

Resonant $dd\mu$ formation in condensed deuterium

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The rate of $dd\mu$ muonic-molecule resonant formation in $d\mu$ atom collisions 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 a 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 orthodeuterium and paradeuterium. 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 recoil-less resonance peaks in the vicinity of 2 meV and very slow deceleration of $d\mu$ atoms below 10 meV. Good agreement between the calculated and experimental spectra is achieved when a broadening of D_2 rotational and vibrational levels in solid deuterium is taken into account. It has been shown that resonant $dd\mu$ formation with simultaneous phonon creation in the 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.

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I. INTRODUCTION

A theoretical study of resonant formation of the muonic molecule $dd\mu$ in condensed deuterium targets is the main subject of this paper. Resonant $dd\mu$ formation, first observed by Dzhelepov and co-workers [1], is a key process of muon catalyzed fusion (μ CF) in deuterium (see, e.g., reviews [2,3]). A muonic deuterium atom $d\mu$ is created when a negative muon μ^- 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 a $d\mu$ 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 $|\varepsilon_{Jv=11}| \approx 1.97$ eV. This energy, according to the Vesman mechanism [4], is completely transferred to excited rotational-vibrational states of the molecular complex $[(dd\mu)dee]$. The scheme of calculation of the $dd\mu$ formation rate in gaseous deuterium has been developed for many years [5–8] and led to 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 [11–13]. 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 the 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 (~ 1 eV) [15,16], the 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, Bragg scattering, phonon scattering, rotational, and vibrational transitions in

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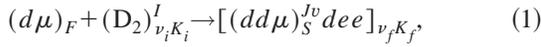
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D_2 molecules are included. We consider a dependence of the resonant formation rate and time spectra on broadening of the rotational-vibrational D_2 energy levels, due to the binding of molecules in the lattice [18].

Since it has been predicted in Refs. [19–21] 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. VI.

II. RESONANT FORMATION IN A FREE MOLECULE

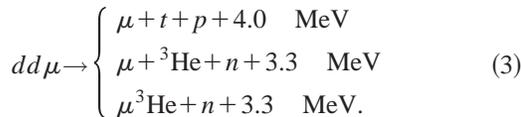
First we consider resonant formation of the $dd\mu$ molecule in the following reaction:



where D_2 is a free deuterium molecule in the initial rotational-vibrational state $(\nu_i K_i)$ with total nuclear spin \mathbf{I} . The muonic atom $d\mu$ has total spin \mathbf{F} and center-of-mass (CMS) kinetic energy ε . The complex $[(dd\mu) vee]$ 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 \mathbf{S} . The rate $\lambda_{\nu_i K_i, \nu_f K_f}^{SF}$ of the process above depends on the elastic width $\Gamma_{\nu_f K_f, \nu_i K_i}^{SF}$ of $[(dd\mu) vee]$ complex decay [22–25] in reactions

$$[(dd\mu)_S^J vee]_{\nu_f K_f} \begin{cases} \xrightarrow{\Gamma_{\nu_f K_f, \nu_i K_i}^{SF}} (d\mu)_F + (D_2)_{\nu_i K_i}^I \\ \rightarrow \text{stabilization processes,} \\ \tilde{\lambda}_f \end{cases} \quad (2)$$

where $\tilde{\lambda}_f$ is the total rate of the stabilization processes, i.e., deexcitation and nuclear fusion in $dd\mu$



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

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

$$\Gamma_{\nu_f K_f, \nu_i K_i}^{SF} = 2\pi A_{if} \int \frac{d^3k}{(2\pi)^3} |V_{if}(\varepsilon)|^2 \delta(\varepsilon_{if} - \varepsilon), \quad (4)$$

where $V_{if}(\varepsilon)$ is the transition-matrix element and ε_{if} is the 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 \mathbf{k} is the momentum of relative $d\mu$ and D_2 motion,

$$\varepsilon = k^2/2\mathcal{M}, \quad (5)$$

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

$$\Gamma_{\nu_f K_f, \nu_i K_i}^{SF} = \frac{\mathcal{M} k_{if}}{\pi} A_{if} |V_{if}(\varepsilon_{if})|^2, \quad k_{if} = k(\varepsilon_{if}). \quad (6)$$

Since $\Gamma_{\nu_f K_f, \nu_i K_i}^{SF}$ and $\tilde{\lambda}_f$ are much lower ($\sim 10^{-3}$ meV) than ε , Vesman's model can be applied and the energy-dependent resonant-formation rate has the Dirac-delta function profile

$$\lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) = 2\pi N B_{if} |V_{if}(\varepsilon)|^2 \delta(\varepsilon - \varepsilon_{if}), \quad (7)$$

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

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

$$B_{if} = 2W_{SF} \frac{2S + 1}{2F + 1}, \quad (8)$$

where

$$W_{SF} = (2F + 1) \begin{Bmatrix} \frac{1}{2} & 1 & F_2 \\ 1 & S & 1 \end{Bmatrix},$$

$$\xi(K_i) = \begin{cases} \frac{2}{3} & \text{for } K_i = 0 \\ \frac{1}{3} & \text{for } K_i = 1, \end{cases} \quad (9)$$

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 $\lambda_{\nu_i K_i, \nu_f K_f}^{SF}(T)$.

III. RESONANT FORMATION IN A CONDENSED TARGET

Since a muonic deuterium atom can be approximately treated as a small neutronlike particle, methods used for the description of neutron scattering and absorption in condensed matter are applicable in the case of $dd\mu$ 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 \mathcal{S}_i [14], for calculation of the resonant- $dd\mu$ -formation rates.

The 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:

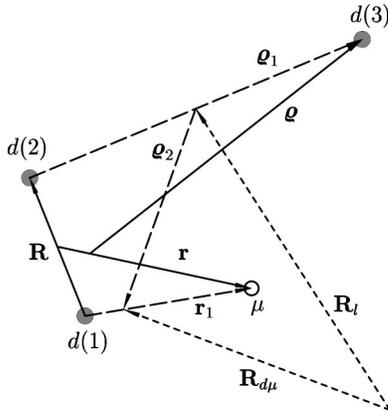


FIG. 1. System of coordinates used for the calculation of resonant formation of the complex $[(dd\mu)dee]$ in a condensed-deuterium target.

