Resonant $dd\mu$ Formation in Condensed Deuterium

Andrzej Adamczak

Institute of Nuclear Physics, Radzikowskiego 152, PL-31-342 Kraków, Poland

Mark P. Faifman

Russian Scientific Center, Kurchatov Institute, RU-123182 Moscow, Russia

(Dated: February 3, 2022)

Abstract

The rate of $dd\mu$ muonic molecule resonant formation in $d\mu$ atom collision with a condensed deuterium target is expressed in terms of a single-particle response function. In particular, $dd\mu$ formation in solid deuterium at low pressures is considered. Numerical calculations of the rate in the case of fcc polycrystalline deuterium at 3 K have been performed using the isotropic Debye model of solid. It is shown that the energy-dependent $dd\mu$ formation rates in the solid differ strongly from those obtained for $D_2$ gaseous targets, even at high $d\mu$ kinetic energies. Monte Carlo neutron spectra from $dd$ fusion in $dd\mu$ molecules have been obtained for solid targets with different concentrations of ortho- and para-deuterium. The recent experimental results performed in low pressure solid targets (statistical mixture of ortho-$D_2$ and para-$D_2$) are explained by the presence of strong recoilless resonance peaks in the vicinity of 2 meV and very slow deceleration of $d\mu$ atoms below 10 meV. A good agreement between the calculated and experimental spectra is achieved when a broadening of $D_2$ rovibrational levels in solid deuterium is taken into account. It has been shown that resonant $dd\mu$ formation with simultaneous phonon creation in solid gives only about 10% contribution to the fusion neutron yield. The neutron time spectra calculated for pure ortho-$D_2$ and para-$D_2$ targets are very similar. A practically constant value of the mean $dd\mu$ formation rate, observed for different experimental conditions, is ascribed to the fact that all the recent measurements have been performed at temperatures $T \lesssim 19$ K, much lower than the target Debye temperature $\Theta_D \approx 110$ K. In result, the formation rate, obtained in the limit $T/\Theta_D \ll 1$, depends weakly on the temperature.

PACS numbers: 34.10+x, 36.10.Dr
I. INTRODUCTION

Theoretical study of resonant formation of the muonic molecule \( dd\mu \) in condensed deuterium targets is the main subject of this paper. The resonant \( dd\mu \) formation, first observed by Dzhelepov and co-workers [1], is a key process of muon catalyzed fusion (\( \mu \)CF) in deuterium (see e.g. reviews [2, 3]). A muonic deuterium atom \( d\mu \) is created when a negative muon \( \mu^- \) is captured into an atomic orbital in a deuterium target. After \( d\mu \) deexcitation to the 1S state and slowing down, the \( dd\mu \) molecule can be formed in \( d\mu \) atom collision with one of the \( D_2 \) target molecules. The resonant formation is possible due to presence of a loosely bound state of \( dd\mu \), characterized by the rotational number \( J = 1 \) and vibrational number \( v = 1 \), with binding energy \( |\varepsilon_{Jv=1}| \approx 1.97 \text{ eV} \). This energy, according to the Vesman mechanism [4], is completely transferred to excited rovibrational states of the molecular complex \( [(dd\mu)\text{dece}] \). The scheme of calculation of \( dd\mu \) formation rate in gaseous deuterium has been developed for many years [5, 6, 7, 8], and has lead to a good agreement with the experiments performed in gaseous targets [9, 10]. On the other hand, this theory, when directly applied to solid deuterium targets, leads to strong disagreement with the experimental results [11, 12, 13]. Therefore, it is necessary to calculate the \( dd\mu \) formation rate with solid state effects taken into account, which is the main purpose of this paper.

Our calculations are based on the theoretical results (transition matrix elements, resonance energies) obtained in the case of \( dd\mu \) formation in a single \( D_2 \) molecule. In Sec. II the main formulas used for this case are briefly reported. A general formula for the energy-dependent \( dd\mu \) formation rate in a \( D_2 \) condensed target is derived in Sec. III using the Van Hove formalism of the single-particle response function [14]. This formula is then applied (Sec. IV) for harmonic solid targets, in particular for a cubic Bravais lattice. A phonon expansion of the response function is used to study phonon contributions to the resonant formation. Numerical results for 3 K zero pressure frozen deuterium targets (TRIUMF experimental conditions [11, 13]), with the fcc polycrystalline structure, are shown in Sec. V. The formation rates have been calculated assuming the isotropic Debye model of the solid and the values of Debye temperature and lattice constant observed in neutron scattering experiments.

The calculated rates of resonant \( dd\mu \) formation and back decay have been used for Monte Carlo simulations of \( dd \) fusion neutron and proton time spectra. Since the initial distributions of 1S muonic atom energy contain contributions from hot \( d\mu \)'s (\( \sim 1 \text{ eV} \)) [15, 16], influence of slow deceleration of \( d\mu \) atoms below 10 meV [17] on these time spectra is investigated in Sec. VI. The simulations take into account the processes of incoherent and coherent \( d\mu \) atom scattering in solid deuterium. In particular, the Bragg scattering, phonon scattering, and rovibrational transitions in \( D_2 \) molecules are included. We consider a dependence of the resonant formation rate and time spectra on broadening of the rovibrational \( D_2 \) energy levels, due to the binding of the molecules in the lattice [18].

Since it has been predicted in Refs. [19, 20, 21] that strong \( dd\mu \) formation takes place only in solid para-\( D_2 \), study of this process in pure ortho-\( D_2 \) and para-\( D_2 \) targets is another aim of this work. The neutron spectra calculated for these two solids are discussed in Sec. VII.
II. RESONANT FORMATION IN A FREE MOLECULE

First we consider resonant formation of the \( dd\mu \) molecule in the following reaction

\[
(d\mu)_F + (D_2)_{v_i K_i} \rightarrow [(dd\mu)_{S^{\nu}dee}]_{\nu_f K_f},
\]

where \( D_2 \) is a free deuterium molecule in the initial rovibrational state \( (\nu_i K_i) \) and the total nuclear spin \( I \). The muonic atom \( d\mu \) has total spin \( F \) and CMS kinetic energy \( \varepsilon \). The complex \( [(dd\mu)_{dee}] \) is created in the rovibrational state \( (\nu_f K_f) \) and the molecular ion \( dd\mu \), which plays the role of a heavy nucleus of the complex, has total spin \( S \). The rate \( \lambda_{v_i K_i, v_f K_f}^{SF} \) of the process above depends on the elastic width \( \Gamma_{v_f K_f, v_i K_i}^{SF} \) of \( [(dd\mu)_{dee}] \) complex decay \( [22, 23, 24, 25] \) in reactions

\[
\Gamma_{v_f K_f, v_i K_i}^{SF} \rightarrow (d\mu)_F + (D_2)_{v_i K_i}^{I}
\]

\[
[(dd\mu)_{S^{\nu}dee}]_{\nu_f K_f} \rightarrow \text{stabilization processes}, \quad \lambda_{f}
\]

where \( \lambda_f \) is the total rate of the stabilization processes, i.e. deexcitation and nuclear fusion in \( dd\mu \)

\[
dd\mu \rightarrow \begin{cases} 
\mu + t + p + 4.0 \text{ MeV} \\
\mu + ^3\text{He} + n + 3.3 \text{ MeV} \\
\mu^3\text{He} + n + 3.3 \text{ MeV}.
\end{cases}
\]

When fusion takes place, the muon is generally released and can again begin the \( \mu \text{CF} \) cycle. However, sometimes the muon is captured into an atomic orbital of helium (sticking), which stops further reactions.

The value of \( \Gamma_{v_f K_f, v_i K_i}^{SF} \) is given in atomic units \( (e = \hbar = m_e = 1) \) by the formula

\[
\Gamma_{v_f K_f, v_i K_i}^{SF} = 2\pi A_{if} \int \frac{d^3k}{(2\pi)^3} |V_{if}(\varepsilon)|^2 \delta(\varepsilon_{if} - \varepsilon),
\]

where \( V_{if}(\varepsilon) \) is a transition matrix element and \( \varepsilon_{if} \) is a resonance energy defined in Ref. [8]. The factor \( A_{if} \) is due to averaging over initial and summing over final projections of spins and angular momenta of the system. Vector \( k \) is the momentum of relative \( d\mu \) and \( D_2 \) motion

\[
\varepsilon = k^2/2M,
\]

and \( M \) is the reduced mass of the system. Integration of Eq. (4) over \( k \) leads to

\[
\Gamma_{v_f K_f, v_i K_i}^{SF} = \frac{\lambda_{f} M k_{if}}{\pi} A_{if} |V_{if}(\varepsilon_{if})|^2, \quad k_{if} = k(\varepsilon_{if}).
\]

Since \( \Gamma_{v_f K_f, v_i K_i}^{SF} \) and \( \lambda_f \) are much lower (\( \sim 10^{-3} \) meV) than \( \varepsilon \), Vesman’s model can be applied and the energy-dependent resonant formation rate has the Dirac delta function profile

\[
\lambda_{v_i K_i, v_f K_f}^{SF}(\varepsilon) = 2\pi N B_{if} |V_{if}(\varepsilon)|^2 \delta(\varepsilon - \varepsilon_{if}).
\]
where $N$ is the density of deuterium nuclei in the target. According to Ref. [8] the coefficients $A_{if}$ and $B_{if}$ in the above equations are equal to

$$A_{if} = 4 W_{SF} \xi(K_i) \frac{2K_i + 1}{2K_f + 1},$$

$$B_{if} = 2 W_{SF} \frac{2S + 1}{2F + 1},$$

where

$$W_{SF} = (2F + 1) \left\{ \frac{1}{2}, 1, \frac{1}{2} \right\}^2,$$

$$\xi(K_i) = \left\{ \begin{array}{ll}
\frac{2}{3} & \text{for } K_i = 0, \\
\frac{1}{3} & \text{for } K_i = 1,
\end{array} \right.$$  \hspace{1cm} (9)

and the curly brackets stand for the Wigner $6j$ symbol. In formula (8) the usual Boltzmann factor describing the population of rotational states in a gas target is omitted because we calculate the formation rate separately for each initial rotational state. If the muonic atoms in a gas have a steady kinetic energy distribution $f(\varepsilon,T)$ at target temperature $T$, Eq. (7) can be averaged over the atom motion leading to a mean resonant rate $\lambda_{\nu_iK_i,\nu_fK_f}(T)$.

