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THEORY OF MOLECULAR TRANSFORMATIONS INDUCED BY NEUTRONS

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A general method is developed for investigating the probabilities of molecular transformations induced by neutron impact. The method is based on an application of the impulse approximation to secondary collisions of the atoms ejected by the neutrons with the other atoms of the molecule. The method is used to calculate the probability of dissociation of triatomic molecules by neutrons accompanied by excitation of the diatomic residue.

1. INTRODUCTION

THE collision between a sufficiently fast neutron and a nucleus in a molecule is accompanied by diverse transformations of this molecule. In this article we develop a theory for such processes, based on the following assumptions:

1. The transformations of the molecule are such that the molecule remains in the electronic ground state. In other words, the atom knocked out by the neutron and also the molecular residue, are not electron-excited. This assumption is valid at neutron energies of the order of the energies of the chemical bonds, if the electronic ground state term does not cross or does not come close to the term of its excited state.

2. The chemical transformation in a polyatomic molecule M, one of whose atoms acquires from the neutron some momentum κ, has the following character: the atom A_1 knocked by the neutron collides with another atom A_2 of the molecule, thereby causing a transformation of the molecular residue $M - A_1$ (excitation or dissociation), and goes off to infinity. In this scheme of the process it is assumed, obviously, that the atoms A_1, which received the momentum from the neutron, are "peripheral" and have a valence bond only with one of the atoms of the residue $M - A_1$ (the terminal atoms in linear molecules, the H atoms in the H_2O molecule, etc., are of this type). We also exclude from consideration such processes as the pick-up of an atom A_1 by an atom A_2 or the substitution of A_2 for A_1 in the residue $M - A_2$, which are possible in principle.

3. The interactions between the atom A_1 and the other atoms of the molecule have a paired character, and the collisions between the atoms A_1 and A_2 are impulsive, i.e., they are characterized by the inequality $A_{1-2}\Delta\epsilon/hv \ll 1$, where A_{1-2}—radius of interaction of the atoms A_1 and A_2, $\Delta\epsilon$—difference of the vibrational levels in the residue molecule, and v—relative velocity of the atoms A_1 and A_2. Both conditions are satisfied starting with $E_{A_1} \gtrsim 10$ eV, which thus defines the region of applicability of our theory.

In this connection it must be noted that an attempt was made in [1] to calculate the probabilities of the processes that we are investigating in terms of "reduced widths" $\Theta(\rho)$, borrowed from nuclear spectroscopy, the statement being made that the region of applicability of the theory in [1] begins with an energy E_{A_1} on the order of several times ten electron volts. Actually (see the end of Sec. 2), as follows from our general formula (14), the disintegration of the molecule, accompanied by some transformation of the residue $M - A_1$, has a cross section that can be expressed in terms of $\Theta(\rho)$ by the formula $\sigma = \int |\Theta(\rho)|^2 d\rho$ (ρ—radius of the reaction channel) only in the limiting case of very high energies $E_{A_1} \gg D^2/\Delta\epsilon$ [D—energy of the dissociation $M \rightarrow A_1 + (M - A_1)$], i.e., only in the limiting Born region of incident-particle energies of the order of 1000 eV, where the probabilities of excitation of the molecular residues are independent of the energy. The most interesting energy region $E_{A_1} \sim D^2/\Delta\epsilon$, in which these probabilities have maxima as functions of E_{A_1} (in accordance with our calculations), is thus not included in the above-mentioned theory.

2. GENERAL METHOD

Assume that a fast neutron collides with one of the atoms (designated by the number 1) of a three-atom molecule, and transfers to the molecule enough energy to break the bonds, so that the molecule goes over as a result of the collision into a continuous-spectrum state $\psi_{\mathrm{k}_1\mathrm{b}}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ ($\mathbf{r}_1$, $\mathbf{r}_2$, $\mathbf{r}_3$—coordinates of the nuclei of the atoms of the molecule), such that

$$\psi_{\mathbf{k}_1 b}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \rightarrow \Phi_{\mathbf{k}_1}(\mathbf{r}_1)\,\varphi_b(\mathbf{r}_2, \mathbf{r}_3) \quad (r_1 \rightarrow \infty),$$