$$H_{\text{tot}} = \frac{1}{2M_{d\mu}} \nabla_{R_{d\mu}}^2 + H_{d\mu}(\mathbf{r}_1) + H_{D_2}(\varrho_1) + V(\mathbf{r}_1, \varrho_1, \varrho_2) + H, \quad (10)$$

where $M_{d\mu}$ is the $d\mu$ mass and $\mathbf{R}_{d\mu}$ 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 and \mathbf{r}_1 is the $d\mu$ internal vector; H_{D_2} denotes the internal Hamiltonian of a free D_2 molecule. It is assumed that $dd\mu$ 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 \mathbf{R}_l ; ϱ_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 $dd\mu$ resonant formation. Vector ϱ_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 ε of the $d\mu$ atom and its momentum \mathbf{k} in the target frame are connected by the relation

$$\varepsilon = k^2/2M_{d\mu}. \quad (11)$$

The Hamiltonian H of a pure D_2 target, corresponding to the initial target energy E_0 , has the form

$$H = \sum_j \frac{1}{2M_{\text{mol}}} \nabla_{R_j}^2 + \sum_j \sum_{j' \neq j} U_{jj'}, \quad (12)$$

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

The coordinate part Ψ_{tot} of the initial wave function of the system can be written as a product

$$\Psi_{\text{tot}} = \psi_{d\mu}^{1S}(\mathbf{r}_1) \psi_{D_2}^{v_i K_i}(\varrho_1) \exp(i\mathbf{k} \cdot \mathbf{R}_{d\mu}) |0\rangle, \quad (13)$$

where $|0\rangle$ stands for the initial wave function of the condensed D_2 target, corresponding to the total energy E_0 .

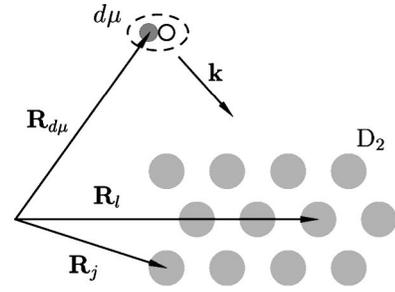


FIG. 2. Position of impinging $d\mu$ atom with respect to the condensed target.

Eigenfunctions of the operators $H_{d\mu}$ and H_{D_2} are denoted by $\psi_{d\mu}^{1S}$ and $\psi_{D_2}^{v_i K_i}$, respectively. Using the relation $\mathbf{R}_{d\mu} = \mathbf{R}_l + \varrho_2$, the wave function Ψ_{tot} takes the form

$$\Psi_{\text{tot}} = \psi_{d\mu}^{1S}(\mathbf{r}_1) \psi_{D_2}^{v_i K_i}(\varrho_1) \exp(i\mathbf{k} \cdot \varrho_2) \exp(i\mathbf{k} \cdot \mathbf{R}_l) |0\rangle, \quad (14)$$

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

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

$$H_{\text{tot}} \approx H'_{\text{tot}} = H_{dd\mu}(\mathbf{r}, \mathbf{R}) + H_C(\varrho) + V(\varrho, \mathbf{r}, \mathbf{R}) + \tilde{H}, \quad (15)$$

where $H_{dd\mu}$ is an internal Hamiltonian of the $dd\mu$ molecular ion, and vectors \mathbf{r} and \mathbf{R} are its Jacobi coordinates. Relative motion of $dd\mu$ and d in the complex is described by a Hamiltonian H_C , which depends on the respective internal vector ϱ . 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

$$\begin{aligned} \tilde{H} &= \frac{1}{2M_C} \nabla_{R_l}^2 + \sum_{j \neq l} \frac{1}{2M_{\text{mol}}} \nabla_{R_j}^2 + \sum_j \sum_{j' \neq j} U_{jj'} \\ &= - \left(1 - \frac{M_{\text{mol}}}{M_C} \right) \frac{1}{2M_{\text{mol}}} \nabla_{R_l}^2 + H = \Delta H + H, \end{aligned} \quad (16)$$

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

$$\Psi'_{\text{tot}} = \psi_{dd\mu}^{Jv}(\mathbf{r}, \mathbf{R}) \psi_C^{v_f K_f}(\varrho) |\tilde{n}\rangle, \quad (17)$$

where $\psi_{dd\mu}^{Jv}$ and $\psi_C^{v_f K_f}$ denote eigenfunctions of the Hamiltonians $H_{dd\mu}$ and H_C , respectively.

The energy-dependent resonant- $dd\mu$ -formation rate $\lambda_{v_i K_i, v_f K_f}^{SF}(\varepsilon)$ 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

$$\lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) = 2\pi N B_{if} |A_{i0,fn}|^2 \delta(\varepsilon - \varepsilon_{if} + E_0 - \tilde{E}_n), \quad (18)$$

with the resonance condition

$$\varepsilon + E_0 = \varepsilon_{if} + \tilde{E}_n, \quad (19)$$

taking into account the initial and final energy of the target. The resonant energy for a free D_2 is denoted by ε_{if} and the transition-matrix element is given by

$$A_{i0,fn} = \langle \Psi'_{\text{tot}} | V | \Psi_{\text{tot}} \rangle. \quad (20)$$

Using Eqs. (14) and (17) the matrix element (20) can be written as a product

$$A_{i0,fn} = \langle \tilde{n} | \exp(i\mathbf{k} \cdot \mathbf{R}_l) | 0 \rangle V_{if}(\varepsilon), \quad (21)$$

where $V_{if}(\varepsilon)$ is the transition-matrix element calculated for a single D_2 molecule [8]. The rate (18) can be additionally averaged over a distribution ρ_{n_0} of the initial target states at a given temperature T and summed over the final target states, which leads to

$$\lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) = 2\pi N B_{if} |V_{if}(\varepsilon)|^2 \sum_{n, n_0} \rho_{n_0} |\langle \tilde{n} | \exp(i\vec{k} \cdot \mathbf{R}_l) | 0 \rangle|^2 \times \delta(\varepsilon - \varepsilon_{if} + E_0 - \tilde{E}_n). \quad (22)$$

The 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 rotational quantum numbers. This factor takes also into account a symmetrization of the total wave function of $d\mu + D_2$ system over three deuterium nuclei.

