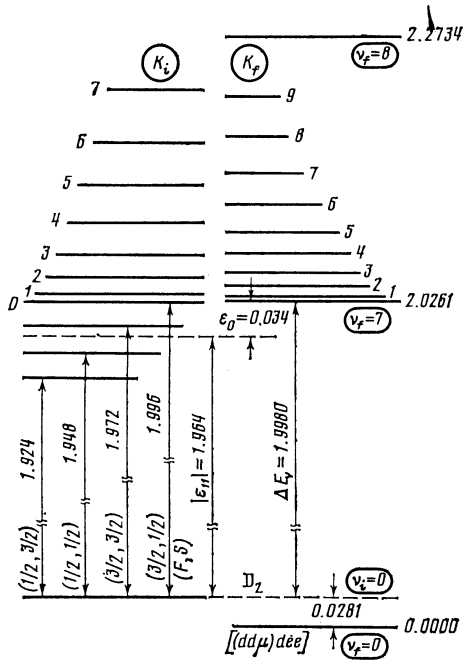


FIG. 3. Resonant transition s in reaction (3) with allowance for the rotational level splitting of the $(D_2)_{v_i K_i}$ molecule and the $[(dd\mu)dee]_{v_f K_f}$ complex. On the left are marked the values of $|\varepsilon_{11}+\Delta\varepsilon_{FS}|+\varepsilon_i$, on the right the values of (11) and (11a), and all values are referred to the ground state of the molecule $(D_2)_{v_i=K_i=0}$

$$\lambda_{FS}=\sum_{K_i,K_f}\lambda_{FK_i,SK_f}\omega(K_i)\theta(\varepsilon_{if}), \tag{13a}$$

where

$$\theta(x)=1 \text{ if } x>0,\ \theta(x)=0 \text{ if } x<0,$$
$$\varepsilon_i=E_{v_iK_i}(D_2)-E_{v_i0}(D_2) \tag{13b}$$

is the rotational energy of the D_2 molecule,

$$\omega(K_i)=\xi(K_i)Z_i^{-1}(2K_i+1)\exp\{-\varepsilon_i/kT\}, \tag{13c}$$

is the Boltzmann distribution of the D_2 molecules over the rotational degrees of freedom. Here Z_i is the partition function over the rotational states of the D_2 molecule at a given temperature T,

$$\xi(K_i)=2/3 \quad \text{for even} \quad K_i,$$
$$\xi(K_i)=1/3 \quad \text{for odd} \quad K_i \tag{13d}$$

(for the case of equal populations of the ortho and para states of the D_2 molecule).

The resonance process (2) is possible only if $\varepsilon_{if}>0$. At temperatures $T\lesssim 10^3$ K the rotational states $K_i<10$ are excited, i.e., at the resonance (11) it is possible to have ~ 100 different values of ΔE_{if} corresponding to different (K_i, K_f) combinations, and 209 terms in the sum (13) that is bounded by the condition $\varepsilon_{if}>0$. Actually, at a given temperature T, a substantial contribution to the sum (13) is made only by 2–3 terms, for which $\varepsilon_{if}\sim T$.

The rate $\Gamma_{SF'}$ of the decay of the complex (4) is equal to[5)]

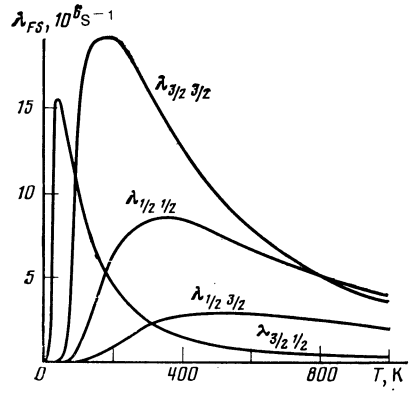


FIG. 4. Temperature dependence of the partial rates λ_{FS} of the process $(d\mu)_F+D_2\to[(dd\mu)_s dee]$, which correspond to the transition scheme of Fig. 2. The sharp peak of the rates $\lambda_{3/2\ 1/2}$ at $T=46$ K corresponds to the resonance energy $\varepsilon_{if}=5.9$ meV of the dipole transition $(F=\frac{3}{2}, K_i=0)\to(S=\frac{1}{2}, K_f=1)$.

$$\Gamma_{SF'}=\sum_{K_i',K_f}\Gamma_{SK_f,F'K_i'}\omega(K_f)\xi(K_i'),$$
$$\omega(K_f)=Z_f^{-1}(2K_f+1)\exp\{-\varepsilon_f/T\},$$
$$Z_f=\sum_{K_f}\omega(K_f), \tag{14}$$

where SK_f, FK_i is defined by the expresion[13,26,27]

$$\Gamma_{SK_f,FK_i}=\frac{2}{\pi}(2\mu_2{}^3\varepsilon_{if})^{1/2}W_{FS}\frac{2F+1}{2S+1}\frac{2K_i+1}{2K_f+1}|V_{fi}|^2\theta(\varepsilon_{if}), \tag{15}$$

where $\mu_2^{-1}=m_{d\mu}^{-1}+m_{D_2}^{-1}$ is the reduced mass of the system $d\mu+D_2$, and the spin factor W_{FS} is defined by Eq. (8).

