Muon catalyzed fusion in solid deuterium

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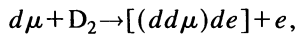
The resonant $dd\mu$ molecule formation process in solid deuterium is considered. It is shown that the rate of this process does not depend on temperature and is estimated to be $\Lambda_{dd\mu} \approx 3 \ \mu s^{-1}$.
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1. INTRODUCTION. THE "STANDARD" THEORY OF THE RESONANT $dd\mu$-MOLECULE FORMATION

Recent measurements of the $dd\mu$ molecule formation rate ($\lambda_{dd\mu}$) at TRIUMF in solid deuterium at $T=3$ K (Ref. 1) and at JINR (Dubna) in solid and liquid deuterium at $T=5.5$–30 K (Ref. 2) show that in this region the value of $\lambda_{dd\mu}$ does not depend on temperature and is equal to 2.5–3 μs^{-1}. This is in strong contradiction with the theoretical predictions[3-10] (Fig. 1).

According to the theory, muonic molecules are formed by the nonresonant Auger process

$$d\mu + D_2 \rightarrow [(dd\mu)de] + e,$$

with a rate depending weakly on the $d\mu$ atom energy, at low temperature being $\approx 0.05 \ \mu s^{-1}$, or by the Vesman resonance mechanism[3]

$$d\mu + D_2 \rightarrow [(dd\mu)d2e]_{K,\nu}. \tag{1}$$

Here symbols K and ν mean the rotational and vibrational quantum numbers, respectively.

In the Vesman scheme the energy $\epsilon_{dd\mu}$ released in $dd\mu$ molecule formation together with the $d\mu$ atom kinetic energy $\epsilon_{d\mu}$ (more precisely, the energy of the relative motion of the $d\mu$ atom and the D_2 molecule) is transferred to vibration and rotation excitation of the compound molecular complex $[(dd\mu)d2e]$ (below it will be called C):

$$\epsilon_{dd\mu} + \epsilon_{d\mu} = \Delta E_{\nu,K}. \tag{2}$$

The values of the $d\mu$ atom energies satisfying Eq. (2) are called the resonance energies and designated as ϵ_r.

In the energy balance (2) one takes into account hyperfine splitting of the $d\mu$ atom (spin $F=3/2$ and 1/2) and $dd\mu$ molecule (spin $S=3/2$ and 1/2) and the energies of the rotational states of the D_2 molecule (subscript i) and the complex (subscript f). Only the transition $F=3/2 \rightarrow S=1/2$ is important at temperatures $T<50$ K. Consequently, we shall assume that the value of $\lambda_{dd\mu}$ is the $dd\mu$ molecule formation rate from the upper $d\mu$ atom spin state $F=3/2$ to the lowest $dd\mu$ molecule spin state.

A statistical 2:1 mixture of orthodeuterium (even K_i) and paradeuterium (odd K_i) is usually used in experiments. At low temperature only the lowest values $K_i=0,1$ are significantly populated. Due to the dipole character of the transition (1), the approximate selection rule $|K_i - K_f| = 1$ is valid at low $d\mu$ atom energies.[5-8] So one should consider only the transitions $0 \rightarrow 1$, $1 \rightarrow 2$, $1 \rightarrow 0$. The values of the resonance energy for these transitions (in meV)[7,14] together with the corresponding matrix elements (in atomic units) are[8]

$$K_i = 0 \rightarrow K_f = 1, \quad \epsilon_r = 6.0, \quad V^2 = 5 \cdot 10^{-11}, \tag{3}$$

$$K_i = 1 \rightarrow K_f = 2, \quad \epsilon_r = 7.4, \quad V^2 = 3 \cdot 10^{-11}, \tag{4}$$

$$K_i = 1 \rightarrow K_f = 0, \quad \epsilon_r = -4.8, \quad V^2 = 1.5 \cdot 10^{-11}. \tag{5}$$

The positive signs in Eqs. (3) and (4) means that the energy defect in the balance (2) is compensated by the $d\mu$ atom motion energy. The negative sign in Eq. (5) corresponds to the case when some additional energy should be transferred. This situation is not realized in the "standard" theory of the binary reaction (1).