Now we introduce a time variable t to eliminate the δ 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 δ function

$$\delta(\varepsilon - \varepsilon_{if} + E_0 - \tilde{E}_n) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \times \exp[-it(\varepsilon - \varepsilon_{if} + E_0 - \tilde{E}_n)], \quad (23)$$

one has

$$\begin{aligned} \lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) &= N B_{if} |V_{if}|^2 \int_{-\infty}^{\infty} dt \\ &\times \exp[-it(\varepsilon - \varepsilon_{if})] \\ &\times \sum_{n, n_0} \rho_{n_0} \langle 0 | \exp(-i\mathbf{k} \cdot \mathbf{R}_l) | \tilde{n} \rangle \\ &\times \langle \tilde{n} | \exp(it\tilde{E}_n) \exp(i\mathbf{k} \cdot \mathbf{R}_l) \exp(-itE_0) | 0 \rangle. \end{aligned} \quad (24)$$

Assuming that the perturbation operator ΔH is well approximated by its mean value

$$\Delta H \approx \langle 0 | \Delta H | 0 \rangle \equiv \Delta \varepsilon_{if} = -(1 - M_{\text{mol}}/M_C) \mathcal{E}_T < 0, \quad (25)$$

which is valid when the target relaxation time is much smaller than the $dd\mu$ lifetime of the order of 10^{-9} s, the matrix element in Eq. (24) can be expressed as

$$\begin{aligned} &\langle \tilde{n} | \exp(it\tilde{E}_n) \exp(i\mathbf{k} \cdot \mathbf{R}_l) \exp(-itE_0) | 0 \rangle \\ &= \langle \tilde{n} | \exp[it(H + \Delta H)] \exp(i\mathbf{k} \cdot \mathbf{R}_l) \exp(-itH) | 0 \rangle \\ &\approx \langle \tilde{n} | \exp(it\Delta \varepsilon_{if}) \exp(itH) \exp(i\mathbf{k} \cdot \mathbf{R}_l) \exp(-itH) | 0 \rangle \\ &= \langle \tilde{n} | \exp(it\Delta \varepsilon_{if}) \exp[i\mathbf{k} \cdot \mathbf{R}_l(t)] | 0 \rangle, \end{aligned} \quad (26)$$

where $\mathbf{R}_l(t)$ denotes the Heisenberg operator and \mathcal{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

$$\begin{aligned} \lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) &= N B_{if} |V_{if}(\varepsilon)|^2 \int_{-\infty}^{\infty} dt \exp[-it(\varepsilon - \varepsilon'_{if})] \\ &\times \langle \exp[-i\mathbf{k} \cdot \mathbf{R}_l(0)] \exp[i\mathbf{k} \cdot \mathbf{R}_l(t)] \rangle_T, \end{aligned} \quad (27)$$

where $\langle \dots \rangle_T$ denotes both the quantum-mechanical and statistical averaging at temperature T , with ε'_{if} being the resonance energy

$$\varepsilon'_{if} = \varepsilon_{if} + \Delta \varepsilon_{if}, \quad (28)$$

shifted by $\Delta \varepsilon_{if} < 0$. Note that such a resonant-energy shift was neglected in papers [26,27], where absorption of neutrons and γ rays by heavy nuclei were considered. An estimation of the shift in the case of γ 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]:

$$\begin{aligned} &\langle \exp[-i\mathbf{k} \cdot \mathbf{R}_l(0)] \exp[i\mathbf{k} \cdot \mathbf{R}_l(t)] \rangle_T \\ &= \int d^3r G_s(\mathbf{r}, t) \exp(i\mathbf{k} \cdot \mathbf{r}), \end{aligned} \quad (29)$$

and the single-particle response function $S_i(\boldsymbol{\kappa}, \omega)$ is given by the formula

$$S_i(\boldsymbol{\kappa}, \omega) = \frac{1}{2\pi} \int d^3r dt G_s(\mathbf{r}, t) \exp[i(\boldsymbol{\kappa} \cdot \mathbf{r} - \omega t)]. \quad (30)$$

Thus, by virtue of Eqs. (27) and (30), the resonant formation rate in a condensed target can be expressed in terms of the response function

$$\lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) = 2\pi N B_{if} |V_{if}(\varepsilon)|^2 S_i(\boldsymbol{\kappa}, \omega), \quad (31)$$

where the momentum transfer $\boldsymbol{\kappa}$ and energy transfer ω to the target are defined as follows:

$$\boldsymbol{\kappa} = \mathbf{k}, \quad \omega = \varepsilon - \varepsilon'_{if}. \quad (32)$$

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 $\mathcal{S}_i(\mathbf{k}, \omega)$. It is possible to rigorously calculate \mathcal{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),

$$\Gamma_{\nu_f K_f, \nu_i K_i}^{SF'} = 2\pi A_{if} \int \frac{d^3k}{(2\pi)^3} |V_{if}(\varepsilon)|^2 \tilde{\mathcal{S}}_i(\mathbf{k}, \omega'),$$

$$\omega' = \tilde{\varepsilon}'_{if} - \varepsilon, \quad \tilde{\varepsilon}'_{if} = \varepsilon_{if} + \Delta \tilde{\varepsilon}_{if}, \quad (33)$$

$\tilde{\mathcal{S}}_i$ is the response function calculated for the state $|\tilde{n}\rangle$ and

$$\Delta \tilde{\varepsilon}_{if} \equiv \langle \tilde{n} | \Delta H | \tilde{n} \rangle = -(M_C/M_{\text{mol}} - 1) \tilde{\mathcal{E}}_T, \quad (34)$$

where $\tilde{\mathcal{E}}_T$ 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

$$G_s(\mathbf{r}, t) = \left[\frac{M_{\text{mol}}}{2\pi\gamma(t)} \right]^{3/2} \exp\left[-\frac{M_{\text{mol}}}{2\gamma(t)} r^2 \right]. \quad (35)$$

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

$$\gamma(t) = \int_{-\infty}^{\infty} dw \frac{Z(w)}{w} n_B(w) \exp(-iwt), \quad (36)$$

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

$$\int_0^{\infty} dw Z(w) = 1, \quad Z(w) = 0 \quad \text{for} \quad w > w_{\text{max}},$$

$$Z(-w) \equiv Z(w), \quad (37)$$

$n_B(w)$ is the Bose factor,

$$n_B(w) = [\exp(\beta w) - 1]^{-1}, \quad \beta = (k_B T)^{-1}, \quad (38)$$

and the Boltzmann constant is denoted by k_B .