### III. RESONANT FORMATION IN A CONDENSED TARGET

Since a muonic deuterium atom can be approximately treated as a small neutron-like particle, methods used for description of neutron scattering and absorption in condensed matter are applicable in the case of formation in dense deuterium targets. Below we adapt the method developed by Lamb [26], and then generalized by Singwi and Sjölander [27] using the Van Hove formalism of the single-particle response function $S_i$ [14], for calculation of the resonant $d\mu$ formation rates.

A Hamiltonian $H_{tot}$ of a system, consisting of a $d\mu$ atom in the $1S$ state and a heavy condensed D$_2$ target, can be written down as follows

$$H_{tot} = \frac{1}{2M_{d\mu}} \nabla_{R_{d\mu}}^2 + H_{d\mu}(r_1) + H_{D_2}(q_1) + V(r_1, q_1, q_2) + H,$$  \hspace{1cm} (10)

where $M_{d\mu}$ is the $d\mu$ mass and $R_{d\mu}$ denotes the position of $d\mu$ center of mass in the coordinate frame connected with the target (see Fig. [1]). Operator $H_{d\mu}$ is the Hamiltonian of a free $d\mu$ atom, $r_1$ is $d\mu$ internal vector; $H_{D_2}$ denotes the internal Hamiltonian of a free D$_2$ molecule. It is assumed that $d\mu$ formation takes place in collision with the $l$-th D$_2$ target molecule. The position of its mass center in the target frame is denoted by $R_l$; $q_1$ is a vector connecting deuterons inside this molecule. Function $V$ stands for the potential of the $d\mu$–D$_2$ interaction [8], leading to $d\mu$ resonant formation. Vector $q_2$ connects the $d\mu$ and D$_2$ centers of mass. We neglect contributions to the potential $V$ from the molecules other than the $l$-th molecule because we assume here that distances between different molecules in the target are much greater than the D$_2$ size. The kinetic energy $\varepsilon$ of the $d\mu$ atom and its momentum $k$ in the target frame are connected by the relation

$$\varepsilon = k^2 / 2M_{d\mu}. $$  \hspace{1cm} (11)
The Hamiltonian $H$ of a pure $D_2$ target, corresponding to the initial target energy $E_0$, has the form

$$H = \sum_j \frac{1}{2M_{\text{mol}}} \nabla^2 R_j + \sum_j \sum_{j' \neq j} U_{jj'},$$

where $R_j$ is the position of $j$-th molecule center of mass in the target frame (Fig. 2), $U_{jj'}$ denotes interaction between the $j$-th and $j'$-th molecule, and $M_{\text{mol}}$ is the mass of a single target molecule.

The coordinate part $\Psi_{\text{tot}}$ of the initial wave function of the system can be written as a product

$$\Psi_{\text{tot}} = \psi_{1S}^{d\mu}(r_1) \psi_{D_2}^{K_1}(\mathbf{q}_1) \exp(i \mathbf{k} \cdot \mathbf{R}_{d\mu}) |0\rangle,$$

where $|0\rangle$ stands for the initial wave function of the condensed $D_2$ target, corresponding to the total energy $E_0$. Eigenfunctions of the operators $H_{d\mu}$ and $H_{D_2}$ are denoted by $\psi_{1S}^{d\mu}$ and $\psi_{D_2}^{K_1}$, respectively. Using the relation $\mathbf{R}_{d\mu} = \mathbf{R}_d + \mathbf{q}_2$, the wave function $\Psi_{\text{tot}}$ takes the form

$$\Psi_{\text{tot}} = \psi_{1S}^{d\mu}(r_1) \psi_{D_2}^{K_1}(\mathbf{q}_1) \exp(i \mathbf{k} \cdot \mathbf{q}_2) \exp(i \mathbf{k} \cdot \mathbf{R}_d) |0\rangle,$$

which is similar to that used in the case of $dd\mu$ formation on a single $D_2$, except the factor $\exp(i \mathbf{k} \cdot \mathbf{R}_d) |0\rangle$. This factor depends only on positions of mass centers of the target molecules.

After formation of $[(dd\mu)\nu\nu]$ complex, the total Hamiltonian of the system is well approximated by the operator $H'_{\text{tot}}$

$$H_{\text{tot}} \approx H'_{\text{tot}} = H_{d\mu}(r, \mathbf{R}) + H_C(\mathbf{q}) + V(\mathbf{q}, r, \mathbf{R}) + \tilde{H},$$

where $H_{d\mu}$ is an internal Hamiltonian of $d\mu$ molecular ion, vectors $r$ and $\mathbf{R}$ are its Jacobi coordinates. Relative motion of $d\mu$ and $d$ in the complex is described by a Hamiltonian $H_C$ which depends on the respective internal vector $\mathbf{q}$. The final Hamiltonian $\tilde{H}$ of the target, with the eigenfunction $|\tilde{n}\rangle$ and energy eigenvalue $\tilde{E}_n$, is expressed by the formula

$$\tilde{H} = \frac{1}{2M_C} \nabla^2 R_i + \sum_{j \neq i} \frac{1}{2M_{\text{mol}}} \nabla^2 R_j + \sum_j \sum_{j' \neq j} U_{jj'}$$

$$= - \left(1 - \frac{M_{\text{mol}}}{M_C}\right) \frac{1}{2M_{\text{mol}}} \nabla^2 R_i + H = \Delta H + H,$$

where $M_C$ is the mass of the complex. The respective coordinate part $\Psi'_{\text{tot}}$ of the total final wave function of the system is

$$\Psi'_{\text{tot}} = \psi_{d\mu}^{f\nu}(r, \mathbf{R}) \psi_{D_2}^{K_f}(\mathbf{q}) |\tilde{n}\rangle,$$

where $\psi_{d\mu}^{f\nu}$ and $\psi_{D_2}^{K_f}$ denote eigenfunctions of the Hamiltonians $H_{d\mu}$ and $H_C$, respectively.

The energy-dependent resonant $d\mu$ formation rate $\lambda^{SF}_{\nu K_i, \nu K_f}(\varepsilon)$ in the condensed target, for the initial $|0\rangle$ and final $|\tilde{n}\rangle$ target states and a fixed $d\mu$ total spin $F$, is calculated using the formula

$$\lambda^{SF}_{\nu K_i, \nu K_f}(\varepsilon) = 2\pi N B_{if} |A_{i0,fn}|^2 \delta(\varepsilon - \varepsilon_{if} + E_0 - \tilde{E}_n),$$

where $B_{if}$ is a resonance width and $A_{i0,fn}$ is a transition matrix element.
with the resonance condition
\[ \varepsilon + E_0 = \varepsilon_{if} + \tilde{E}_n, \]  
(19)
taking into account the initial and final energy of the target. The resonant energy for a free D\textsubscript{2} is denoted by \( \varepsilon_{if} \) and the transition matrix element is given by
\[ \mathcal{A}_{\delta_0 fn} = \langle \Psi'_{\text{tot}} | V | \Psi_{\text{tot}} \rangle. \]  
(20)
Using Eqs. (14) and (17) the matrix element (20) can be written as a product
\[ \mathcal{A}_{\delta_0 fn} = \langle \tilde{n} | \exp(i\mathbf{k} \cdot \mathbf{R}_l) | 0 \rangle V_{if}(\varepsilon) \]  
(21)
where \( V_{if}(\varepsilon) \) is the transition matrix element calculated for a single D\textsubscript{2} molecule [8]. The rate (18) can be additionally averaged over a distribution \( \rho_{n_0} \) of the initial target states at a given temperature \( T \) and summed over the final target states, which leads to
\[ \lambda^{SF}_{n_i,K_i,n_f,K_f}(\varepsilon) = 2\pi NB_{if} |V_{if}(\varepsilon)|^2 \sum_{n,n_0} \rho_{n_0} |\langle \tilde{n} | \exp(i\mathbf{k} \cdot \mathbf{R}_l | 0 \rangle|^2 \times \delta(\varepsilon - \varepsilon_{if} + E_0 - \tilde{E}_n). \]  
(22)
Factor \( B_{if} \), defined by Eqs. (8), is due to the averaging over the initial projections and summation over the final projections of spin and rovibrational quantum numbers. This factor takes also into account a symmetrization of the total wave function of \( d\mu + \text{D}_2 \) system over three deuterium nuclei.