The value of $\lambda_{dd\mu}$ as a function of $\epsilon_{d\mu}$ is shown in Fig. 2. The theoretical dependence $\lambda_{dd\mu}(T)$ was obtained by integrating the function $\lambda_{dd\mu}(\epsilon_{d\mu})$ over the Maxwellian distributions in the $d\mu$ atom thermal energy. These distributions for $T=5$ K and $T=20$ K are also shown in Fig. 2. As can be seen from this figure, the thermal energy distribution at $T=5$ K does not overlap the nearest resonance. This means that the "standard" theory cannot explain the high value of $\lambda_{dd\mu}$ measured in solid deuterium. At the same time, it is in good agreement with the data obtained for gaseous and liquid deuterium.[10-13] So it is natural to suppose that the effect revealed in (Ref. 1,2) is due to the influence of the intermolecular forces in solid deuterium, i.e., the phonon processes.

2. CALCULATION OF THE $dd\mu$ MOLECULE FORMATION RATE IN SOLID DEUTERIUM

Let us write the resonance condition for reaction (1) in a solid:

$$\epsilon_{d\mu} = \epsilon_r + \Delta E, \tag{6}$$

where $\Delta E = \widetilde{E}_f - E_i$, E_i and $\widetilde{E}_f$ are the lattice energies in the initial and final states, respectively. In the initial state the lattice Hamiltonian has a form

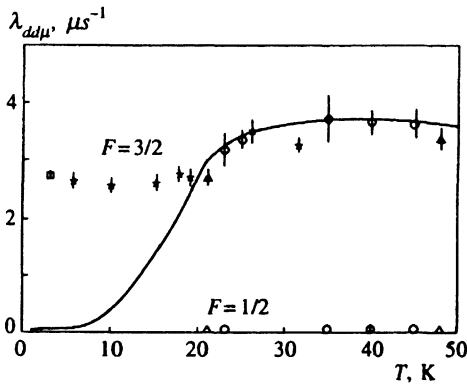$$\hat{H}_i = \sum_a \frac{\mathbf{p}_a^2}{2M} + U,$$

FIG. 1. The effective rate of muonic deuterium formation as a function of temperature. Measurements: circles,[11,12] triangles,[13] square,[1] stars.[2] Curve—calculations.[7-9]

where M is the D_2 molecule mass, the summation is made over D_2 molecules ($a=1,2,...$), and U is the lattice potential energy.

Strictly speaking, one should assume that the $d\mu$ atom interacts with the whole lattice and forms an intermediate complex C with it, i.e., one should take into account the effect of coherent formation of the μ-molecule on a lattice. However, this effect is negligible. The reason is the smallness of the overlap integral of the wave functions of the $d\mu$ atoms joined to the neighboring D_2 molecules. So let us distinguish one D_2 molecule, label it zero, and assume that the $d\mu$ atom interacts with it only. The $dd\mu$ formation rate calculated in this manner should be multiplied by the total number of the D_2 molecules in a lattice. In the following calculations we put the coordinate origin in the equilibrium position of the zero molecule.

In the final lattice state the mass of the zero molecule is changed, $M \to M_C$, so

$$\hat{H}_f = \sum_{a \neq 0} \frac{\mathbf{p}_a^2}{2M} + \frac{\mathbf{p}_0^2}{2M_C} + U = \hat{H}_i + \hat{V}_L,$$

$$\hat{V}_L = \frac{1}{2}\left(\frac{1}{M_C} - \frac{1}{M}\right)\mathbf{p}_0^2.$$
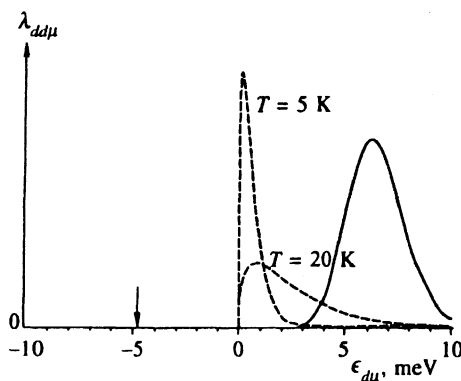

FIG. 2. Muonic deuterium formation rate as a function of $d\mu$ atom kinetic energy (solid curve) for Maxwellian distributions with $T=5$ K and 20 K (dashed curves). The arrow indicates the position of the transition (5).