The details of the calculations of the rates λ_{FS} and Γ_{SF} are given in Refs. 11–13, and their dependences on the temperature T are shown in Figs. 4 and 5. The observed rate $\lambda_{dd\mu}$ of formation of $dd\mu$ molecules is expressed in terms of the quantities λ_{FS} and Γ_{SF}, the rate $\lambda_f^{(S)}$ of the nuclear fusion (5), the rate (4b) of the Auger transitions, the rate λ_{nr} of the nonresonant formation of $dd\mu$ molecules[28] in the reaction (1), and the rates $\lambda_{FF'}$ of the spin flip of the $d\mu$ atom in collisions with deuterium nuclei. To find the corresponding

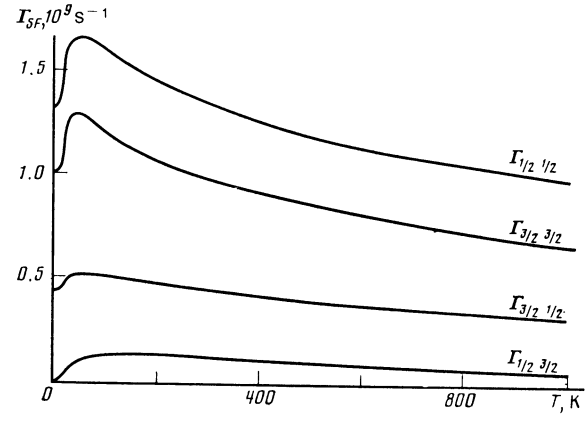


FIG. 5. Temperature dependence of the rate Γ_{SF} of the inverse decay of the complexes in the reaction $[(dd\mu)_s dee]\to(d\mu)_F+D_2$. The valid approximations are $\sum_F\Gamma_{(3/2)F}\sim\sum_F\Gamma_{(1/2)F}\approx$ const.

expressions for $\lambda_{dd\mu}$ it is necessary to examine the kinetics of the muon-catalysis processes in deuterium.

3. KINETICS OF μ-CATALYSIS PROCESSES IN DEUTERIUM

Muons of energy ~ 10 keV produce with deuterium, within a time $t_0 \sim 10^{-12}\varphi^{-1}$ s (Ref. 29), $d\mu$ atoms in highly excited states $n \gtrsim 14$ (Ref. 30). After a time $t_a \sim 10^{-11}\varphi^{-1}$ s they go over to the ground $1s$ state of the $d\mu$ atom[31] and populate statistically the sublevels $F = \frac{1}{2}$ and $F = \frac{3}{2}$ of its hyperfine structure[32] (these sublevels will hereafter by labeled 1 and 2) with weights $\eta_1 = \frac{1}{3}$ and $\eta_2 = \frac{2}{3}$. This is followed by processes (1) and (2) of $dd\mu$-molecule formation and by spin-flip processes[33,34]

$$d\mu\,(F=3/2) + d \underset{\lambda_{12}}{\overset{\lambda_{21}}{\rightleftarrows}} d\mu\,(F=1/2) + d, \tag{16}$$

with the rates of the direct and inverse processes (16) connected by the detailed-balancing relation

$$\lambda_{12} = \gamma\lambda_{21}, \quad \gamma = 2\exp\{-\Delta E/T\}, \quad \Delta E \equiv \Delta E_{d\mu}^{hfs} = 0.0485 \text{ eV}. \tag{17}$$

The general case of kinetics of μ-catalysis proceses in a mixture of hydrogen isotopes is quite complicated and is considered in Refs. 35 and 36. The sequence of the processes in pure deuterium is shown in Fig. 6, and the corresponding set of equations is

$$dN_F/dt = -(\lambda_0 + \lambda_{nr}\varphi + \lambda_F\varphi + \lambda_{FF'}\varphi)N_F + \lambda_{F'F}\varphi N_{F'} + \sum_\alpha \{\Gamma_{\alpha F} + \eta_F(1-\omega)\lambda_f^{(\alpha)}\}N_\alpha, \quad F' \neq F,$$

$$dN_\alpha/dt = -\Big(\lambda_0 + \lambda_f^{(\alpha)} + \Gamma_\alpha + \sum_{\alpha'}\lambda_{\alpha\alpha'}\Big)N_\alpha + \sum_{\alpha'\neq\alpha}\lambda_{\alpha'\alpha}N_{\alpha'} + \varphi\sum_F \lambda_{F\alpha}N_F,$$

$$dN_n/dt = \beta\sum_\alpha \lambda_f^{(\alpha)}N_\alpha, \quad \lambda_F = \sum_S \lambda_{FS}, \quad \Gamma_S = \sum_F \Gamma_{SF},$$

$$\Gamma_{\alpha F} = 0 \quad \text{if} \quad \alpha \neq S, \quad \lambda_{nr} = \sum_{\alpha\neq S}\lambda_{F\alpha} \tag{18}$$

with initial conditions

$$N_F(0) = \eta_F, \quad N_\alpha(0) = N_n(0) = 0. \tag{19}$$

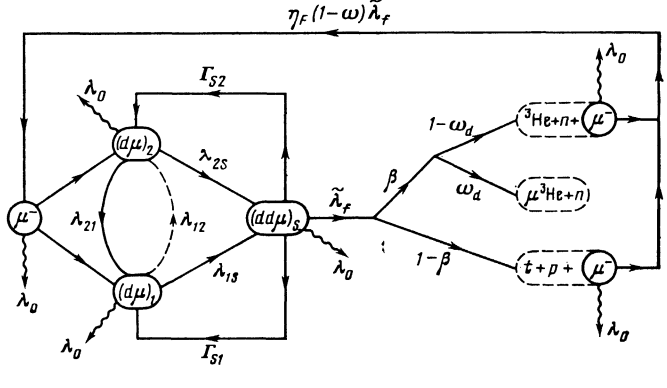


FIG. 6. Kinetics of the μ-catalysis processes in deuterium. The rates λ_{FS}, Γ_{SF}, and $\tilde{\lambda}_f$ are determined by Eqs. (13a), (14) and (21) with $\varphi = 1$.