Now we introduce a time variable \( t \) to eliminate the \( \delta \) function in the equation above and then we involve time-dependent operators, which is familiar in scattering theory (see, e.g., Refs [28, 29]). Using the Fourier expansion of the \( \delta \) function
\[ \delta(\varepsilon - \varepsilon_{if} + E_0 - \tilde{E}_n) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \exp(\varepsilon t - \varepsilon_{if} t + E_0 t - \tilde{E}_n t) \]  
(23)
one has
\[ \lambda^{SF}_{n_i,K_i,n_f,K_f}(\varepsilon) = NB_{if} |V_{if}(\varepsilon)|^2 \int_{-\infty}^{\infty} dt \exp(\varepsilon t - \varepsilon_{if} t) \sum_{n,n_0} \rho_{n_0} 
\times \langle 0 | \exp(-i\mathbf{k} \cdot \mathbf{R}_l | \tilde{n} \rangle \langle \tilde{n} | \exp(it\tilde{E}_n) \exp(i\mathbf{k} \cdot \mathbf{R}_l) \exp(-itE_0) | 0 \rangle. \]  
(24)
Assuming that the perturbation operator \( \Delta H \) is well-approximated by its mean value
\[ \Delta H \approx \langle 0 | \Delta H | 0 \rangle \equiv \Delta \varepsilon_{if} = -(1 - M_{mol}/M_C) \varepsilon_T < 0, \]  
(25)
which is valid when the target relaxation time is much smaller than the \( \text{dd} \mu \) lifetime of the order of \( 10^{-9} \) s, the matrix element in Eq. (24) can be expressed as
\[ \langle \tilde{n} | \exp(it\tilde{E}_n) \exp(i\mathbf{k} \cdot \mathbf{R}_l) \exp(-itE_0) | 0 \rangle 
= \langle \tilde{n} | \exp(it(H + \Delta H)) \exp(i\mathbf{k} \cdot \mathbf{R}_l) \exp(-itH) | 0 \rangle 
\approx \langle \tilde{n} | \exp(it\Delta \varepsilon_{if}) \exp(itH) \exp(i\mathbf{k} \cdot \mathbf{R}_l) \exp(-itH) | 0 \rangle 
= \langle \tilde{n} | \exp(it\Delta \varepsilon_{if}) \exp(i\mathbf{k} \cdot \mathbf{R}_l(t)) | 0 \rangle, \]  
(26)
where $R_l(t)$ denotes the Heisenberg operator and $E_T$ in formula (25) is the mean kinetic energy of the target molecule at temperature $T$.

Using the identity $\sum_n |\tilde{n}\rangle\langle\tilde{n}| = 1$ in Eq. (24) we obtain
\begin{equation}
\lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) = NB_{if}|V_{if}(\varepsilon)|^2 \int_{-\infty}^{\infty} dt \exp\left(-it(\varepsilon - \varepsilon'_{if})\right) \times \langle \exp\left(-i\mathbf{k} \cdot \mathbf{R}_l(0)\right) \exp\left(i\mathbf{k} \cdot \mathbf{R}_l(t)\right) \rangle_T,
\end{equation}
where $\langle \cdots \rangle_T$ denotes both the quantum mechanical and the statistical averaging at temperature $T$, and $\varepsilon'_{if}$ being the resonance energy
\begin{equation}
\varepsilon'_{if} = \varepsilon_{if} + \Delta\varepsilon_{if},
\end{equation}
shifted by $\Delta\varepsilon_{if} < 0$. Note that such a resonant energy shift was neglected in papers [26, 27], where absorption of neutrons and $\gamma$-rays by heavy nuclei were considered. An estimation of the shift in the case of $\gamma$ emission from a nucleus bound in a solid, similar to Eq. (25) was given in Ref. [30].

A self pair correlation function $G_s(\mathbf{r}, t)$ is defined by the following equation [14]
\begin{equation}
\langle \exp\left(-i\mathbf{k} \cdot \mathbf{R}_l(0)\right) \exp\left(i\mathbf{k} \cdot \mathbf{R}_l(t)\right) \rangle_T = \int d^3 r G_s(\mathbf{r}, t) \exp(i\mathbf{k} \cdot \mathbf{r}),
\end{equation}
and the single-particle response function $S_i(\mathbf{k}, \omega)$ is given by the formula
\begin{equation}
S_i(\mathbf{k}, \omega) = \frac{1}{2\pi} \int d^3 r dt G_s(\mathbf{r}, t) \exp(i(\mathbf{k} \cdot \mathbf{r} - \omega t)).
\end{equation}
Thus, by virtue of Eqs. (27) and (30), the resonant formation rate in a condensed target can by expressed in terms of the response function
\begin{equation}
\lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) = 2\pi NB_{if}|V_{if}(\varepsilon)|^2 S_i(\mathbf{k}, \omega),
\end{equation}
where the momentum transfer $\mathbf{k}$ and energy transfer $\omega$ to the target are defined as follows
\begin{equation}
\mathbf{k} = \mathbf{k}, \quad \omega = \varepsilon - \varepsilon'_{if}.
\end{equation}
The advantage of the Van Hove method is that all properties of the target, for given momentum and energy transfers, are contained in the factor $S_i(\mathbf{k}, \omega)$. It is possible to rigorously calculate $S_i$ in the case of a perfect gas and in the case of a harmonic solid. However, a liquid target or a dense gas target is a difficult problem to solve.

Proceeding as above one can obtain a similar formula for $\Gamma_{\nu_f K_f, \nu_i K_i}^{SF'}$ in a condensed target (in general, $d\mu$ spin $F'$ after back decay can be different from $d\mu$ spin $F$ before the formation)
\begin{equation}
\Gamma_{\nu_f K_f, \nu_i K_i}^{SF'} = 2\pi A_{if} \int \frac{d^3 k}{(2\pi)^3} |V_{if}(\varepsilon)|^2 \tilde{S}_i(\mathbf{k}, \omega'),
\end{equation}
where $\tilde{S}_i$ is the response function calculated for the state $|\tilde{n}\rangle$
\begin{equation}
\Delta\varepsilon_{if} \equiv \langle \tilde{n}|\Delta H|\tilde{n}\rangle = -(M_C/M_{mol} - 1) \tilde{E}_T,
\end{equation}
where $\tilde{E}_T$ denotes the mean kinetic energy of the complex bound in the target.
IV. RESONANT FORMATION IN A HARMONIC SOLID

It has been shown by Van Hove [14] that the self correlation function in the case of a gas or a solid with cubic symmetry takes the general form

\[ G_s(r, t) = \left( \frac{M_{\text{mol}}}{2\pi \gamma(t)} \right)^{3/2} \exp \left( -\frac{M_{\text{mol}}}{2\gamma(t)} r^2 \right). \]  

(35)

For a cubic Bravais lattice, in which each atom is at a center of inversion symmetry, \( \gamma(t) \) is given by the formula

\[ \gamma(t) = \int_{-\infty}^{\infty} dw \frac{Z(w)}{w} n_B(w) \exp(-iwt), \]

(36)

where \( Z(w) \) is the normalized vibrational density of states such that

\[ \int_{0}^{\infty} dw \, Z(w) = 1, \quad Z(w) = 0 \quad \text{for} \ w > w_{\text{max}}, \]

\[ Z(-w) \equiv Z(w), \]

(37)

\( n_B(w) \) is the Bose factor

\[ n_B(w) = \left[ \exp(\beta w) - 1 \right]^{-1}, \quad \beta = (k_B T)^{-1}. \]

(38)

and the Boltzmann constant is denoted by \( k_B \).

The response function (30), after substitution of Eqs. (35), (36) and integration over \( r \), can be written as follows

\[ S_i(\kappa, \omega) = \frac{1}{2\pi} \exp \left( -\frac{\kappa^2}{2M_{\text{mol}}} \gamma(\infty) \right) \times \int_{-\infty}^{\infty} dt \, \exp(-i\omega t) \exp \left( \frac{\kappa^2}{2M_{\text{mol}}} \left[ \gamma(\infty) - \gamma(t) \right] \right), \]

(39)

\( \gamma(\infty) \) denotes the limit of \( \gamma(t) \) at \( t \to \infty \). This formula can be expanded in a power series of the momentum transfer \( \kappa \), which leads to

\[ S_i(\kappa, \omega) = \exp(-2W) \left[ \delta(\omega) + \sum_{n=1}^{\infty} g_n(\omega, T) \frac{(2W)^n}{n!} \right], \]

(40)

where \( 2W \) is the Debye-Waller factor, familiar in the theory of neutron scattering,

\[ 2W = \frac{\kappa^2}{2M_{\text{mol}}} \gamma(\infty) = \frac{\kappa^2}{2M_{\text{mol}}} \int_{0}^{\infty} dw \frac{Z(w)}{w} \coth \left( \frac{\kappa}{2} \beta w \right), \]

(41)

and the functions \( g_n \) are given by

\[ g_1(w, T) = \frac{1}{\gamma(\infty)} \frac{Z(w)}{w} [n_B(w) + 1], \]

\[ g_n(w, T) = \int_{-\infty}^{\infty} dw' g_1(w - w', T) g_{n-1}(w', T), \]

\[ \int_{-\infty}^{\infty} dw g_n(w) = 1. \]

(42)
In the case of a cubic crystal structure $2W$ can also be expressed as

$$2W = \frac{1}{3} \langle 0 | u^2 | 0 \rangle \kappa^2,$$

(43)

where $u$ is the displacement of a molecule from its lattice site. Substitution of Eq. (40) to Eq. (31) leads to the following formation rate

$$\lambda_{\nu_i K_i, \nu_f K_f}^{S, E, \nu_i K_i, \nu_f K_f}(u) = 2\pi N B_{\nu f} | V_{\nu f} (u) |^2 \exp(-2W) \left[ \delta(\omega) + \sum_{n=1}^{\infty} g_n(\omega, T) \frac{(2W)^n}{n!} \right],$$

(44)

The first term in expansion (44) represents a sharp peak describing the $\delta$ profile recoilless formation. The next terms give broad distributions corresponding to subsequent multi-phonon processes. In particular, the term with $n = 1$ describes formation connected with creation or annihilation of one phonon.