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

$$\mathcal{S}_i(\mathbf{k}, \omega) = \frac{1}{2\pi} \exp\left[-\frac{\kappa^2}{2M_{\text{mol}}} \gamma(\infty) \right] \int_{-\infty}^{\infty} dt$$

$$\times \exp(-i\omega t) \exp\left\{ \frac{\kappa^2}{2M_{\text{mol}}} [\gamma(\infty) - \gamma(t)] \right\}, \quad (39)$$

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

$$\mathcal{S}_i(\mathbf{k}, \omega) = \exp(-2W) \left[\delta(\omega) + \sum_{n=1}^{\infty} g_n(\omega, T) \frac{(2W)^n}{n!} \right], \quad (40)$$

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

$$2W = \frac{\kappa^2}{2M_{\text{mol}}} \gamma(\infty) = \frac{\kappa^2}{2M_{\text{mol}}} \int_0^{\infty} dw \frac{Z(w)}{w} \coth\left(\frac{1}{2}\beta w\right), \quad (41)$$

and the functions g_n are given by

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

$$g_n(w, T) = \int_{-\infty}^{\infty} dw' g_1(w-w', T) g_{n-1}(w', T), \quad (42)$$

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

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

$$2W = \frac{1}{3} \langle 0 | \mathbf{u}^2 | 0 \rangle \kappa^2, \quad (43)$$

where \mathbf{u} is the displacement of a molecule from its lattice site. Substitution of Eq. (40) in Eq. (31) leads to the following formation rate:

$$\lambda_{\nu_i K_i, \nu_f K_f}^{SF}(\varepsilon) = 2\pi N B_{if} |V_{if}(\varepsilon)|^2 \exp(-2W)$$

$$\times \left[\delta(\omega) + \sum_{n=1}^{\infty} g_n(\omega, T) \frac{(2W)^n}{n!} \right]. \quad (44)$$

The first term in expansion (44) represents a sharp peak describing the δ -profile recoil-less formation. The next terms give broad distributions corresponding to subsequent multiphonon 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 [26] 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 multiphonon terms give comparable contribu-

tions to Eq. (44). Therefore, for sufficiently large κ^2 , it is convenient to use the impulse approximation in which $\gamma(t)$ is replaced by its value near $t=0$

$$\gamma(t) \approx \gamma(0) + it - \frac{2}{3}\mathcal{E}_T. \quad (45)$$

This leads to the asymptotic formula for \mathcal{S}_i

$$\mathcal{S}_i(\mathbf{\kappa}, \omega) = \frac{1}{\Delta\sqrt{\pi}} \exp\left[-\left(\frac{\omega - \mathcal{R}}{\Delta}\right)^2\right], \quad (46)$$

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

$$\mathcal{E}_T = \frac{3}{2} \int_0^\infty dw Z(w) w \left[n_B(w) + \frac{1}{2} \right]. \quad (48)$$

The energy \mathcal{E}_T contains a contribution from the zero-point vibrations, approaching $3k_B T/2$ only at high temperatures $T \gg w_{\text{max}}/k_B$. Function (46) 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 Placzek¹ for resonant absorption of neutrons in gas targets [31]. However, the resonance width (47) 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_{eff}

$$T_{\text{eff}} = \frac{2}{3}\mathcal{E}_T/k_B. \quad (49)$$

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's 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]. As a 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

$$Z(w) = \begin{cases} 3w^2/w_D^3 & \text{if } w \leq w_D \\ 0 & \text{if } w > w_D, \end{cases} \quad (50)$$

with the Debye energy $w_D = k_B \Theta_D$ and Debye temperature Θ_D taken from 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

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

$$T_{\text{eff}} = \frac{3}{8}\Theta_D \approx 40 \text{ K} \quad (51)$$

are very good approximations of Eqs. (41), (48), and (49). The Debye-Waller factor and mean kinetic energy \mathcal{E}_T at low-est 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 $T \rightarrow 0$. 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 rotational and vibrational 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 the $(dd\mu)d$ 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

¹In fact, formula (46) is the limit of the Bethe formula in the case of a very narrow natural resonance width $\Gamma \rightarrow 0$.

TABLE I. The lowest resonance energies of $dd\mu$ formation in $d\mu$ scattering from a single D_2 molecule (ε_{if}) and from 3-K solid-deuterium target (ε'_{if}). These energies are given in the respective CMS systems.

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

=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 ε_{if} and ε'_{if} for fixed ν_i , ν_f and different values of F , K_i , S , and K_f are shown in Table I [10]. 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 the energy of neighboring molecules can be increased. Such an effect, due to triple collisions in gas targets, has been first discussed in Ref. [39]. In a solid, the energy excess is lost through incoherent phonon creation. According to Eqs. (25), (28), and (51), in the considered 3-K solid deuterium all resonant energies ε'_{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

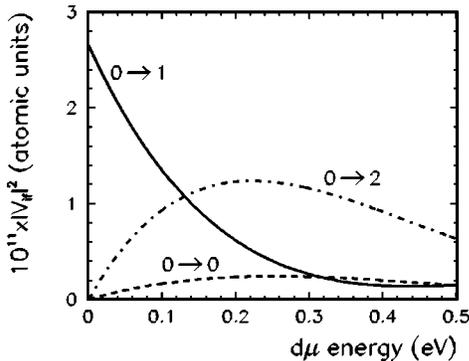


FIG. 3. Transition-matrix elements $|V_{if}(\varepsilon)|^2$ for $K_i=0$ and $K_f=0,1,2$ versus $d\mu$ energy.

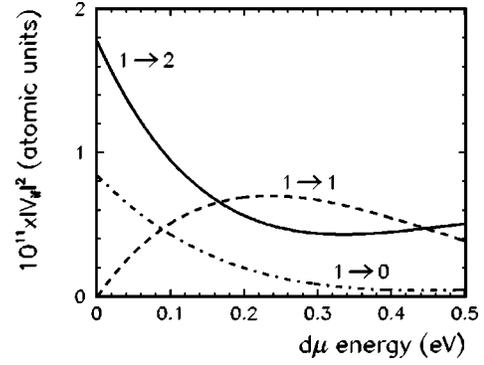


FIG. 4. Transition-matrix elements $|V_{if}(\varepsilon)|^2$ for $K_i=1$ and $K_f=0,1,2$ versus $d\mu$ energy.

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 ($\varepsilon \leq w_D$) are calculated using formula (44) with a few most significant terms of the response-function expansion (40) taken into account. Figure 5 shows the function $S_i(\boldsymbol{\kappa}, \varepsilon - \varepsilon'_{if})$ corresponding to the two dipole transitions in para- D_2 . The subthreshold 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 nonphonon 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 Eq. (40) is proportional to κ^{2n} , the $dd\mu$ formation rate tends to zero at $\varepsilon \rightarrow 0$. Note that phonon annihilation gives a 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,

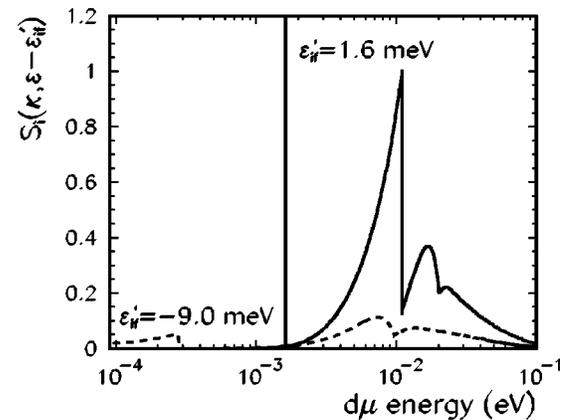


FIG. 5. Response function $S_i(\boldsymbol{\kappa}, \varepsilon - \varepsilon'_{if})$ (in arbitrary units) for the para- D_2 crystal at 3 K. The dashed line is obtained for the subthreshold resonance $\varepsilon'_{if} \approx -9.0$ meV, the solid line corresponds to $\varepsilon'_{if} \approx 1.6$ meV. The vertical line represents the rigid-lattice term $\delta(\varepsilon - \varepsilon'_{if}) \exp(-2W)$.