Our notation here is: N_F is the number of $(d\mu)_F$ muonic atoms; N is the number of $(dd\mu)$ muonic molecules in a state $\alpha = \{S, Jv\}$, and for the resonant state $J = v = 1$ we put hereafter $\alpha = S$; N_n is the number of neutrons emitted in the reaction (5); $\omega = \beta\omega_d$ is the probability of muon sticking in reactions (5); $\lambda_0 = 0.455\cdot 10^6\ \text{s}^{-1}$ is the μ^- decay rate; $\lambda_f^{(\alpha)}$ is the rate of the nuclear reaction in the state of the muonic molecule $(dd\mu)_\alpha$; λ_{nr} is the total rate of nonresonant formation of $dd\mu$ molecules in reaction (1) in all the states $\alpha = (S, Jv)$ [with exception of the state $(J = v = 1)$], for which $\Gamma_{\alpha F} = 0$; $\lambda_{\alpha\alpha'}$ is the rate of the Auger transitions (4b); $\lambda_{FF'}$ is the rate of the spin flip in reactions (16). All the rates λ_F, $\lambda_{FF'}$, $\lambda_{F\alpha}$ and λ_{nr} are normalized to the liquid-hydrogen density $N_0 = 4.25\cdot 10^{22}\ \text{cm}^{-3}$. In the calculations that follow we have used the following numerical values of the characteristics of the process:

$$\lambda_{FF'} = 4.7\cdot 10^7\ \text{s}^{-1}\ [\text{Ref. 34}], \quad \lambda_{nr} = 0.4\cdot 10^5\ \text{s}^{-1}\ [\text{Refs. 4, 28}],$$
$$\lambda_f^{(s)} = 0.43\cdot 10^9\ \text{s}^{-1}\ [\text{Ref. 9}], \quad \lambda_f = 1.5\cdot 10^9\ \text{s}^{-1}\ [\text{Ref. 9}],$$
$$\lambda_{S\alpha} = 0.08\cdot 10^9\ \text{s}^{-1}\ [\text{Ref. 10}], \quad \omega = 0.071\ [\text{Refs. 17, 18}]. \tag{20}$$

The rates $\lambda_{FS}(T) \approx 10^6$–$10^7\ \text{s}^{-1}$ and $\Gamma_{SF} \approx 10^8$–$10^9\ \text{s}^{-1}$ were calculated in the present paper (see Figs. 4 and 5).

With the inequalities $\{\Gamma_{SF}, \lambda_f^{(\alpha)}\} \gg \{\lambda_{FS}, \lambda_0\}$ taken into account, it follows from Eqs. (18) that at $t \gg (\tilde{\lambda}_f + \Gamma_S)^{-1} \approx 0.5\cdot 10^{-9}$ s there is established in the system a qusistationary regime in which the conditions $dN_\alpha/dt \approx 0$ are met and the following relations hold:

$$N_S \approx \varphi\sum_F \frac{\lambda_{FS}}{\tilde{\lambda}_f + \Gamma_S}N_F, \tag{21}$$

$$\sum_\alpha \lambda_f^{(\alpha)}N_\alpha = \sum_F(\lambda_{\cdot F} + \lambda_{nr})\varphi N_F - \sum_S \Gamma_S N_S,$$

$$\tilde{\lambda}_f = \lambda_f^{(S)} + \sum_\alpha \lambda_{S\sigma}.$$

In this regime, the system (18) takes, accurate to $\{\lambda_{FS}, \lambda_0\}/\{\lambda_{FS}, \lambda_f^{(\alpha)}\} \approx 10^{-2}$, the simpler form

$$dN_n/dt = \beta\tilde{\lambda}_f N_{dd\mu}, \quad N_{dd\mu} = \sum_\alpha N_\alpha = \tilde{\lambda}_f^{-1}\varphi\sum_F N_F\tilde{\lambda}_F, \tag{22}$$

which corresponds to the scheme of the processes in Fig. 7, with effective rates

$$\tilde{\lambda}_F = \lambda_{nr} + \sum_S\{\lambda_{FS}\tilde{\lambda}_f/(\tilde{\lambda}_f + \Gamma_S)\},$$

$$\tilde{\lambda}_{FF'} = \lambda_{FF'} + \sum_S\{\lambda_{FS}\Gamma_{SF'}/(\tilde{\lambda}_f + \Gamma_S)\},$$

$$\tilde{\lambda}_f = \Big(\sum_\alpha \lambda_f^{(\alpha)}N_\alpha\Big)\Big/\sum_\alpha N_\alpha, \tag{23}$$

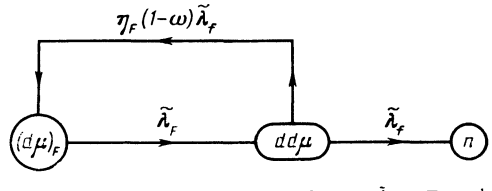


FIG. 7. Skeleton diagram, valid for $t \gg (\tilde{\lambda}_f + \Gamma_S)^{-1}$, of the μ-catalysis processes in deuterium.

where $\tilde{\lambda}_f \approx \lambda_f^{(S)}$ accurate to $\sim \lambda_{nr}/\tilde{\lambda}_F$.