If $2W \ll 1$ we deal with so-called strong binding [20] where only the few lowest terms in the above expansion are important. On the other hand, in the limit $2W \gg 1$ (weak-binding) many multi-phonon terms give comparable contributions to (44). Therefore, for sufficiently large $\kappa^2$ it is convenient to use the impulse approximation in which $\gamma(t)$ is replaced by its value near $t = 0$

$$\gamma(t) \approx \gamma(0) + it - \frac{2}{3} \mathcal{E}_T.$$

(45)

This leads to the asymptotic formula for $S_i$

$$S_i(\kappa, \omega) = \frac{1}{\Delta \sqrt{\pi}} \exp \left( - \frac{\left( \omega - \mathcal{R} \right)^2}{\Delta^2} \right),$$

(46)

where

$$\Delta = 2 \sqrt{\frac{2}{3} \mathcal{E}_T \mathcal{R}}, \quad \mathcal{R} = \frac{\kappa^2}{2M_{\text{mol}}}. \quad (47)$$

The mean kinetic energy $\mathcal{E}_T$ of a molecule in the solid, which also determines the resonance energy shift (25), is equal to

$$\mathcal{E}_T = \frac{3}{2} \int_0^\infty dw Z(w) w \left[ n_B(w) + \frac{1}{2} \right].$$

(48)

The energy $\mathcal{E}_T$ contains a contribution from the zero-point vibrations and it approaches $3k_B T/2$ only at high temperatures $T \gg w_{\text{max}}/k_B$. Function (16) is a Gaussian with response centered at the recoil energy $\mathcal{R}$. Therefore in the weak binding region the resonant formation rate takes the Doppler form obtained by Bethe and Placzek [1] for resonant absorption of neutrons in gas targets [31]. However, the resonance width (17) in the solid at temperature $T$ is different from the Doppler width in a Maxwellian gas $\Delta_{\text{gas}} = 2 \sqrt{k_B T \mathcal{R}}$ unless the temperature is sufficiently high. This phenomenon was pointed out by Lamb in his paper [26] concerning resonant neutron absorption in solid crystals. By virtue of the equations above one can introduce for the solid an effective temperature $T_{\text{eff}}$

$$T_{\text{eff}} = \frac{2}{3} \mathcal{E}_T / k_B.$$

(49)

---

1 In fact, formula (16) is the limit of the Bethe formula in the case of a very narrow natural resonance width $\Gamma \rightarrow 0$. 

9
V. RESONANT FORMATION IN FROZEN DEUTERIUM

The following considerations concern the solid deuterium crystals used in the TRIUMF experiments [32, 33], though the results presented below can be applied to targets obtained in similar conditions [12, 34]. At TRIUMF thin solid deuterium layers have been formed by rapid freezing of gaseous D$_2$ on gold foils at $T = 3$ K and zero pressure. According to Ref. [35] such deuterium layers have the face-centered cubic (fcc) polycrystalline structure. Since the distance between the neighboring molecules is a few times greater than the diameter of a D$_2$ molecule and the Van der Waals force that binds the solid is weak, one can neglect perturbations of the resonant formation potential $V$ due to these neighbors.

The deuterium crystals at zero pressure are quantum molecular crystals. The amplitude of zero-point vibration at 3 K equals 15% of the nearest neighbor distance. A single-particle potential in this case is not harmonic and the standard lattice dynamics leads to imaginary phonon frequencies. However, the standard dynamics can be applied after a renormalization of the interaction potential, taking into account the short-range pair correlations between movement of the neighbors [35]. In result, the theoretical calculations [36] of the phonon dispersion relations give a good agreement with the neutron scattering experiments [37] and the Debye model for solid deuterium can be used as a good approximation of the phonon energy distribution

$$Z(w) = \begin{cases} \frac{3}{w_D^2} w_D^3 & \text{if } w \leq w_D, \\ 0 & \text{if } w > w_D, \end{cases}$$

with the Debye energy $w_D = k_B \Theta_D$ and Debye temperature $\Theta_D$ taken from the neutron experiments. For $T = 3$ K we use the Debye model of an isotropic solid with $\Theta_D = 108$ K corresponding to the maximal phonon energy $w_D = 9.3$ meV. Thus, we are dealing with the limit $T/\Theta_D \ll 1$ where

$$\gamma(\infty) = \frac{3}{2} w_D^{-1}, \quad \mathcal{E}_T = \frac{9}{16} w_D \approx 5.2 \text{ meV}, \quad T_{\text{eff}} = \frac{3}{8} \Theta_D \approx 40 \text{ K},$$

are very good approximations of Eqs. (41), (48) and (49). The Debye-Waller factor and mean kinetic energy $\mathcal{E}_T$ at lowest temperatures are determined by contributions from the zero-point D$_2$ vibration in the lattice, and therefore these quantities do not tend to zero at $T \to 0$. The zero-point energy is not accessible energy, but its effects are always present.

The values of the resonance energies depend on initial and final rovibrational quantum numbers of the system. In solid hydrogens at low pressures these quantum numbers remain good quantum numbers, but excited energy levels broaden into energy bands (rotons and vibrons) due to coupling between neighboring molecules [18]. The calculations presented in the literature concern pure solid H$_2$, HD and D$_2$ targets and only lowest quantum numbers. The problem of a heavier impurity, such as $(dd\mu)d$ complex in D$_2$, has not been considered yet. However, knowing that the width of the rotational bands can reach about 1 meV [18], a possible influence of this effect on the calculated formation rates and fusion neutron time spectra is discussed in the next section.

At low temperatures all D$_2$ molecules are in the ground vibrational state $\nu_i = 0$ and $dd\mu$ is formed via the excitation of the complex to the state $\nu_f = 7$. Unless a catalyst is applied, rapidly frozen deuterium is a mixture of ortho-D$_2$ ($K_i = 0$) and para-D$_2$ ($K_i = 1$). In the TRIUMF experiments gaseous deuterium was pumped through a hot palladium filter before freezing. Therefore the solid target was a statistical mixture (2:1) of the ortho- and para-states ($K_i = \text{stat}$). Since the para-ortho relaxation without a catalyst is very slow (0.06%/h)
in solid deuterium \[38\], the population of these states is not changed during experiments of a few days.

The lowest resonance energies \(\varepsilon_{if}\) and \(\varepsilon'_{if}\), for fixed \(\nu_i, \nu_f\) and different values of \(F, K_i, S\) and \(K_f\) are shown in Table \[11\]. A few of them have negative values, which means that to satisfy the resonance condition \(\varepsilon = \varepsilon_{if}\) an energy excess in the \(d\mu+D_2\) system should be transferred to external degrees of freedom. This is possible in dense targets, where energy of neighboring molecules can be increased. Such an effect, due to triple collisions in gas targets, has been firstly discussed in Ref. \[40\]. In a solid, the energy excess is lost through incoherent phonon creation. According to \(25\), \(28\), and \(51\), in the considered 3 K solid deuterium all resonant energies \(\varepsilon'_{if}\) are shifted by \(\Delta \varepsilon_{if} \approx -1.81\) meV. One can see that all resonances for \(F = \frac{1}{2}\) are placed at higher energies, which is caused by \(d\mu\) hyperfine splitting \(\Delta E_{\text{hfs}} = 48.5\) meV. All resonance energies \(\varepsilon'_{if} \leq w_D \approx 10\) meV are connected with formation from the upper spin state \(F = \frac{3}{2}\) of \(d\mu\). However, only resonances corresponding to the dipole transitions \(K_i = 0 \rightarrow K_f = 1\) and \(K_i = 1 \rightarrow K_f = 0, 2\) can give a significant contribution to the formation rate at lowest energies. Other transition matrix elements described in Ref. \[40\] tend to zero when \(\varepsilon \rightarrow 0\) (see Figs. \[3\] and \[4\] obtained for \(K_i = 0\) and \(K_i = 1\)).

The low energy rates \((\varepsilon \lesssim w_D)\) are calculated using formula (44) with a few most significant terms of the response function expansion (41) taken into account. Fig. \[2\] shows the function \(S_i(\kappa, \varepsilon, \varepsilon'_{if})\) corresponding to the two dipole transitions in \(\text{para-}D_2\). The sub-threshold resonance, with \(\varepsilon'_{if} \approx -9.0\) meV, gives contributions to the formation rate only through the phonon creation processes. For \(\varepsilon'_{if} \approx 1.6\) meV, the non-phonon process is possible and it is represented by a vertical line. Different peaks in this figure describe processes connected with different numbers of created phonons. In particular, one-phonon processes, which are proportional to \(Z(w)\) with the characteristic Debye cutoff, can be clearly distinguished. Since the \(n\)-phonon term in (41) is proportional to \(\kappa^{2n}\), the \(dd\mu\) formation rate tends to zero at \(\varepsilon \rightarrow 0\). Note that the phonon annihilation gives negligible contribution to the rate at very low target temperatures \(T \ll \Theta_D\).