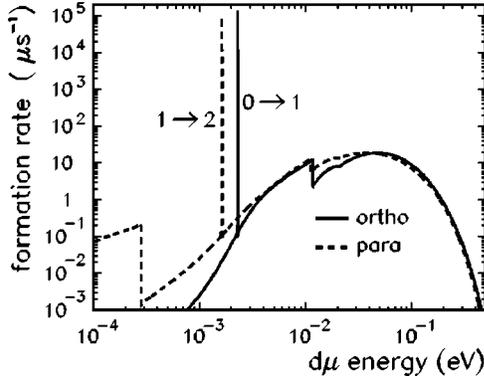


FIG. 6. Formation rate $\lambda_{K_i}^F(\varepsilon)$ for $F=\frac{3}{2}$ in 3-K ortho- D_2 (solid line) and para- D_2 (dashed line). The labels “1→2” and “0→1” denote the rotational transition $K_i \rightarrow K_f$ corresponding to the lowest nonphonon processes.

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

In Fig. 6 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 recoil-less 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 $0.1061 \text{ eV } \mu\text{s}^{-1}$ 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 $0.07544 \text{ eV } \mu\text{s}^{-1}$ as the resonance strength. Higher resonance energies involve many multiphonon 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 liquid-hydrogen density, $N_0 = 4.25 \times 10^{22}$ atoms/cm³.

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 $\bar{\lambda}_{K_i}^F(\varepsilon)$, which leads to the nuclear dd fusion in the $[(dd\mu)dee]$ complex. Back decay of the complex to the $d\mu + D_2$ system, characterized by the quantum numbers K'_i and F' , strongly influences the fusion process because the back-decay rates are comparable with the effective fusion rate $\bar{\lambda}_f \approx 374 \mu\text{s}^{-1}$ [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:

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

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

$$\mathcal{P}_S^{\text{fus}} = \frac{\bar{\lambda}_f}{\Gamma^S}, \quad \Gamma^S = \bar{\lambda}_f + \sum_{F'} \Gamma^{SF'},$$

$$\Gamma^{SF'} = \sum_{K'_i, K_f=0} \Gamma_{\nu_f K_f, \nu_i K'_i}^{SF'}. \quad (54)$$

Since the frequency of lattice vibrations ($\sim w_D/\hbar \sim 10^7 \mu\text{s}^{-1}$) 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 $w \geq k_B T \approx 0.26$ meV is strongly suppressed by the Bose factor $n_B(w)$. 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 the $d\mu$ state $F=\frac{3}{2}$ due to subthreshold resonances, is closed because this would require 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 ($\Delta E^{\text{hfs}} \gg w_D$), the $dd\mu$ decay rate is dominated by contributions from simultaneous phonon-creation processes.

After integration of formula (33) over the direction of vector \mathbf{k} one obtains

$$\Gamma_{\nu_f K_f, \nu_i K_i}^{SF'} = \frac{A_{if}}{\pi} \int_0^\infty dk k^2 |V_{if}(\varepsilon)|^2 \bar{S}_i(k^2, \omega'), \quad (55)$$

and then substitution of expansion (40) and integration of the recoil-less term lead to

$$\Gamma_{\nu_f K_f, \nu_i K_i}^{SF'} = \frac{A_{if}}{\pi} \left[M \bar{k}_{if} |V_{if}(\tilde{\varepsilon}'_{if})|^2 \exp(-2\tilde{W}_{if}) + \sum_{n=1}^\infty \int_0^\infty dk k^2 |V_{if}(\varepsilon)|^2 \times \exp(-2\tilde{W}) g_n(\omega', T) \frac{(2\tilde{W})^n}{n!} \right], \quad (56)$$

where

$$2\tilde{W} = \frac{k^2}{2M_C} \gamma(\infty), \quad 2\tilde{W}_{if} = 2\tilde{W}(\bar{k}_{if}), \quad \bar{k}_{if} = \sqrt{2M\tilde{\varepsilon}'_{if}}. \quad (57)$$

It is assumed in the formula above that the phonon-energy spectrum of solid deuterium containing $[(dd\mu)dee]$ 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 the 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 $\bar{\mathcal{E}}_T$ of the complex reaches

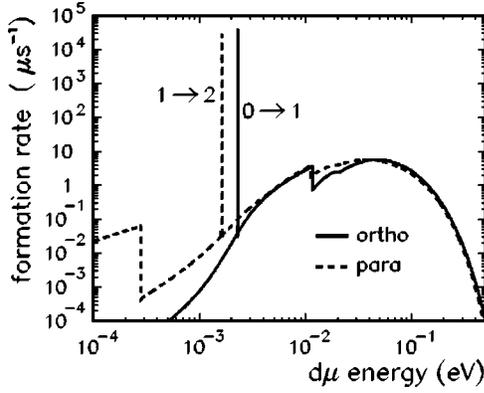


FIG. 7. Effective formation rate $\bar{\lambda}_{K_i}^F(\varepsilon)$ for $F=\frac{3}{2}$ in 3-K solid ortho-D₂ and para-D₂. The labels “1→2” and “0→1” denote the rotational transition $K_i \rightarrow K_f$ corresponding to the lowest non-phonon processes.

the energy \mathcal{E}_T characterizing a pure deuterium solid. Thus the resonance energy shift (34) is approximated by

$$\Delta \tilde{\varepsilon}_{if} \approx -(M_C/M_{\text{mol}} - 1)\mathcal{E}_T \approx -2.77 \text{ meV}, \quad (58)$$