It can be seen from (23) that allowance for the inverse decay (4a) of the $dd\mu$ molecules at the rates (20) decrease to about one-third the rates λ_{FS} calculated from Eqs. (13) in which this process is not taken into account. (The importance of process (4a) was first pointed out in Refs. 26 and 27; see also Ref. 37.)

For the same reason, the effective rate $\tilde{\lambda}_{FF'}$ of spin flip differs from the rate $\lambda_{FF'}$ of the reaction (16), since the complex-decay process (4a) alters the populations of the $(d\mu)_F$ states and leads to an additional depolarization whose rate is determined by the second term of Eq. (23) for $\tilde{\lambda}_{FF'}$. (This question is treated in greater detail in Ref. 38.)

In the stationary regime at $t \gg (\lambda_{21}\varphi)^{-1}$, when the population P_F of the $(d\mu)_F$ spin states reaches equilibrium because of the dynamic equilibrium of the processes (16), the time dependence of the populations N_F takes the universal form

$$N_F(t) = P_F N_{d\mu}(t), \quad \sum_F P_F = \sum_F \eta_F = 1. \tag{24}$$

When (24) is taken into account, it follows from (22) that

$$dN_{d\mu}/dt = -\lambda_c N_{d\mu}, \quad \lambda_c = \lambda_0 + \omega\lambda_{dd\mu}\varphi,$$
$$dN_n/dt = \beta\lambda_{dd\mu}\varphi N_{d\mu} = \beta\lambda_{dd\mu}\varphi e^{-\lambda_c t},$$
$$\lambda_{dd\mu} = \sum_F P_F \tilde{\lambda}_F, \tag{25}$$

where the populations P_F are given by

$$P_1 = (1+\tilde{\gamma})^{-1}, \quad P_2 = 1 - P_1, \quad \tilde{\gamma} = (\tilde{\lambda}_{12} + \eta_2\tilde{\lambda}_1)/(\tilde{\lambda}_{21} + \eta_1\tilde{\lambda}_2).$$

In the quasistationary regime, neglecting the terms $\sim\omega^2$, the solutions of (22) can be expressed in analytic form

$$N_F(t) = P_F e^{-\lambda_c t} + Q_F e^{-\Lambda t}, \tag{26}$$

where

$$Q_F = \eta_F - P_F, \quad \Lambda = \lambda_0 + \varphi(\tilde{\lambda}_{21} + \tilde{\lambda}_{12} + \eta_1\tilde{\lambda}_2 + \eta_2\tilde{\lambda}_1).$$

With allowance for (22)–(25), the neutron distribution in time becomes

$$dN_n/dt = \beta\varphi\{\lambda_{dd\mu}e^{-\lambda_c t} + (\eta_2 - P_2)(\tilde{\lambda}_2 - \tilde{\lambda}_1)e^{-\Lambda t}\}, \tag{27}$$

yielding for the integrated neutron yield per muon

$$\chi_c = \beta\{(\omega + \lambda_0/\lambda_{dd\mu}\varphi)^{-1} + \varphi(\eta_2 - P_2)(\tilde{\lambda}_2 - \tilde{\lambda}_1)\Lambda^{-1}\}. \tag{28}$$

At low deuterium temperatures $T \lesssim 100$ K, when the relations $\tilde{\lambda}_{3/2} \gg \tilde{\lambda}_{1/2} \gg \lambda_{21} \gg \{\tilde{\lambda}_{12}, \tilde{\lambda}_1, \tilde{\lambda}_2\}$, $P_1 \approx 1$, $P_2 \approx 0$, are satisfied, the neutron distribution in time becomes

$$dN_n/dt \approx \beta\varphi e^{-\lambda_0 t}(\tilde{\lambda}_1 \exp\{-\omega\tilde{\lambda}_1\varphi t\} + {}^2/_3\tilde{\lambda}_2 \exp\{-(\tilde{\lambda}_{21} + {}^1/_3\tilde{\lambda}_2)\varphi t\}).$$

4. DISCUSSION OF RESULTS

It follows from the foregoing that the calculated values of $\lambda_{dd\mu}(T)$ depend, within the framework of the calculation method used, only on two parameters: the binding energy $|\varepsilon_{11}|$ of the state $(J = v = 1)$ of the $dd\mu$ molecule and the effective rate $\tilde{\lambda}_f$ of the nuclear fusion reaction in the $dd\mu$ molecule.