In order to compare the calculated formation rates with experiments the summed rates \(\lambda_{K_i}^F(\varepsilon)\) are introduced

\[
\lambda_{K_i}^F(\varepsilon) = \sum_{K_f, S} \lambda_{\nu_i K_i, \nu_f K_f}^{SF}, \quad \nu_i = 0, \quad \nu_f = 7. \tag{52}
\]

In Fig. \[3\] the formation rates \(\lambda_{K_i}^F(\varepsilon)\) in the solid ortho-\(D_2\) and para-\(D_2\) are shown for \(F = \frac{3}{2}\). In the case of resonances satisfying the condition \(\varepsilon'_{if} \leq w_D\) we have \(2W < 1\) and the expansion (44) is used. The two strong peaks represent the recoilless formation process, without phonon excitations. The delta function profile of every peak is shown as a rectangle with a height equal to the formation rate strength divided by the total decay width \((\approx 0.8 \times 10^{-3}\) meV). The strength defined as the value of the factor standing before \(\delta(\omega)\) in the expansion (44), is equal to 0.1061 eV\(\cdot\)µs\(^{-1}\) for the resonance \(K_i = 0 \rightarrow K_f = 1\) in solid ortho-\(D_2\). The transition \(K_i = 1 \rightarrow K_f = 2\) in para-\(D_2\) gives 0.07544 eV\(\cdot\)µs\(^{-1}\) as the resonance strength. Higher resonance energies involve many multi-phonon terms and therefore we use the asymptotic form (46) of \(S_i\) for \(\varepsilon'_{if} > w_D\). All formation rates presented in the figures are normalized to the liquid hydrogen density \(N_0 = 4.25 \times 10^{22}\) atoms/cm\(^3\).

Though in Monte Carlo simulations, involving energy-dependent rates of different processes, the “absolute” formation rates \(\lambda_{K_i}^F(\varepsilon)\) should be used, it is convenient to introduce an
effective formation rate $\bar{\lambda}_{K_f}^F(\varepsilon)$ which leads to the nuclear $dd$ fusion in \([(dd\mu)\text{dec}]\) complex. Back decay of the complex to the $d\mu+D_2$ system, characterized by the quantum numbers $K_f'$ and $F'$, strongly influences the fusion process because the back-decay rates are comparable with the effective fusion rate $\bar{\lambda}_f \approx 374 \text{ }\mu s^{-1}$ [7]. Since in a solid target rotational deexcitation of the asymmetric complex is much faster than back decay and fusion, it is assumed that back decay takes place only from the state $K_f = 0$. The effective formation rate is then defined by the following formula

$$\bar{\lambda}_{K_i}^F(\varepsilon) = \sum_{K_f,S} \lambda_{\nu_f,K_i\nu_f,K_f}^{SF',F}(\varepsilon) \mathcal{P}_S^{\text{fus}}, \quad \nu_i = 0, \nu_f = 7,$$  \hspace{1cm} (53)

where the fusion fraction $\mathcal{P}_S^{\text{fus}}$ is given by

$$\mathcal{P}_S^{\text{fus}} = \frac{\bar{\lambda}_f}{\Gamma_S}, \quad \Gamma_S = \bar{\lambda}_f + \sum_{F'} \Gamma_i^{SF'}, \quad \Gamma_i^{SF'} = \sum_{K_i',K_f=0} \Gamma_{\nu_f,K_i',\nu_f,K_i}'^{SF'}.$$

Since the frequency of lattice vibrations ($\sim \omega_d/\hbar \sim 10^7 \text{ }\mu s^{-1}$) is many orders of magnitude greater than the back-decay and fusion rates, energetic phonons created during the $dd\mu$ formation process are dissipated. At 3 K the number of phonons with energies $w \gtrsim k_B T \approx 0.26$ meV is strongly suppressed by the Bose factor $n_w(w)$. Therefore back decay with phonon annihilation at $T \ll \Theta_D$ is negligible. In particular, the phonon channel of decay of $dd\mu$, formed from $d\mu$ state $F = \frac{3}{2}$ due to the subthreshold resonances, is closed because this would require an annihilation of a phonon with energy of a few meV. In this case back decay is connected with the spin-flip transition to $F' = \frac{1}{2}$. Since the corresponding energy release of a few tens of meV is much greater than the Debye energy ($\Delta E^{\text{hfs}} \gg \omega_D$), the $dd\mu$ decay rate is dominated by contributions from simultaneous phonon creation processes.

After integration of formula (33) over direction of vector $k$ one obtains

$$\Gamma_{\nu_f,K_i',\nu_f,K_i}^{SF'} = \frac{A_{ij}}{\pi} \int_0^\infty dk k^2 |V_{ij}(\varepsilon)|^2 \tilde{S}_i(k^2,\omega'),$$

and then substitution of expansion (10) and integration of the recoiless term lead to

$$\Gamma_{\nu_f,K_i',\nu_f,K_i}^{SF'} = \frac{A_{ij}}{\pi} \left[ M\tilde{k}_{ij} |V_{ij}(\varepsilon)|^2 \exp(-2\tilde{W}_{ij}) + \sum_{n=1}^\infty \int_0^\infty dk k^2 |V_{ij}(\varepsilon)|^2 \exp(-2\tilde{W}) g_n(\omega',T) \frac{(2\tilde{W})^n}{n!} \right],$$

where

$$2\tilde{W} = \frac{k^2}{2M_C} \gamma(\infty), \quad 2\tilde{W}_{ij} = 2\tilde{W}(\tilde{k}_{ij}), \quad \tilde{k}_{ij} = \sqrt{2M\varepsilon_{ij}'}. \hspace{1cm} (57)$$

It is assumed in the formula above that the phonon energy spectrum of solid deuterium containing \([(dd\mu)\text{dec}]\) is similar to that of a pure deuterium lattice. The problem of lattice dynamics of a quantum solid deuterium crystal containing a small admixture of a heavier isotope has not been considered yet in literature, at least to the knowledge of the authors. However, this approximation is reasonable since the Debye temperatures of solid hydrogen
and deuterium at 3 K are very similar [35], independently of the mass difference of these isotopes. Therefore it is assumed that during the $dd\mu$ lifetime the mean kinetic energy $\tilde{E}_T$ of the complex reaches the energy $E_T$ characterizing a pure deuterium solid. Thus the resonance energy shift (34) is approximated by

$$\Delta \tilde{\varepsilon}_{if} \approx - \left( M_C / M_{\text{mol}} - 1 \right) E_T \approx -2.77 \text{ meV},$$

which gives $\varepsilon'_{if} = \varepsilon_{if} - 2.77 \text{ meV}$.

The effective formation rates in 3 K solid deuterium for $F = 3/2$ are shown in Fig. 7. The phonon part of the rates below a few meV is about two orders of magnitude lower than the average rate of $2.7 \mu s^{-1}$ derived from the experiment [11, 13]. This means that at $\varepsilon \ll w_D$ the phonon contribution to the total resonant formation rate is even smaller than the non resonant $dd\mu$ formation rate $\lambda_{nr} \approx 0.44 \mu s^{-1}$ [9], and that the estimation of the phonon contribution given in Ref. [20] is strongly overestimated. Therefore, the experimental results can only be explained by resonant $dd\mu$ formation at energies $\varepsilon \gtrsim 1 \text{ meV}$, where the rate exceeds significantly the value of $1 \mu s^{-1}$. A cusp at 0.3 meV in para-$D_2$ is due to the formation with simultaneous one-phonon creation, connected with the subthreshold resonance $K_i = 1 \to K_f = 0$. This implies a significant difference between the resonant formation in ortho-$D_2$ and para-$D_2$ below 1 meV. However, this difference is difficult to measure because of a broad distribution of $d\mu$ energy. Note that a similar subthreshold phonon effect in the case of resonant $dt\mu$ formation in solid deuterium has been discussed in Ref [41].

In the solid target the fusion fraction $P_{\text{fus}}^{S} \approx 0.3$ and the total resonance width $\Gamma^S \approx 0.8 \times 10^{-3} \text{ meV}$ for both $S = 1/2$ and $S = 3/2$. The back-decay rate $\Gamma^{SF'}$ from $S = 1/2$ to $F' = 1/2$ equals about $843 \mu s^{-1}$. Decay $S = 1/2 \to F' = 1/2$ is impossible. In the case of $S = 3/2$ we have obtained $\Gamma^{SF'} \approx 281 \mu s^{-1}$ for $F' = 1/2$ and $\Gamma^{SF'} \approx 610 \mu s^{-1}$ for $F' = 3/2$. Phonon creation processes give dominant contributions to the back-decay rates, e.g., the non-phonon part of $\Gamma^{SF'}$, given by the first term of expansion (56), equals $169 \mu s^{-1}$. Therefore the $d\mu$ energy spectrum, after back decay in the solid, is not discrete.

In Fig. 8 the effective rates in solid deuterium for $F = 1/2$ are presented. For the sake of comparison the formation rate for 3 K ortho-$D_2$ gas is also plotted. The “gas” curve has been calculated using the asymptotic formula (45) for $S_i$ with $T_{\text{eff}} = 3 \text{ K}$. This figure shows that in a real solid deuterium target the rates are smeared much more than in a gas target with the same temperature, because of the zero-point vibrations. Therefore even at relatively high $d\mu$ energies of some 0.1 eV one should not neglect the solid effects and use the formation rates calculated for a 3 K Maxwellian gas.