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

The effective formation rates in 3-K solid deuterium for $F=\frac{3}{2}$ 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 $2.7 \mu\text{s}^{-1}$ 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 nonresonant $dd\mu$ formation rate $\lambda_{\text{nr}} \approx 0.44 \mu\text{s}^{-1}$ [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 $1 \mu\text{s}^{-1}$. A cusp at 0.3 meV in para-D₂ is due to the formation with simultaneous one-phonon creation, connected with the subthreshold resonance $K_i=1 \rightarrow K_f=0$. This implies a significant difference between the resonant formation in ortho-D₂ and para-D₂ 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 $\mathcal{P}_S^{\text{fus}} \approx 0.3$ and the total resonance width $\Gamma^S \approx 0.8 \times 10^{-3} \text{ meV}$ for both $S=\frac{1}{2}$ and $S=\frac{3}{2}$. The back-decay rate $\Gamma^{SF'}$ from $S=\frac{1}{2}$ to $F'=\frac{1}{2}$ equals about $843 \mu\text{s}^{-1}$. Decay $S=\frac{1}{2} \rightarrow F'=\frac{1}{2}$ is impossible. In the case of $S=\frac{3}{2}$ we have obtained $\Gamma^{SF'} \approx 281 \mu\text{s}^{-1}$ for $F'=\frac{1}{2}$ and $\Gamma^{SF'} \approx 610 \mu\text{s}^{-1}$ for $F'=\frac{3}{2}$. Phonon-creation processes give dominant contributions to the back-decay rates, e.g., the nonphonon part of $\Gamma^{SF'}$, given by the first term of expansion (56), equals $169 \mu\text{s}^{-1}$. 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 $F=\frac{1}{2}$ are presented. For the sake of comparison, the formation rate

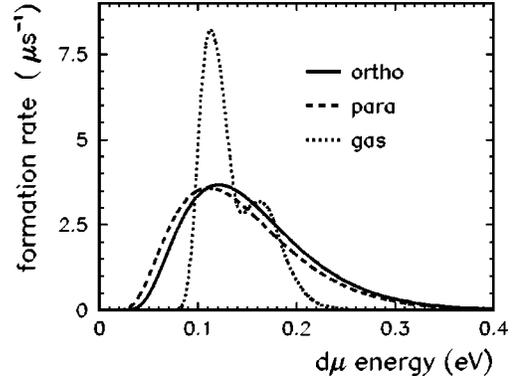


FIG. 8. Effective formation rate $\bar{\lambda}_{K_i}^F(\varepsilon)$ for $F=\frac{1}{2}$ in 3-K solid ortho-D₂ and para-D₂. The label “gas” denotes the curve obtained for 3-K gaseous deuterium ($K_i=0$), using the asymptotic formula (46) for the response function \mathcal{S}_i with $T_{\text{eff}}=3 \text{ K}$.

for 3 K ortho-D₂ gas is also plotted. The “gas” curve has been calculated using the asymptotic formula (46) for \mathcal{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 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 μ 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 $S=\frac{1}{2}$, $K_f=0$, and $F'=\frac{1}{2}$. The rotational transitions to $K'_i=0,1,2$ with no phonon-creation are seen as delta peaks. The continuous energy spectrum describes phonon creation contribution to the $d\mu$ energy. Note that, opposite to $dd\mu$ formation rates, this phonon contribution (for a given rotational transition peak) extends towards lower energies. The

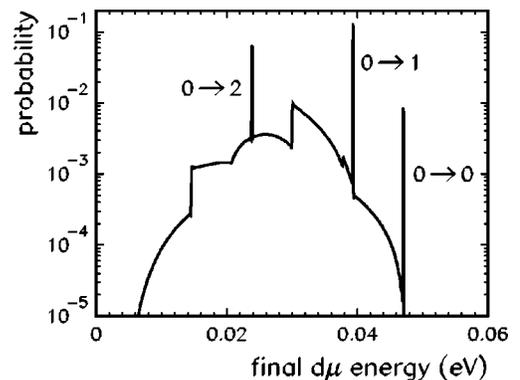


FIG. 9. Distribution of final $d\mu$ energy after $dd\mu$ back decay from $S=\frac{1}{2}, K_f=0$ to $F'=\frac{1}{2}, K'_i=0,1,2$. The three peaks describe the rotational transitions without simultaneous phonon excitation.

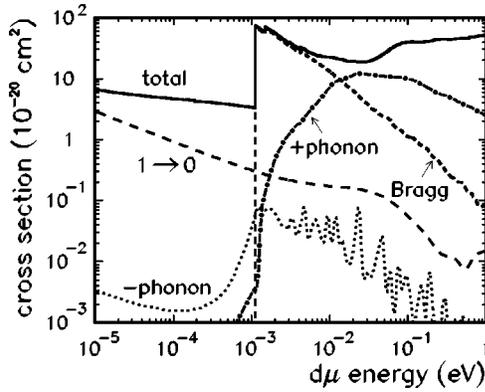


FIG. 10. Total cross section for $d\mu(F=\frac{3}{2})$ scattering in a statistical mixture of solid ortho- D_2 and para- D_2 . The label “ $1 \rightarrow 0$ ” denotes the rotational deexcitation $K=0 \rightarrow 1$ of a target D_2 molecule. The curves “-phonon” and “+phonon” stand for $d\mu$ scattering with phonon annihilation and creation, respectively. The Bragg cross section is calculated for the fcc polycrystalline lattice.

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, rotational and vibrational 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 incoherent processes, such as spin-flip or rovibrational transitions, are described by the self-pair-correlation function $G_s(\mathbf{r}, t)$ defined by Eq. (29). The Bragg scattering and coherent phonon scattering are connected with a pair correlation function $G(\mathbf{r}, t)$ [14].

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 the $d\mu$ 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 “ $0 \rightarrow 1$ ” in Fig. 10, describing the rotational deexcitation, includes contributions from simultaneous incoherent phonon processes. This cross section at $\varepsilon = 2.5$ meV equals 0.22×10^{-20} cm², 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 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

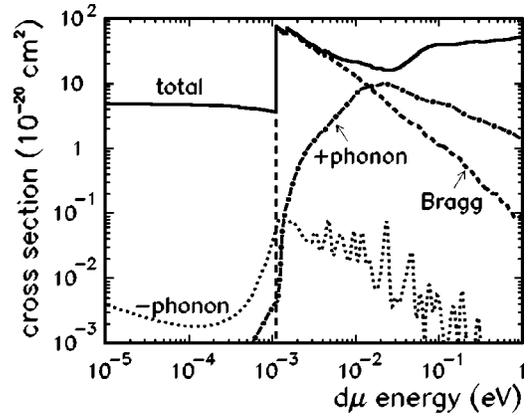


FIG. 11. Cross section for $d\mu(F=\frac{3}{2})$ scattering in solid ortho- D_2 . The labels are identical to those in Fig. 10.