Figure 8 shows the theoretical values of $|\varepsilon_{11}| \pm \delta\varepsilon_{11}$ and

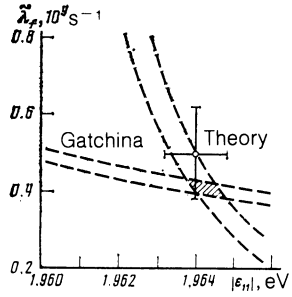


FIG. 8. Region, which follows from the experiments of Refs. 17 and 39 (shaded), in which the values of $|\varepsilon_{11}| \pm \delta\varepsilon_{11}$ and $\tilde{\lambda}_f \pm \delta\tilde{\lambda}_f$ are defined. Cross—theoretical values of ε_{11} and $\tilde{\lambda}_f$ with their probable errors.

$\tilde{\lambda}_f \pm \delta\tilde{\lambda}_f$, and also the corridors of their allowed values, which follow from the most accurate experiments of Refs. 17 and 39. The lower corridor corresponds to the pair of values $|\varepsilon_{11}|$ and $\tilde{\lambda}_f$ at which the $\lambda_{dd\mu}$ calculated from the theoretical equations (23)–(26) land on the boundary of the interval of the measured values[17]

$$\lambda_{dd\mu}(T = 293\ \text{K}) = (2.76 \pm 0.08) \cdot 10^6\ \text{s}^{-1}.$$

The upper corridor corresponds to the measured ratio[39]

$$\frac{\tilde{\lambda}_{3/2}}{\tilde{\lambda}_{1/2}}(T = 34K) = 79.5 \pm 8.0$$

of the rates of formation of the $dd\mu$ molecules from the states $F = \frac{3}{2}$ and $F = \frac{1}{2}$ of the muonic atom $(d\mu)_F$. It is easily seen that the shaded region of values of $|\varepsilon_{11}|$ and $\tilde{\lambda}_f$, which is compatible with the experimental results of both Refs. 17 and 39, does not go beyond the error limits of the theoretical values:

$$|\varepsilon_{11}| = (1.964 \pm 0{,}001)\ \text{eV}, \quad \tilde{\lambda}_f = (0.48 \pm 0{,}10) \cdot 10^9\ \text{s}^{-1}. \tag{29a}$$

Figure 9 shows the experimental[5,17,40–42] values of $\lambda_{dd\mu}(T)$. (The data of Ref. 5 were multiplied by the normalization ratio $B = 3.6$ of the rates $\lambda_{dd\mu}$ measured in Refs. 17 and 5 at $T = 293$ K.) The solid line shows the calculated $\lambda_{dd\mu}(T)$ for the values

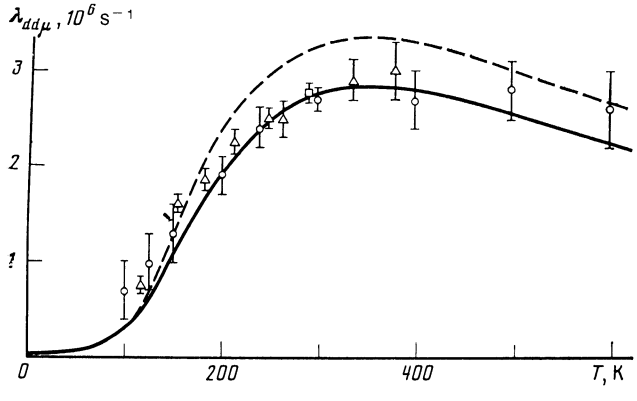


FIG. 9. Dependence of the rate $\lambda_{dd\mu}(T)$ of the reaction (2) on the temperature T. Experimental points: Δ—Dubna,[5] □—Gatchina,[17] ○—Los Alamos[40] (the Dubna data are renormalized to the value $\lambda_{dd\mu}(T = 293$ K) measured in Gatchina). Dashed curve—our calculation for $\varepsilon_{11} = -1.964$ eV, $\tilde{\lambda}_f = 0.51 \cdot 10^9$ sec^{-1}, solid curve—calculation at $\varepsilon_{11} = -1.964$ eV and $\tilde{\lambda}_f = 0.41 \cdot 10^9$ s^{-1}.

Source	$\tilde{\lambda}_2/\tilde{\lambda}_1$	$\tilde{\lambda}_{21}, 10^6\ s^{-1}$
[39]	79.5±8.0	37.4±1.5
Present paper		
$\lambda_f = 0.41\cdot 10^9\ s^{-1}$	73.3	48.6
$\tilde{\lambda}_f = 0.51\cdot 10^9\ s^{-1}$	87.7	48.1

$$|\varepsilon_{11}| = 1.964\text{eV and}\tilde{\lambda}_f = 0.41\cdot 10^9\ s^{-1}, \quad (29b)$$

taken from the shaded region of Fig. 8. It is easily seen that at these values of $|\varepsilon_{11}|$ and $\tilde{\lambda}_f$ the calculated values of $\lambda_{dd\mu}(T)$ are in good agreement with the entire aggregate of th experiments of Refs. 5, 17, and 40 (see also Table I). An additional χ^2 test confirms that the best agreement between theory and experiment is reached just at these values of $|\varepsilon_{11}|$ and $\tilde{\lambda}_f$.