It follows from the expression for $\hat{H}_i$ and $\hat{H}_f$ that the characteristic parameter of the problem is $\xi=(M_C-M)/M=1/2-1/3$ (either M_C or M may be in the denominator).

Hence, $\hat{H}_i \neq \hat{H}_f$. This is indicated by the tilde in the notation for the energy of the final lattice state (see Eq. (6)). It follows from Eqs. (3), (4), and (5) that one can neglect the $d\mu$ atom energy in Eq. (6), which is very small in comparison with $|\epsilon_r|$. Hence, the simplified resonance condition is

$$\Delta E \simeq -\epsilon_r. \tag{7}$$

If $\epsilon_r > 0$ holds, then, according to Eq. (7), we have $\Delta E < 0$, that is, phonons of energy $\sim \epsilon_r$ are absorbed. However, at the lowest energies the number of such phonons is exponentially small, $\sim\exp(-\epsilon_r/kT)$. That is why one can neglect the transitions (3) and (4) for the experimental conditions of Refs. 1, 2 and should consider only the transition (5), which goes with an escape of the even phonon numbers (the last circumstance is obvious from the form of $\hat{V}_L$). Besides, as follows from the previous consideration, one can neglect the existing lattice phonons, that is, one can consider the initial lattice state as the vacuum: $\hat{H}_i|0\rangle = E_0|0\rangle$.

The distance between the D_2 molecules in a lattice is large (≈ 6 a.u.) compared with the size of the molecule, so one can consider $d\mu$, D_2 and C to be point-like, and the interaction operator which is responsible for the transition (1) can be written in the form

$$\hat{H} = A\delta(\mathbf{r}), \tag{8}$$

where A is a constant which will be obtained later, $\mathbf{r}=\mathbf{r}_d-\mathbf{U}_0$, $\mathbf{r}_d$ are the $d\mu$ atom coordinates, $\mathbf{U}_0$ is the displacement of the D_2 molecule from the equilibrium point. To obtain the value of A let us first calculate the $dd\mu$ molecule formation rate in the double-collision approximation (1) with $\epsilon_r > 0$. The wave functions of the initial and the final state are

$$\Psi_i = e^{i\mathbf{p}\cdot\mathbf{r}}|0\rangle, \quad \Psi_f = \Psi_0(r),$$

where $\Psi_0(r)$ is the wave function of the bound state of the $d\mu$ atom and the D_2 molecule, that is the wave function of the complex C. In first order of perturbation theory with respect to the operator $\hat{V}$, we have ($\hbar=1$, the normalized volume is chosen equal to unity)

$$\Lambda_{dd\mu} = n_d \int_0^\infty f(\epsilon)d\epsilon 2\pi\delta(\epsilon - \epsilon_r)|V_{fi}|^2 = 2\pi n_d f(\epsilon_r)B, \tag{9}$$

where n_d is the number of deuterium nuclei in a unit volume, $\epsilon_r = p^2/2\mu$,

$$\mu^{-1} = (M_{d\mu})^{-1} + (M_{D_2})^{-1}, \quad f(\epsilon) = \frac{2\sqrt{2}}{\sqrt{\pi}T^{3/2}}e^{-\epsilon/T},$$

and $B^2 = A^2\Psi_0^2(0)$. Comparing Eq. (9) with Eq. (32) in (Ref. 7) we obtain $B = W_{3/2,1/2}V^2$, where $W_{3/21/2} = 1/9$ is the spin term introduced in (Ref. 7).

Taking the value of V^2 from (Ref. 7), for the transition $K_i = 0 \to K_f = 1$ one obtains $B^2 = 5 \cdot 10^{-12}$ a.u. From this and Eqs. (3) and (5) for $K_i = 1 \to K_f = 0$ one determines

$$B^2 = 1.5 \cdot 10^{-12} \text{ a.u.} \tag{10}$$

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Let us now calculate the value of $\Lambda_{dd\mu}$ for a lattice. In this case

$$\Psi_i = e^{i\mathbf{q}\cdot\mathbf{r}_d}, \quad \Psi_f = \Psi_0(r)|\tilde{n}\rangle,$$

where $\mathbf{q}$ is the $d\mu$ atom momentum in the lattice rest frame and $|\tilde{n}\rangle$ is the vector of the lattice final state: $\tilde{H}_f|\tilde{n}\rangle = \tilde{E}_n|\tilde{n}\rangle$.