VI. MONTE CARLO CALCULATIONS

The calculated energy-dependent $dd\mu$ formation rates have been applied in our Monte Carlo simulations of $\mu$CF in 3 K solid deuterium targets. The final $d\mu$ energy distribution after back decay, including simultaneous phonon creation processes, has been determined through a numerical integration of Eq. (56). The calculated distribution is shown in Fig. 9 for $S = 1/2$, $K_f = 0$ and $F' = 1/2$. The rotational transitions to $K'_i = 0, 1, 2$ with no phonon creation are seen as the delta peaks. The continuous energy spectrum describes phonon creation contribution to $d\mu$ energy. Note that, opposite to $dd\mu$ formation rates, this phonon
contribution (for a given rotational transition peak) extends towards lower energies. The average $d\mu$ energy after $dd\mu$ back decay equals about 30 meV, for the presented spectrum.

The $dd$ fusion neutron and proton spectra depend on the time evolution of $d\mu$ energy. This energy is determined by differential cross sections of different scattering processes of $d\mu$ atoms in a given solid target, including elastic scattering, rovibrational transitions, spin-flip reactions and phonon processes. The scattering cross sections in a solid are calculated using the Van Hove method. Some results of such calculations for $d\mu$ atoms in fcc solid deuterium have been presented and discussed in Ref. \[42\].

The $dd\mu$ fusion neutron and proton spectra depend on the time evolution of $d\mu$ energy. This energy is determined by differential cross sections of different scattering processes of $d\mu$ atoms in a given solid target, including elastic scattering, rovibrational transitions, spin-flip reactions and phonon processes. The scattering cross sections in a solid are calculated using the Van Hove method. Some results of such calculations for $d\mu$ atoms in fcc solid deuterium have been presented and discussed in Ref. \[42\].

In Fig. 10 is shown the total cross section for $d\mu(F = \frac{3}{2})$ scattering in the statistical mixture of 3 K solid ortho-D$_2$ and para-D$_2$. Bragg scattering, with the Bragg cutoff at $\varepsilon_B = 1.1$ meV, and incoherent elastic scattering do not change $d\mu$ energy because of the very large mass of the considered solid target. Below 1.7 meV the $d\mu$ atom is effectively accelerated, mainly due to the rotational deexcitation of para-D$_2$ molecules \[21, 42\]. This transition is enabled by muon exchange between deuterons in $d\mu+D_2$ scattering. The curve “0 → 1” in Fig. 10 describing the rotational deexcitation, includes contributions from simultaneous incoherent phonon processes. This cross section at $\varepsilon = 2.5$ meV equals $0.22 \times 10^{-20}$ cm$^2$, which is about three times less (taking into account the statistical factor of 1/3 for $K = 1$ states) than the estimation given in paper \[21\]. Phonon annihilation is a much weaker $d\mu$ acceleration mechanism than the rotational deexcitation.

Since the coherent amplitude for $d\mu$ elastic scattering on a single D$_2$ molecule is greater by two orders of magnitude than the incoherent amplitude, the coherent processes involving conservation of momentum dominate low energy $d\mu$ scattering in solid deuterium. It is especially important below a few meV, where the shapes of coherent and incoherent cross sections differ strongly. The small phonon creation cross section below 1.1 meV, leading to $d\mu$ energy loss, is due to the incoherent amplitude. Coherent phonon creation is impossible below $\varepsilon_B$. This limit is obtained in the case of coherent one-phonon creation process, for the total momentum conservation involving the smallest (non-zero) inverse lattice vector $\tau$, which also fixes the position of the first peak of the Bragg scattering at $\varepsilon_B = 1.1$ meV. For $\tau = 0$ one-phonon creation is possible only if the $d\mu$ velocity is not lower than the sound velocity in the crystal, which is well-known in neutron physics. According to Ref. \[38\] the mean sound velocity in solid deuterium equals about $1.2 \times 10^5$ cm/s and this corresponds to $d\mu$ energy of 15 meV. Therefore, neglecting the inverse lattice contribution to the one-phonon creation cross section in Ref. \[21\] leads to the severe underestimation of $d\mu$ slowing down at lowest energies and subsequent overestimation of $d\mu$ kinetic energy.

Above 1.7 meV phonon creation already prevails over all acceleration processes. However, the effective deceleration rate below $w_D$ is strongly suppressed by the dominating Bragg elastic scattering. At energies above some 10 meV subsequent rotational and then vibrational excitations of D$_2$ molecules become important and they provide a very fast mechanism of $d\mu$ deceleration at higher energies.

The total cross section for $d\mu(F = \frac{3}{2})$ scattering in a pure 3 K ortho-D$_2$ target (see Fig. 11) is quite similar to that shown in Fig. 10. A significant difference is the lack of rotational deexcitation. Therefore phonon annihilation is the only, and weak, acceleration mechanism. It dominates the inelastic cross section below 1.4 meV.

Fig. 12 presents the time evolution of average $d\mu(F = \frac{3}{2})$ atom energy $\varepsilon_{avg}$, obtained from
our Monte Carlo calculations. It is assumed that the target is infinite and that \( d\mu \) atoms have initially a Maxwellian energy distribution with a mean energy of 1 eV. A statistical initial population of \( d\mu \) total spin is used and the theoretical non-resonant part of the total spin-flip rate \( \lambda_{3\rightarrow 1/2} \) is multiplied by a single scaling factor of 0.4, in order to keep agreement with the experimental values [11, 13] of the spin-flip rate. The calculations have been performed for ortho-D\(_2\), para-D\(_2\) and their statistical mixture (stat). One can see that \( d\mu \) mean energy of 10 meV is reached already after 5 ns. Then, below the Debye energy, deceleration become very slow. The lowest value of \( \varepsilon_{\text{avg}} \) is determined by the intersection point of the cross sections of the acceleration processes and phonon creation process. In the case of a statistical mixture \( \varepsilon_{\text{avg}} \approx 1.7 \) meV, for \( K = 0 \) we have \( \varepsilon_{\text{avg}} \approx 1.4 \) meV. Finally, for pure para-D\(_2\), with a contribution to the total cross section from the rotational transition \( K = 1 \rightarrow 0 \) three times greater than that shown in Fig. 10, \( \varepsilon_{\text{avg}} \approx 2.2 \) meV. Thus, \( d\mu \) atoms are never thermalized and their energy is significantly greater than 1 meV. For para-D\(_2\) the mean energy is always greater than the energy of the lowest resonance peak \( \varepsilon'_{i_f} = 1.6 \) meV. However, even if \( \varepsilon_{\text{avg}} \) is smaller than \( \varepsilon'_{i_f} \), a significant part of \( d\mu \) atoms has energy \( \varepsilon \geq \varepsilon'_{i_f} \) because of a large admixture of hot \( d\mu \) atoms at \( t = 0 \) [13, 16] and slow deceleration below 10 meV.

Since at energies of a few meV the lowest delta peaks are dominant in the resonant formation, their contributions to the mean effective formation rate are shown in Fig. 13 for gas and solid deuterium (stat) targets, assuming steady Maxwell distributions of \( d\mu(F = \frac{3}{2}) \) energy, with different \( \varepsilon_{\text{avg}} \). The maximum average rate of about 6 \( \mu s^{-1} \) in the solid is due to the resonance energy shift of \(-1.8 \) meV. The experimental result of 3 \( \mu s^{-1} \) can be explained because \( \varepsilon_{\text{avg}} \) is greater than 1 meV. However, in order to obtain large fusion neutron and proton yields through resonant \( dd\mu \) formation, the width \( \Gamma^S \) of the resonance peaks in solid can not be too narrow. The peak resonant rates of a few \( 10^4 \mu s^{-1} \) have been obtained assuming the discrete values of the rovibrational D\(_2\) energies in solid deuterium and \( \Gamma^S \sim 10^{-3} \) meV. These resonant rates are many orders of magnitude greater than the inelastic scattering rate \( \sim 10 \mu s^{-1} \). In such a case \( d\mu \) atoms are very quickly (compared to \( d\mu(F = \frac{3}{2}) \) lifetime) removed from the regions of resonance peaks and the contribution of the recoilless resonances to the neutron yield is negligible. The Monte Carlo simulations have shown that the neutron yield from the phonon part of the resonant rates gives only some 10% of the yield observed in the experiments. In result, the calculated time spectra, obtained for the small \( \Gamma^S \), are dominated by weak non-resonant \( dd\mu \) formation, which disagrees with the experimental data. Therefore, we have investigated influence of a broadening of the non-phonon resonant peaks, due to the presence of molecular rovibrational bands in solid, discussed in Ref. [16]. Since in the literature there is no information concerning the profile of such bands, we have assumed a rectangular shape of the resonance peaks. The resonance strengths have been fixed and their widths have been changed in the limits 0.001–1 meV. It turns out that good Monte Carlo results are obtained for \( \Gamma^S \approx 0.5 \) meV, which is consistent with the rotational bandwidths of about 1 meV reported in Ref. [18]. This gives the resonant formation rate of 294 \( \mu s^{-1} \) for the recoilless peak in ortho-D\(_2\), and respectively 214 \( \mu s^{-1} \) in para-D\(_2\). In Fig. 14 one sees the resonant formation rate at lowest energies for \( \Gamma^S = 0.5 \) meV and for the statistical mixture of ortho- and para-states. Also shown is the Monte Carlo distribution of \( d\mu(F = \frac{3}{2}) \) energy, calculated for times \( t = 10 \) ns and \( t = 30 \) ns. The Maxwell distribution of initial \( d\mu \) energy, with \( \varepsilon_{\text{avg}} = 1 \) eV, has been assumed. Two minima in the \( d\mu \) energy distribution appear quickly at the positions of the resonance peaks since the respective \( dd\mu \) formation rates are comparable with the total inelastic scattering rate of
about 30 $\mu$s$^{-1}$.