Figure 9 shows also the calculated $\lambda_{dd\mu}(T)$ dependence (dashed curve) for the theoretical values

$$|\varepsilon_{11}| = 1.964\text{ eV and}\tilde{\lambda}_f = 0.51\cdot 10^9\ s^{-1}. \quad (30)$$

It can be seen that the deviations of the dashed curve from the solid one does not exceed ~20% in the entire range of T. Since the accuracy of the calculation in Ref. 13 is estimated at 1%, the errors are due mainly to the inaccuracy of $\tilde{\lambda}_f$, which amounts at present to 20–30% (Ref. 9).

The foregoing calculation of $\lambda_{dd\mu}(T)$ was carried out for an equilibrium mixture of ortho- and para-states of the D_2 molecules, corresponding to values $\xi(K_i)$ (13c). The curves for the pure ortho (odd K_i, $\xi(K_i) = 1$) and para (even K_i, $\xi(K_i) = 1$) states of D_2 differ by not more than ~5%. (See also Ref. 43, which deals with the question exclusively.)

It follows from our calculations[4] that at $T = 34$ K

$$\lambda_{dd\mu} \approx \tilde{\lambda}_{1/2} \approx \lambda_{dd\mu}^{nr} = 0.40\cdot 10^5\ s^{-1}, \quad (31)$$

which differs quite appreciably from the experimental values $(0.76 \pm 0.15)\cdot 10^5\ s^{-1}$ (Ref. 41) and $(1.03 \pm 0.04)\cdot 10^5\ s^{-1}$ (Ref. 42). One of the possible cases of such a difference is the method of reducing the experimental data. In particular, both exponentials must be taken into account in the distribution (27) of the neutrons in time, etc. More up-to-date measurements of $\lambda_{dd\mu}$ in liquid deuterium, as well as the temperature dependences of $\tilde{\lambda}_{3/2}(T)$ and $\tilde{\lambda}_{1/2}(T)$, are therefore necessary under these conditions. (See Fig. 10, where the results of our calculations are shown.) The absolute value fo $\tilde{\lambda}_{3/2}$ is extremely sensitive to the value of ε_{11} (see Fig. 8), so that measurement of $\tilde{\lambda}_{3/2}$ can yield $|\varepsilon_{11}|$ with high accuracy $\sim 10^{-4}$ eV.

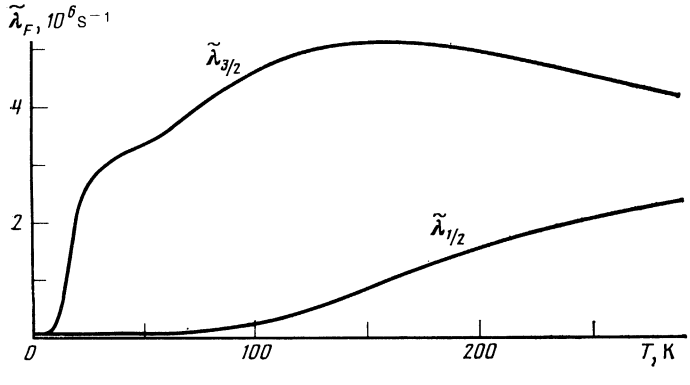


FIG. 10. Temperature dependence of the rates $\tilde{\lambda}_F(T)$ of formation of the complexes $[(dd\mu)dee]$.

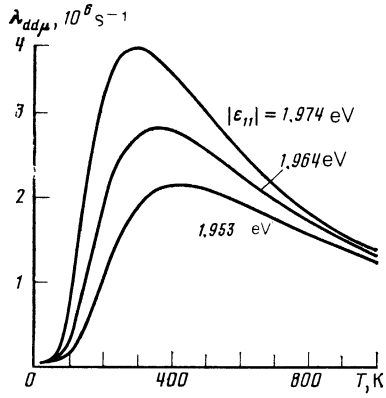


FIG. 11. Dependence of the shape of the $\lambda_{dd\mu}(T)$ curve for reaction (2) on the value of ε_{11} at $\tilde{\lambda}_f = 0.41\cdot 10^9\ s^{-1}$.

At low deuterium densities and temperatures, a contribution to the sum (13) for the partial rates λ_{FS} is made in practice by only one term corresponding to the dipole transition $(F = \frac{3}{2}, K_i = 0) \to (F = \frac{1}{2}, K_i = 1)$ with resonant energy = 5.9 meV, corresponding to a resonant temperature $T = 69$ K.

In this case $\omega(K_i) \approx 1$ and expressions (6) and (13) for $\lambda_{3/2\,1/2}(T)$ at $T \lesssim 100$ K can be represented in analytic form:

$$\lambda_{\eta_2^{1/2}}(T) \approx 4\pi^{1/2} N_0 W_{\eta_2^{1/2}} |V_{fi}|^2 \varepsilon_{if}^{1/2} T^{-3/2} \times \exp\{-\varepsilon_{if}/T\} = 2.5\cdot 10^{10}\ T^{-3/2} \exp\{-69/T\}\ [s^{-1}], \quad (32)$$

where $W_{3/2\,1/2} = \frac{1}{9}$, $|V_{fi}|^2 = 4.63\cdot 10^{-11}$ a.u.

The form of the $\lambda_{dd\mu}(T)$ plot is quite sensitive to the value of ε_{11}, therefore highly accurate measurement of the temperature dependence of the rate of resonant formation of $dd\mu$ molecules will make possible, for the first time, experimental study of relativistic effect in a three-body system.[44] The degree of sensitivity of $\lambda_{dd\mu}(T)$ the value of ε_{11} is demonstrated in Fig. 11: when ε_{11} changes by 10 meV the rate $\lambda_{dd\mu}(T)$ at $T \lesssim 100$ K is approximately doubled.