As in the case of the double collision (1), one obtains in the first order of perturbation theory

$$\Lambda_{dd\mu} = n_d w_1 \sum_n \int F(\mathbf{q}) d^3 q \, 2\pi \delta(\epsilon_d + E_0 - \tilde{E}_n$$

$$- \epsilon_r)|V_{fi}|^2 \simeq 2\pi n_d w_1 Q(-\tilde{\epsilon}_r) B^2, \tag{11}$$

where $F(\mathbf{q})$ is the distribution of the $d\mu$ atoms in their momentum $\mathbf{q}$, $\epsilon_d = q^2/2M_{d\mu}$ is the mesoatom kinetic energy (which we neglect according to (7)), $\tilde{\epsilon}_r = \epsilon_r + \tilde{E}_0 - E_0$, $w_1 = 1/3$ is the statistical weight of the paradeuterium, and

$$Q(\omega) = \sum_{\tilde{n}} |\langle \tilde{n}|\exp(i\mathbf{q}\mathbf{u}_0)|0\rangle|^2 \delta(\omega - \tilde{E}_n + \tilde{E}_0)$$

$$\simeq \sum_{\tilde{n}} |\langle \tilde{n}|0\rangle|^2 \delta(\omega - \tilde{E}_n + \tilde{E}_0). \tag{12}$$

Here we put $\exp(i\mathbf{q}\cdot\mathbf{u}_0) \simeq 1$ neglecting the small terms in the expression for $\Lambda_{dd\mu}$, which are $\sim T/\omega_D$, where $\omega_D \simeq 100$ K is the Debye frequency for solid deuterium. It follows from Eqs. (11) and (12) that for $T \ll \omega_D$ the value of $\Lambda_{dd\mu}$ does not depend on temperature, which agrees with the experiments.[1,2]

The physical meaning of the term Q is clear. Let the mesomolecular complex C be formed at $t=0$ in the collision of a $d\mu$ atom with a D_2 molecule. Then at $t=0$ an instantaneous change in the lattice Hamiltonian occurs: $\tilde{H}_i \rightarrow \tilde{H}_f$, that is a "shaking". For $t<0$, the lattice was in the vacuum state $|0\rangle$. So the probability of being in the state $|\tilde{n}\rangle$ is $W_n = |\langle \tilde{n}|0\rangle|^2$. It follows from this that the probability of finding the lattice in the states with energies in the interval $(\omega, \omega+d\omega)$ is $dW = Q(\omega)d\omega$. Equation (12) provides the necessary condition

$$\int_0^\infty Q(\omega)d\omega = 1. \tag{13}$$

One should add that $Q(\omega)$ differs from zero only for $\omega > 0$ (as can also be seen from Eq. (12)).

At $\xi=0$ the term $Q(\omega)$ coincides with $\delta(\omega)$. In this case there is no energy transfer to the lattice. For $\xi \neq 0$, the term $Q(\omega)$ differs from zero in the whole interval $(0, +\infty)$. At $\xi \simeq 1$, it concentrates mainly in the region of frequencies $\omega \sim \omega_D$ characteristic of this problem. Taking into account $|\epsilon_r| \sim \omega_D$, from this and from Eq. (13) we obtain an estimate

$$Q(-\tilde{\epsilon}_r) \sim 1/\omega_D \sim 5 \cdot 10^3 \text{ a.u.} \tag{14}$$

Rigorous calculation of $Q(\omega)$ will be performed later. Unfortunately, it can be done only numerically.

In obtaining the estimate (14) we neglected the difference between the values of ϵ_r and $\tilde{\epsilon}_r$. This difference can be easily estimated from perturbation theory using the Debye phonon model:

$$\tilde{\epsilon}_r - \epsilon_r = \tilde{E}_0 - E_0 \simeq \langle 0|\tilde{V}_L|0\rangle$$

$$\simeq \left(\frac{M}{M_C} - 1\right)\frac{3}{16}\omega_D \simeq -6 \text{ K}.$$

It is seen that one can indeed neglect this difference.

From Eqs. (10), (11), and (14) we finally obtain that for $n_d = 4 \cdot 10^{22}$ cm$^{-3} = 4 \cdot 10^{-3}$ a.u.

$$\Lambda_{dd\mu} \simeq 6 \cdot 10^{-11} \text{ a.u.} = 3 \cdot 10^6 \text{ s}^{-1}. \tag{15}$$

This is an order-of-magnitude estimate.