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 ε_B . This limit is obtained in the case of the coherent one-phonon-creation process, for the total-momentum conservation involving the smallest (nonzero) inverse lattice vector τ , which also fixes the position of the first peak of the Bragg scattering at $\varepsilon_B = 1.1$ meV. For $\tau = \mathbf{0}$ 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 1.2×10^5 cm/s and this corresponds to a $d\mu$ 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.

Figure 12 presents the time evolution of average $d\mu(F=\frac{3}{2})$ atom energy ε_{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 nonresonant part of the total spin-flip rate $\lambda_{3/2,1/2}$ is multiplied by a single scaling factor of 0.4, in order to keep agreement with the experimental values [10,43] 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 a $d\mu$ mean energy of 10 meV is reached after only 5 ns. Then, below the Debye en-

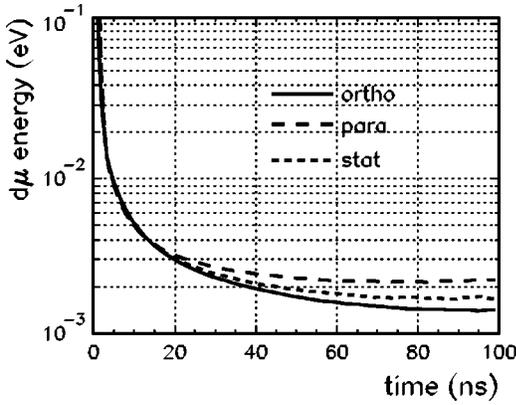


FIG. 12. Calculated time evolution of average $d\mu$ energy ε_{avg} for $F=\frac{3}{2}$ in 3 K solid ortho- D_2 , para- D_2 , and their statistical mixture (stat). A Maxwell distribution of $d\mu$ initial energy, with mean energy of 1 eV, has been assumed.

ergy, deceleration becomes very slow. The lowest value of ε_{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 $\varepsilon'_{if} = 1.6$ meV. However, even if ε_{avg} is smaller than ε'_{if} , a significant fraction of $d\mu$ atoms has energy $\varepsilon \geq \varepsilon'_{if}$ because of a large admixture of hot $d\mu$ atoms at $t=0$ [15,16] and slow deceleration below 10 meV.

Since at energies of a few meV the lowest delta peaks are dominant in 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 ε_{avg} . The

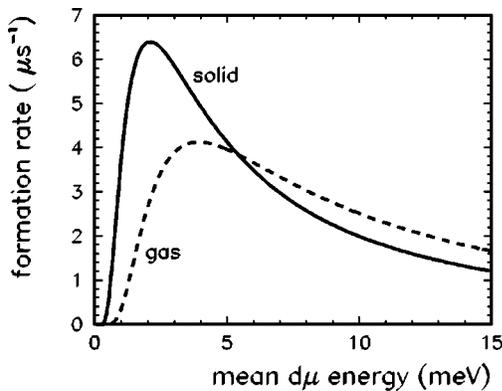


FIG. 13. The effective resonant- $dd\mu$ -formation rate as a function of mean CMS energy ε_{avg} of $d\mu(F=\frac{3}{2})$ atom for gas- and solid-deuterium targets. A steady Maxwell distribution of $d\mu$ energy is assumed for a given ε_{avg} . The contributions from the two lowest resonant peaks to the formation rate are taken into account.

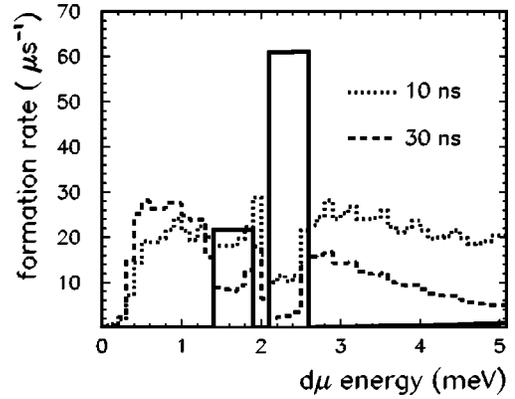


FIG. 14. Resonant- $dd\mu$ -formation rate for $F=\frac{3}{2}$ in a statistical mixture of ortho- D_2 and para- D_2 for the resonance peak width $\Gamma^S = 0.5$ meV. The Monte Carlo distribution of $d\mu$ energy at $t = 10$ ns and $t = 30$ ns after the muon stop is plotted (in arbitrary units).

maximum average rate of about $6 \mu\text{s}^{-1}$ in the solid is due to the resonance-energy shift of -1.8 meV. The experimental result of $3 \mu\text{s}^{-1}$ can be explained because ε_{avg} is greater than 1 meV. However, in order to obtain large fusion-neutron and -proton yields through resonant $dd\mu$ formation, the width Γ^S of the resonance peaks in the solid cannot be too narrow. The peak resonant rates of a few $10^4 \mu\text{s}^{-1}$ have been obtained assuming discrete values of the rotational-vibrational 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 $\sim 10 \mu\text{s}^{-1}$. In this case $d\mu$ atoms are very quickly [compared to the $d\mu(F=\frac{3}{2})$ lifetime] removed from regions of the resonance peaks and the contribution of the recoil-less 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. As a result, the calculated time spectra, obtained for small Γ^S , are dominated by weak nonresonant $dd\mu$ formation, which disagrees with the experimental data. Therefore, we have investigated the influence of a broadening of the non-phonon resonant peaks due to the presence of molecular rotational and vibrational bands in the solid, discussed in Ref. [18]. 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 within 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 a resonant formation rate of $294 \mu\text{s}^{-1}$ for the recoil-less peak in ortho- D_2 and $214 \mu\text{s}^{-1}$ in para- D_2 . In Fig. 14 one sees the resonant formation rate at lowest energies for $\Gamma^S = 0.5$ meV and for a 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. A Maxwell distribution of initial $d\mu$ energy, with $\varepsilon_{\text{avg}} = 1$ eV, has been assumed. Two minima in the $d\mu$ energy distribu-

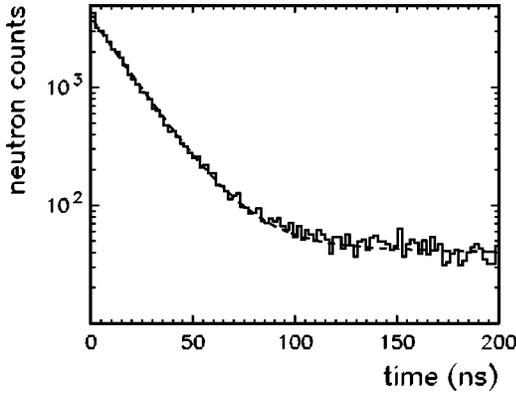


FIG. 15. The Monte Carlo fusion-neutron spectrum for a statistical mixture of 3-K solid ortho-D₂ and para-D₂ (solid line). The dashed line represents the spectrum obtained using an analytical steady-state-kinetics model with $\bar{\lambda}_{\text{stat}}^{3/2} = 3 \mu\text{s}^{-1}$. The initial $d\mu$ energy is given by a Maxwell distribution with mean energy of 1 eV. The width Γ^S of the nonphonon resonances is fixed at 0.5 meV. A 3.2×10^{-6} concentration of nitrogen is included.

tion 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 $30 \mu\text{s}^{-1}$.