Figure 12 shows the rates of the spin flip $(F = \frac{3}{2}) \to (F = \frac{1}{2})$ of $(d\mu)_F$ atoms without allowance for formation and decay of $dd\mu$ molecules (λ_{21}) and with allowance for these processes $(\tilde{\lambda}_{21})$. It can be seen that the values of $\tilde{\lambda}_{21}$ at low temperatures (see Table I) are ~20% higher than the values of λ_{21} (Ref. 34) and their agreement with experiment[39] is worse than that of λ_{21}. The cause of this fact is not yet clear (see Ref. 45 on this subject).

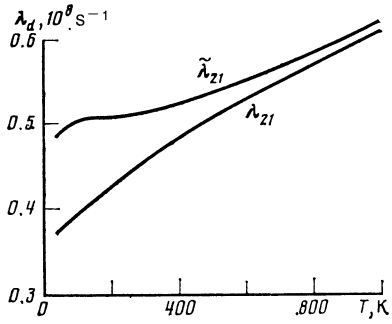


FIG. 12. Rates $\lambda_{FF'}$ and $\tilde{\lambda}_{FF'}$ (23) of the spin flip in the direct reaction (16) and with allowance for the inverse decay (4a). The contribution of the latter process reaches $\sim 30\%$ at $T \lesssim 34$ K.

5. RESONANT FORMATION OF $dd\mu$ MOLECULES IN $d\mu + DT$ AND $d\mu + HD$ COLLISIONS

The calculation procedure of Ref. 13 can be used directly also to calculate the rates λ_F of resonant formation of $dd\mu$ molecules in the reactions

$$(d\mu)_F + DT \to [(dd\mu)_s tee], \tag{33}$$

$$(d\mu)_F + HD \to [(dd\mu)_s pee]. \tag{34}$$

The resonance defects of these reactions for the reaction (33) are

$$\varepsilon_0 = -0.1789 \text{ eV} \quad \text{for} \quad v_f = 7,$$
$$\varepsilon_0 = 0.0478 \text{ eV} \quad \text{for} \quad v_f = 8,$$

and for the reaction (34)

$$\varepsilon_0 = -0.1078 \text{ eV} \quad \text{for} \quad v_f = 5,$$
$$\varepsilon_0 = 0.2151 \text{ eV} \quad \text{for} \quad v_f = 6.$$

The temperature dependences $\lambda_F(T)$ for reactions (33) and (34) are shown in Figs. 13 and 14, respectively. It must be borne in mind, however, that they represent only certain peculiarities and the scale of the rates of resonant formation of the $dd\mu$ molecules in reactions (33) and (34). To calculate the observed values of $\lambda_{dd\mu}(T)$ it is necessary to consider the kinetics of $dd\mu$ formation in the mixtures $D_2 + T_2$ and $H_2 + D_2$.

In particular, attention must be paid in the $H_2 + D_2$ mixture to the deep maximum in the cross section for $d\mu + p$ elastic scattering at $E \approx 16$ eV (Ref. 46), owing to which the thermalization time of the $d\mu$ atoms increases substantially. This in turn influences the processes of resonant $dd\mu$ molecule formation in reaction (34) and the spin flip of the $d\mu$ atoms in the reaction chain

$$(d\mu)_F + (HD)_{v_i K_i} \to [(dd\mu)_s pee]_{v_f K_f} \to (d\mu)_{F'} + (HD)_{v_i K_i'}. \tag{35}$$

A detailed examination of the mechanism of $d\mu$-atom depolarization in the $H_2 + D_2$ mixture may turn out to be necessary for a consistent description of the experimental results[47] of the characteristic of the nuclear reaction in the $pd\mu$ molecule, and also for the understanding of the measured[48] rate of μ capture by deuterium in an $H_2 + D_2$ mixture.

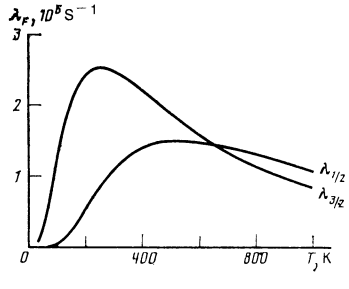


FIG. 13. Partial rates $\lambda_F(T)$ of the reaction $d\mu + DT \to [(dd\mu)tee]$ of formation of the $[(dd\mu)tee]$ complexes in the state $v_f = 8$.

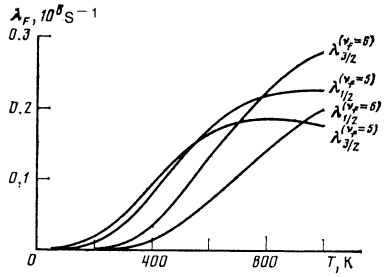


FIG. 14. Partial rates $\lambda_F(T)$ of the reaction $d\mu + DH \to [(dd\mu)pee]$ of formation of the $[(dd\mu)pee]$ complexes in the states $v_f = 5$ and 6 assuming complete thermalization of the $d\mu$ atom.