The $dd$ fusion neutron spectrum, calculated assuming the same initial $d\mu$ energy and resonance profiles, is shown in Fig. [12]. A $3.2 \times 10^{-6}$ concentration of nitrogen is included in order to fit the TRIUMF target conditions. The solid line plotted in this figure has been calculated using the steady-state kinetics model with the effective formation rate $\lambda_{stat}^{3/2} = 3 \mu s^{-1}$ and total spin-flip rate $\lambda_{Z+} = 36 \mu s^{-1}$ taken from the fits to the experimental data [13]. The slope of the spectrum at $t \lesssim 80$ ns is determined by the rates $\lambda_{stat}^{3/2}$, $\lambda_{Z+}$, and $d\mu$ scattering rate which also changes the population of $d\mu(F = \frac{3}{2})$ atoms in the vicinity of the resonant peaks. The steady-state kinetics model does not include the process of $d\mu$ deceleration. Therefore, fits using this model could entangle the deceleration rate with the formation and spin-flip rates. The mean formation rate, calculated directly in the Monte Carlo runs, is a function of time, and it stays at the level of 1–3 $\mu s^{-1}$. The spectrum slope at large times $t \gtrsim 100$ ns, when $d\mu(F = \frac{3}{2})$ atoms practically disappear, are due to the nonresonant $dd\mu$ formation from $F = \frac{1}{2}$ and to the muon transfer to nitrogen contamination.

The shape of the time spectra practically does not change when the mean energy $\varepsilon_{avg}$ of the initial single Maxwell distribution varies in the limits 0.01–1 eV. On the other hand, the spectra change strongly if a significant part of $d\mu$ atoms at $t = 0$ has energy smaller than the energy of the lowest resonant peak, which can be observed using a more complicated (e.g. two-Maxwell distribution). Assuming that $\Gamma^S$ is greater than 0.5 meV we obtain results which begin to differ significantly from the analytical curve calculated with the experimental parameters. In particular, the ratios of neutron yields from the short and large times begin to disagree. Fits of the calculated spectra to the experimental data would enable a better determining of $\Gamma^S$ and a shape of the initial $d\mu$ energy. However, this is not the purpose of this work. A qualitative comparison of Monte Carlo spectra with the experimental data has already been performed in article [13]. In this case good fits were not obtained since at that time the resonant $dd\mu$ formation rates in solid $D_2$ and $d\mu$ scattering rates including coherent effects in the solid were not yet available.

Our calculations show that strong resonant $dd\mu$ formation takes place both in ortho-$D_2$ and para-$D_2$. There are certain differences between the neutron time spectra from these targets (see Fig. [12]), caused by the different positions and strengths of the lowest resonance peaks. Also $d\mu$ slowing down process differs slightly in the two cases. The neutron yield at larger times is smaller for ortho-$D_2$ since in this case the resonance peak is placed at higher energy of 2.3 meV. Therefore, $d\mu$ atoms are removed faster from the peak compared to the situation in para-$D_2$, where the resonance is observed at 1.6 meV. A greater mean $d\mu$ energy in para-$D_2$ (cf. Fig. [12]) leads also to a stronger overlap of the resonance peak and $d\mu$ energy distribution at $t \gtrsim 20$ ns. However, the differences between the spectra can be clearly seen only in high-statistics experiments.

VII. CONCLUSIONS

The methods used for description of resonant neutron and $\gamma$-ray absorption in condensed matter have been directly applied for calculation of resonant $dd\mu$ formation and back-decay rates in condensed deuterium targets. These rates are expressed in terms of the Van Hove single-particle function, which depends on properties of a given target. In particular, we have derived the analytical formulas for the rate in the case of resonant $dd\mu$ formation in a harmonic solid deuterium. The calculations show great differences between resonant $dd\mu$
formation in 3 K solid deuterium and in 3 K D₂ gas. In solid, the formation at a few meV, which determines the experimental results, is dominated by presence of the strong recoilless resonant peaks. On the other hand, the formation with simultaneous phonon creation is important above the Debye energy. The resonance profiles in the solid at higher energies are similar to that in D₂ gas, but with the effective temperature equal to 40 K. This temperature is determined by the energy of zero-point vibration of D₂ molecule in the lattice. Phonon creation is always important in the case of ddμ back decay because it is connected with energy release of a few tens meV, which is much greater than the Debye energy.

A condition \( T/\Theta_D \ll 1 \) is fulfilled for any solid deuterium target at low pressure. Therefore, the parameters determining solid state effects (Debye-Waller factor, mean energy of D₂ vibration in solid) weakly depend on target temperature \( T \). They are expressed in terms of the Debye energy \( w_D \) which does not significantly change with the varying solid temperature \( T \). In result, the resonant ddμ formation rates in solid deuterium for different \( T \) are very similar and one may expect that the average formation rates, derived from measurements performed at different temperatures, will also be very close. This is confirmed by the results of experiments carried out at TRIUMF and at JINR.

The structure of a solid deuterium target depends on its temperature and history. Targets maintained at \( T \gtrsim 4 \) K have the hcp structure [35]. Though our calculations have been performed for fcc crystals, the obtained results are also good approximations of the resonant rates in hcp polycrystals since the Debye temperature and nearest neighbor distance are similar for these two lattices. In general, the formulas derived in this paper can be used in a wide range of target temperature and density, with appropriate experimental values of the Debye temperature and lattice constant taken into account.

The Monte Carlo calculations show that dμ deceleration below the Debye energy is very slow and that mean energy of dμ(\( F = \frac{3}{2} \)) atom is always significantly greater than 1 meV. The energy distribution of dμ’s during their lifetime is very broad (at least a few meV), therefore a strong overlap of this distribution and lowest resonance peaks takes place, leading to a large mean ddμ formation rate in solid deuterium. However, explanation of the experiments is possible only if the broadening of rovibrational molecular levels in solid is taken into account. We obtained reasonable results assuming that the strengths of the recoilless resonant peaks are constant and that the rotational bands increase the resonance peak width to 0.5 meV. Note that, according to Ref. [18], high pressures lead to a greater broadening and even to a mixing of rotational states. This could complicate a comparison of theory and high-pressure experiments. The phonon part of the resonant rate give only about 10% contribution to the calculated neutron time spectra.

The dd fusion neutron spectra calculated for ortho-D₂ and para-D₂ solid targets are quite similar. Small differences between the spectra are due to the different energies and strengths of the lowest resonant peaks, and to a slightly higher mean dμ energy in para-D₂. These differences can be clearly seen only in high-statistics experiments. Our calculations do not confirm a lack of strong resonant ddμ formation in solid ortho-D₂, predicted in the papers [20, 21]. In order to verify the theory it is necessary to perform measurements in pure ortho-D₂ and para-D₂ solid targets under the same conditions.

Acknowledgments

We wish to thank L. I. Ponomarev for stimulating discussions. We are grateful to G. M. Marshall for a critical reading of the manuscript. This work was supported in part
through Grant INTAS 97-11032.