The dd fusion-neutron spectrum, calculated assuming the same initial $d\mu$ energy and resonance profiles, is shown in Fig. 15. A 3.2×10^{-6} concentration of nitrogen is included in order to fit the TRIUMF target conditions. The dashed line plotted in this figure has been calculated using the steady-state kinetics model with an effective formation rate $\bar{\lambda}_{\text{stat}}^{3/2} = 3 \mu\text{s}^{-1}$ and total spin-flip rate $\lambda_{3/2,1/2} = 36 \mu\text{s}^{-1}$ taken from the fits to the experimental data [13]. The slope of the spectrum at $t \lesssim 80$ ns is determined by the rates $\bar{\lambda}_{\text{stat}}^{3/2}$, $\lambda_{3/2,1/2}$, and the $d\mu$ -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 $1-3 \mu\text{s}^{-1}$. The spectrum slope at large times $t \gtrsim 100$ ns, when $d\mu(F = \frac{3}{2})$ atoms practically disappear, is due to nonresonant $dd\mu$ formation from $F = \frac{1}{2}$ and to muon transfer to nitrogen contamination.

The shape of the time spectra is practically unchanged when the mean energy ε_{avg} of the initial single Maxwell distribution varies within the limits 0.01–1 eV. On the other hand, the spectra change substantially if a significant fraction 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 Γ^S is greater than 0.5 meV we obtain results that begin to differ significantly from the analytical curve calculated with the experimental parameters. In particular, the ratios of neutron yields from the short and long times begin to disagree. Fits of the calculated spectra to the experimental data would enable a better determination of Γ^S and a shape of the initial $d\mu$ energy. However, this is not the purpose of

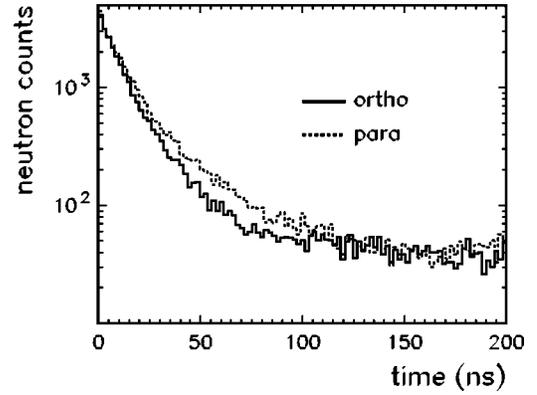


FIG. 16. Calculated neutron spectra from 3-K solid ortho-D₂ and para-D₂. A Maxwell distribution of initial $d\mu$ energy with $\varepsilon_{\text{avg}} = 1$ eV and $\Gamma^S = 0.5$ meV has been assumed for the both targets.

this work. A qualitative comparison of Monte Carlo spectra with the experimental data has already been performed in Ref. [13]. In this case good fits were not obtained since at that time, the resonant $dd\mu$ formation rates in solid D₂ 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-D₂ and para-D₂. There are certain differences between the neutron time spectra from these targets (see Fig. 16), caused by the different positions and strengths of the lowest resonance peaks. Also the $d\mu$ slowing-down process differs slightly in the two cases. The neutron yield at longer times is smaller for ortho-D₂ since in this case the resonance peak is placed at a higher energy of 2.3 meV. Therefore, $d\mu$ atoms are removed faster from the peak compared to the situation in para-D₂, where the resonance is observed at 1.6 meV. A greater mean $d\mu$ energy in para-D₂ (cf. Fig. 12) leads also to a stronger overlap of the resonance peak with the $d\mu$ energy distribution at $t \gtrsim 20$ ns. Strong resonant $dd\mu$ formation in ortho-D₂ has been recently observed at RIKEN-RAL Muon Facility and at TRIUMF [44]. The effective formation rate in ortho-D₂, derived from these experiments, is lower by about 25% than an experimental value of the rate found in a statistical mixture. These results confirm our calculations, which predict a somewhat lower fusion-neutron yield for targets containing more ortho-D₂ molecules.

VII. CONCLUSIONS

The methods used to describe resonant neutron and γ -ray absorption in condensed matter have been directly applied to the 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 response function, which depends on the 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 harmonic solid deuterium. The calculations show great differences between resonant $dd\mu$ formation in 3-K solid deuterium and in 3-K

D_2 gas. In solid deuterium, the formation at a few meV, which determines the experimental results, is dominated by the presence of strong recoil-less resonant peaks. On the other hand, 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 D_2 gas, but with the effective temperature equal to 40 K. This temperature is determined by the energy of zero-point vibration of a D_2 molecule in the lattice. Phonon creation is always important in the case of $dd\mu$ back decay because it is connected with an energy release of a few tens of meV, which is much greater than the Debye energy.

The 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 D_2 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 . As a result, the resonant $dd\mu$ 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 $T \geq 4$ K have an hcp structure [35]. Though our calculations have been performed for fcc crystals, the results obtained 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 $d\mu$ deceleration below the Debye energy is very slow and that mean energy of the $d\mu$ ($F = \frac{3}{2}$) atom is always significantly greater than 1 meV. The energy distribution of $d\mu$'s during their lifetime is very broad (at least a few meV), therefore a strong overlap of this distribution with the lowest resonance peaks takes place, leading to a large mean $dd\mu$ formation rate in solid deuterium. However, an explanation of the experiments is possible only if the broadening of rotational-vibrational molecular levels in the solid is taken into account. We obtained reasonable results assuming that the strengths of the recoil-less 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 gives only about a 10% contribution to the calculated neutron time spectra.

The dd fusion-neutron spectra calculated for ortho- D_2 and para- D_2 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 $d\mu$ energy in para- D_2 . These differences can be clearly seen only in high-statistics experiments. Our calculations and recent experiments [44] do not confirm the lack of strong resonant $dd\mu$ formation in solid ortho- D_2 predicted in papers Refs. [20,21].

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