6. CONCLUSION

The foregoing calculation of the rates of resonant formation of $dd\mu$ molecules completes the research program planned in Ref. 35. Numerous characteristics of muonic molecules and muon-molecular complexes, which are needed for the calculations (e.g., the nonrelativistic energy of the level $(J = v = 1)$ of the $dd\mu$ molecule, its relativistic shifts due to polarization of vacuum, to recoil of the nuclei, and to other effects, the influence of the form factors of the nuclei, spin, and other relativistic effects, energy levels of muonic-molecule complexes, allowance for the finite dimensions of the $dd\mu$ molecule in the complexes, rates of cascade transitions in the $dd\mu$ molecule and of the nonresonant formation of $dd\mu$ molecule, and others, were calculated without the use of adjustment parameters. An exception is the rate of the nuclear reaction of the $dd\mu$ molecule, which was calculated from the measured cross section of the reaction $d(d, {}^3\text{He})n$. Apart from these exceptions, ours is an *ab initio* calculation.

The agreement between the theoretical and experimental values of $\lambda_{dd\mu}(T)$ is evidence both of use of a correct computation scheme as a whole (choice of wave functions, of the transition operator, of the averaging method, etc.) and of high accuracy of the theoretical calculations of the energy-level structure of the muonic molecules and muon-molecule complexes.

Our investigation demonstrates the feasibility of principle of precision measurements and can serve as a theoretical basis for them. The binding energy of the $(J = v = 1)$ state of the $dd\mu$ molecule can be measured accurate to $\sim 10^{-4}$ eV, i.e., with relative accuracy $\sim 10^{-7}$ (Ref. 44). Analysis of the available experimental data (Fig. 8) outlines the corridor of the admissible values of the binding energy of the state $(J = 1, v = 1)$ of the $dd\mu$ molecule:

$$|\varepsilon_{11}| = 1.9643 \pm 0.0005 \text{ eV}, \tag{36}$$

which is in very good agreement with the theoretical value (see Sec. 2). Since, however, no account is taken in this analysis of the fine structure of the $dd\mu$ molecules (splitting ~ 0.5 meV) and since the theoretical value of $\lambda_{dd\mu}^{nr}$ was used, the cited value of $|\varepsilon_{11}|$ cannot be regarded as final. The values in (36) can be made more precise by measurements of $\lambda_{dd\mu}(T)$ in wider ranges of φ and T. Particular interest attaches to precision measurements of $\lambda_{dd\mu}$ at low deuterium densities and temperatures.

The present calculation is a numerical realization of a theoretical scheme developed in Refs. 11–13 for the general case of resonant formation of $dd\mu$ and $dt\mu$ molecules. The attained agreement between theory and experiment for the $dd\mu$ molecules allows it to be used with confidence for the more complicated case of $dt\mu$ molecules. The corresponding calculations are about to be completed.

In conclusion, we take pleasure in thanking S. I. Vinitskiĭ, V. S. Melezhik, I. V. Puzynin, T. P. Puzinina, and L. B. Somov for all-around help during various stages of the work, S. S. Gershteĭn and Yu. V. Petrov for helpful discussions, and V. M. Bystritskiĭ, A. A. Vorob'ev, V. P. Dzhelepov, V. G. Zinov, G. G. Semenchuk, V. V. Fil'chenkov, and B. A. Khomenko for numerous consultations.

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[1)]The first attempt to take into account the hyperfine structure of the levels $(d\mu)_F$ and $(dd\mu)_S$ was made in Ref. 6.

[2)]Another expression for (6) with $\delta(E_i - E_f)$ replaced by a Breit-Wigner factor was proposed in Ref. 19. For a discussion of this question see Refs. 13 and 20.

[3)]The cross section $\sigma_{el} \approx 2 \cdot 10^{-10}$ cm^2 (Ref. 34) for $d\mu + d$ elastic scattering is large enough to permit a Maxwellian distribution to set in within a time $\sim 3 \cdot 10^{-8}\, \varphi^{-1}$ s (see, however, Ref. 45, where the kinetics in the $D_2 + T_2$ mixture is considered at small φ.)

[4)]This value of $\varepsilon_{11} = \varepsilon_{11}^{nrel} + \Delta\varepsilon_{\varepsilon_{11}}^{rel} + \Delta\varepsilon_{11}^{dim}$ is made up of the nonrelativistic energy $\varepsilon_{11}^{nrel} = 1.9750$ eV for pointlike nuclei,[8] the sum of the relativistic corrections $\Delta\varepsilon_{11}^{rel} = 0.0097$ eV,[22] and the corrections $\Delta\varepsilon_{11}^{dim} = 0.0010$ eV for the finite dimensions of the $dd\mu$ molecule in the muon-molecular complex.[24,25] This yields $\varepsilon_{11} = -1.9643$ eV with a probable error $\delta\varepsilon_{11} = \pm 0.0005$ eV,[8,25] which is comparable with the fine splitting (0.0006 eV) of the ε_{11} level of the $dd\mu$ molecule[22] (see Fig. 2), which is not taken into account in the present calculation.

[5)]The rates of the rotational relaxation of the produced complex ($\sim 10^{13}\varphi$ s^{-1} (Ref. 26) exceed substantially the rates λ_f, λ_{dex}, and Γ_{SF}, so that a Boltzmann distribution in the rotational states K_f is established in the complex $[(dd\mu)dee]$ during its lifetime.

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