3. DISCUSSION

In the present paper it has been shown that in the limit $T \rightarrow 0$ the value of the $dd\mu$ molecule formation rate $\Lambda_{dd\mu}$ approaches a constant, and the estimate (15) is obtained for the value of $\Lambda_{dd\mu}$ in solid deuterium. This has been done on the basis of the pseudopotential method, which originates from the work of E. Fermi and N. N. Bogoliubov.[14] As in these papers, the formula for $\Lambda_{dd\mu}$ is rigorous because a comparison with the rigorous calculation was used in obtaining the value for A in it. Doubt may arise whether the formulas (9) and (11) are applicable in our consideration. They coincide in form with the Fermi golden rule of perturbation theory, but in our case the problem parameter is not small ($\xi \sim 1$). However, as has been shown in,[5,6,9] this fact is indeed too coincidental. The validity of these formulas is not ensured by the smallness of V_L but is due to the following circumstances.
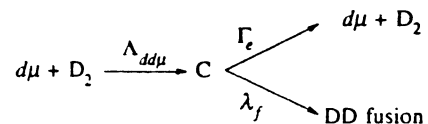
a) The smallness of the width Γ_e decay of the complex C via the elastic channel:

$$\Gamma_e \ll \omega_C, \quad \Gamma_e \ll \omega_D, \tag{16}$$

where ω_C is the vibrational quantum of the complex C.

b) The short-range character of the interaction of the neutral $d\mu$ atom with the nuclei of the D_2 molecule.

In its entirety, the $dd\mu$ molecule formation process is written as

Here λ_f is the DD fusion reaction rate in the $dd\mu$ molecule. The products of this reaction are detected in experiments. That is why the observed muonic molecule formation rate is indeed the effective rate

$$\lambda_{dd\mu} = w_f \Lambda_{dd\mu}, \quad w_f = \lambda_f/(\lambda_f + \Gamma_e).$$

According to Ref. 7, $w_f \simeq 0.3$ holds for positive resonance energies $\epsilon_r > 0$. In our case these energies satisfy $\epsilon_r < 0$, so the complex cannot decay into the initial state of the $d\mu + D_2$ system where the $d\mu$ atom spin is $F=3/2$. However, it can decay into the state with $F=1/2$. As follows from Ref. 7, the back decay rate is $\Gamma_e \sim 10^9$ s^{-1} (10^{-2} K), i.e., condition (16) is satisfied.

Another possible channel of the complex decay is the transition from its bound state with $K_f=0$ to the decay state with $K_f=1$ due to inelastic scattering on the D_2 molecule (in

our case of solid deuterium, this occurs by means of phonon absorption). Since the rate of this process is proportional to $\exp(-\Delta E_r/T)$, where ΔE_r is the transition energy, it is small.

Thus, in the case considered we have $w_f \approx 0.3$ (according to Ref. 7), and

$$\lambda_{dd\mu} \approx 10^6 \ \text{s}^{-1}.$$

It follows from this that the mechanism considered for $dd\mu$ molecule formation with lattice excitation can play an essential role in solid deuterium.

The important role of many-particle effects and transitions with $\epsilon_r < 0$ was first pointed out in Ref. 15, where the reaction (1) was considered involving a third particle (in our case it is the D_2 molecule) carrying away the excess energy. This mechanism turned out to be efficient for the $d+t$ fusion cycle at low temperature but not for a pure deuterium. But if one understands it more broadly, just the same process is realized in a solid deuterium where multiphonon phenomena take place.

In conclusion we would like to mention that there is a possibility of directly checking the results of the present work. For this one should measure the $dd\mu$ molecule formation rate with solid deuterium in the pure orthostate, which is the natural equilibrium state for the lowest temperatures. Since in solid orthodeuterium practically all D_2 molecules are in the rotational state with $K_f=0$, one should expect a sharp decrease of $\lambda_{dd\mu}$ in comparison with the nonequilibrium deuterium usually used in experiments. Note that measurements with orthodeuterium would also be important in liquid and gaseous deuterium. It would allow one to examine the theory of resonant $dd\mu$ molecule formation in a full measure.

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