[1] V. P. Dzelepov et al., Zh. Eksp. Teor. Fiz. 50, 1235 (1966) [Sov. Phys. JETP 23, 820 (1966)].
[2] W. H. Breunlich et al., Annu. Rev. Nucl. Part. Sci. 39, 311 (1989).
[3] L. I. Ponomarev, Contemp. Phys. 31, 219 (1990).
[4] E. A. Vesman, Zh. Eksp. Teor. Fiz. Pisma 5, 113 (1967) [Sov. Phys. JETP Letters 5, 91 (1967)].
[5] L. I. Ponomarev and M. P. Faifman, Zh. Eksp. Teor. Fiz. 71, 1689 (1976) [Sov. Phys. JETP 44, 886 (1976)].
[6] L. I. Menshikov and M. P. Faifman, Yad. Fiz. 43, 650 (1986) [Sov. J. Nucl. Phys. 43, 414 (1986)].
[7] L. I. Menshikov et al., Zh. Eksp. Teor. Fiz. 92, 1173 (1987) [Sov. Phys. JETP 65, 656 (1987)].
[8] M. P. Faifman, L. I. Menshikov, and T. A. Strizh, Muon Catalyzed Fusion 4, 1 (1989).
[9] A. Scrinzi et al., Phys. Rev. A47, 4691 (1993).
[10] C. Petitjean et al., Hyp. Interact. 118, 127 (1999).
[11] P. E. Knowles et al., Hyp. Interact. 101/102, 21 (1996).
[12] D. L. Demin et al., Hyp. Interact. 101/102, 13 (1996).
[13] P. E. Knowles et al., Phys. Rev. A56, 1970 (1997).
[14] L. Van Hove, Phys. Rev. 95, 249 (1954).
[15] V. E. Markushin, Phys. Rev. A50, 1137 (1994).
[16] D. J. Abbott et al., Phys. Rev. A55, 214 (1997).
[17] A. Adamczak, Hyp. Interact. 101/102, 113 (1996).
[18] J. Van Kranendonk, Solid Hydrogen (Plenum Press, New York and London, 1983).
[19] V. V. Filchenkov, Hyp. Interact. 101/102, 37 (1996).
[20] L. I. Menshikov and V. V. Filchenkov, Hyp. Interact. 101/102, 207 (1996).
[21] C. L. Gurin and L. I. Menshikov, Hyp. Interact. 118, 147 (1999).
[22] V. N. Ostrovski and V. I. Ustimov, Zh. Eksp. Teor. Fiz. 79, 1228 (1980) [Sov. Phys. JETP 52, 620 (1980)].
[23] A. M. Lane, Phys. Lett. A98, 337 (1983).
[24] A. Gula, A. Adamczak, and M. Bubak, Phys. Lett. A109, 224 (1985).
[25] N. T. Padial et al., Phys. Rev. A37, 329 (1988).
[26] W. E. Lamb, Phys. Rev. 55, 190 (1939).
[27] K. S. Singwi and A. Sjöland, Phys. Rev. 120, 1093 (1960).
[28] A. Akhiezer and I. Pomeranchuk, Zh. Eksp. Teor. Fiz. 17, 769 (1947) [Sov. Phys. JETP 11, 167 (1947)].
[29] G. C. Wick, Phys. Rev. 94, 1228 (1954).
[30] B. D. Josephson, Phys. Rev. Lett. 4, 341 (1960).
[31] H. Bethe and G. Placzek, Phys. Rev. 51, 462 (1937).
[32] G. M. Marshall et al., Hyp. Interact. 82, 529 (1993).
[33] P. E. Knowles et al., Nucl. Instrum. Methods A368, 604 (1996).
[34] P. Strasser et al., Phys. Lett. B368, 32 (1996).
[35] I. F. Silvera, Rev. Modern Phys. 52, 393 (1980), and references therein.
[36] M. L. Klein and R. Koehler, J. Phys. C3, L102 (1970).
[37] M. Nielsen, Phys. Rev. B7, 1626 (1973).
[38] P. C. Souers, *Hydrogen Properties for Fusion Energy* (University of California Press, Berkeley, 1986).

[39] L. I. Menshikov and L. I. Ponomarev, Phys. Lett. **B167**, 141 (1986).

[40] M. P. Faifman *et al.*, Hyp. Interact. **101/102**, 179 (1996).

[41] K. Fukushima, Phys. Rev. **A48**, 4130 (1993).

[42] A. Adamczak, Hyp. Interact. **119**, 23 (1999).

[43] N. I. Voropaev *et al.*, Hyp. Interact. **118**, 135 (1999).

---

**FIG. 1**: System of coordinates used for the calculation of resonant formation of the complex \([(dd\mu)dee]\) in a condensed deuterium target.

**FIG. 2**: Position of impinging \(d\mu\) atom with respect to the condensed target.
FIG. 3: Transition matrix elements $|V_{if}(\varepsilon)|^2$ for $K_i = 0$ and $K_f = 0, 1, 2$ versus $d\mu$ energy.

FIG. 4: Transition matrix elements $|V_{if}(\varepsilon)|^2$ for $K_i = 1$ and $K_f = 0, 1, 2$ versus $d\mu$ energy.
FIG. 5: Response function $S_i(\kappa, \varepsilon - \varepsilon_if)$ (in arbitrary units) for the para-D$_2$ crystal at 3 K. The dashed line is obtained for the subthreshold resonance $\varepsilon_if \approx -9.0$ meV, the solid line corresponds to $\varepsilon_if \approx 1.6$ meV. The vertical line represents the rigid lattice term $\delta(\varepsilon - \varepsilon_if) \exp(-2W)$.

FIG. 6: Formation rate $\lambda_{Kf}^{K_i}(\varepsilon)$ for $F = \frac{3}{2}$ in 3 K ortho-D$_2$ (solid line) and para-D$_2$ (dashed line). The labels “1 $\rightarrow$ 2” and “0 $\rightarrow$ 1” denote the rotational transition $K_i \rightarrow K_f$ corresponding to the lowest non-phonon processes.
FIG. 7: Effective formation rate $\bar{\lambda}_{K_i}^F(\varepsilon)$ for $F = \frac{3}{2}$ in 3 K solid ortho-$D_2$ and para-$D_2$. The labels “1 → 2” and “0 → 1” denote the rotational transition $K_i \rightarrow K_f$ corresponding to the lowest non-phonon processes.

FIG. 8: Effective formation rate $\bar{\lambda}_{K_i}^F(\varepsilon)$ for $F = \frac{1}{2}$ in 3 K solid ortho-$D_2$ and para-$D_2$. The label “gas” denotes the curve obtained for 3 K gaseous deuterium ($K_i = 0$), using the asymptotic formula (46) for the response function $S_i$ with $T_{\text{eff}} = 3$ K.
FIG. 9: Distribution of final $d\mu$ energy after $dd\mu$ back decay from $S = \frac{1}{2}, K_f = 0$ to $F' = \frac{1}{2}, K' = 0, 1, 2$. The three peaks describe the rotational transitions without a simultaneous phonon excitation.

FIG. 10: Total cross section for $d\mu (F = \frac{3}{2})$ scattering in statistical mixture of solid ortho-D$_2$ and para-D$_2$. The label “1 → 0” denotes the rotational deexcitation $K = 0 \rightarrow 1$ of a target D$_2$ molecule. The curves “−phonon” and “+phonon” stand for $d\mu$ scattering with phonon annihilation and creation, respectively. The Bragg cross section is calculated for the fcc polycrystalline lattice.
FIG. 11: Cross section for $d\mu (F = \frac{3}{2})$ scattering in solid ortho-D$_2$. The labels are identical to those in Fig. 10.

FIG. 12: Calculated time evolution of average $d\mu$ energy $\varepsilon_{avg}$ for $F = \frac{3}{2}$ in 3 K solid ortho-D$_2$, para-D$_2$, and their statistical mixture (stat). A Maxwell distribution of $d\mu$ initial energy, with mean energy of 1 eV, has been assumed.
FIG. 13: The effective resonant $d\mu$ formation rate as a function of mean CMS energy $\varepsilon_{\text{avg}}$ of $d\mu(F = \frac{3}{2})$ atom for gas and solid deuterium targets. A steady Maxwell distribution of $d\mu$ energy is assumed for a given $\varepsilon_{\text{avg}}$. The contributions from the two lowest resonant peaks to the formation rate are taken into account.

FIG. 14: Resonant $d\mu$ formation rate for $F = \frac{3}{2}$ in the statistical mixture of ortho-$D_2$ and para-$D_2$ for the resonance peak width $\Gamma^S = 0.5$ meV. Monte Carlo distribution of $d\mu$ energy at $t=10$ ns and $t=30$ ns after the muon stop is plotted (in arbitrary units).
FIG. 15: The Monte Carlo fusion neutron spectrum for the statistical mixture of 3 K solid ortho-D$_2$ and para-D$_2$ (solid line). The dashed line represents the spectrum obtained using an analytical steady state kinetics model with $\tilde{\chi}_{stat}^{3/2} = 3 \mu s^{-1}$. The initial $d\mu$ energy is given by a Maxwell distribution with mean energy of 1 eV. The width $\Gamma^S$ of the non-phonon resonances is fixed at 0.5 meV. A $3.2 \times 10^{-6}$ concentration of nitrogen is included.

FIG. 16: Calculated neutron spectra from 3 K solid ortho-D$_2$ and para-D$_2$. The Maxwell distribution of initial $d\mu$ energy with $\varepsilon_{avg} = 1$ eV and $\Gamma^S = 0.5$ meV have been assumed for the both targets.
TABLE I: The lowest resonance energies of $dd\mu$ formation in $d\mu$ scattering from single D$_2$ molecule ($\epsilon_{if}$) and from 3 K solid deuterium target ($\epsilon'_{if}$). These energies are given in the respective CMS systems.

| $\epsilon_{if}$ (meV) | $\epsilon'_{if}$ (meV) | F | $K_i$ | $K_f$ | S |
|-----------------------|------------------------|---|------|------|---|
| −7.218                | −9.028                 | 3 | 1    | 0    | $\frac{1}{2}$ |
| −3.667                | −5.477                 | 3 | 1    | 1    | $\frac{1}{2}$ |
| 0.5368                | −1.272                 | 3 | 0    | 0    | $\frac{1}{2}$ |
| 3.422                 | 1.612                  | 3 | 1    | 2    | $\frac{1}{2}$ |
| 4.088                 | 2.279                  | 3 | 0    | 1    | $\frac{1}{2}$ |
| 11.18                 | 9.368                  | 3 | 0    | 2    | $\frac{1}{2}$ |
| 42.10                 | 40.30                  | 1 | 1    | 0    | $\frac{1}{2}$ |
| 45.66                 | 43.85                  | 1 | 1    | 1    | $\frac{1}{2}$ |
| 49.86                 | 48.05                  | 1 | 0    | 0    | $\frac{1}{2}$ |
| 52.74                 | 50.94                  | 1 | 1    | 2    | $\frac{1}{2}$ |
| 53.41                 | 51.60                  | 1 | 0    | 1    | $\frac{1}{2}$ |
| 60.50                 | 58.69                  | 1 | 0    | 2    | $\frac{1}{2}$ |