where $\varphi_{\mathrm{b}}(\mathbf{r}_2, \mathbf{r}_3)$—wave function of the excited molecular residue, and $\Phi_{\mathrm{k}_1}(\mathbf{r}_1)$—function that consists as $\mathbf{r}_1 \to \infty$ of a plane and converging spherical wave. The function $\psi_{\mathrm{k}_1\mathrm{b}}$ corresponds, obviously, to the knock-out of a recoil atom with final momentum $\mathbf{k}_1$. The type of the excitation of the molecular residue is not specified here [$\varphi_{\mathrm{b}}(\mathbf{r}_2, \mathbf{r}_3)$ can correspond also to disintegration of this residue].

We denote the wave function of the initial molecule by $\psi_0(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$. The cross section of the process in the Fermi pseudopotential approximation is (see, for example [2]; we use a system of units in which h = 1)

$$d\sigma_1 = a_1^2 \frac{k}{k_0} \left| \langle \psi_{\mathbf{k}_1 b}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)\, e^{i\varkappa \mathbf{r}_1} \psi_0(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \rangle \right|^2 d\mathbf{k}_1 do_n, \tag{1}$$

where a_1—amplitude for the scattering of neutrons by atom 1 which is rigidly fixed in an infinite mass; $\mathbf{k}_0$ and $\mathbf{k} = \mathbf{k}_0 - \kappa$—neutron momentum before and after the scattering; o_{n}—solid angle of the scattered neutrons.

The function $\psi_{\mathrm{k}_1\mathrm{b}}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ with the above-mentioned asymptotic property describes (accurate to within the sign of $\mathbf{k}_1$ and the complex conjugation operation) the scattering of particle 1 by the system of atoms 2 and 3, which makes up the residue molecule. As shown earlier [3], in the case of sufficiently high incident-atom energies, for which the condition $A\Delta\epsilon/v \ll 1$ is satisfied, it is possible to use the impulse approximation to describe the collision of A_1 with the residue $M - A_1$. As applied to our problem, this means that we can write down the wave function of the final state of the molecule in the impulse approximation [4]:

$$\psi_{\mathbf{k}_1 b}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) = \iint \varphi_b(\mathbf{p}_2, \mathbf{p}_3) \left[\exp\left(i\frac{\mathbf{k}_1\mathbf{r}_1 + \mathbf{p}_2\mathbf{r}_2}{m_1 + m_2} \right) \right.$$
$$\times \psi_{\mathbf{q}_2}(\mathbf{r}_1 - \mathbf{r}_2)\, e^{i\mathbf{p}_3\mathbf{r}_3} + \exp\left(i\frac{\mathbf{k}_1\mathbf{r}_1 + \mathbf{p}_3\mathbf{r}_3}{m_1 + m_3} \right)$$
$$\left. \times \psi_{\mathbf{q}_3}(\mathbf{r}_1 - \mathbf{r}_3)\, e^{i\mathbf{p}_2\mathbf{r}_2} \right] d\mathbf{p}_2 d\mathbf{p}_3,$$
$$\mathbf{q}_2 = (m_2\mathbf{k}_1 - m_1\mathbf{p}_2)\,/\,(m_1 + m_2),$$
$$\mathbf{q}_3 = (m_3\mathbf{k}_1 - m_1\mathbf{p}_3)\,/\,(m_1 + m_2). \tag{2}$$

From now on replacement of $\mathbf{r}_2$ or $\mathbf{r}_3$ with the momentum $\mathbf{p}_2$ or $\mathbf{p}_3$ in the functions φ_{b} and ψ_0 denotes that the Fourier transform in the corresponding variable was taken. The function $\psi_{\mathrm{q}}(\rho)$ describes the paired scattering of the recoil atom and one of the atoms of the molecular residue in their center-of-mass system.

Let us consider for concreteness the case when the final state is produced as a result of interaction between atoms 1 and 2. The corresponding matrix element takes after simple transformations the form

$$\iiiint\!\!\int \varphi_b^*(\mathbf{p}_2, \mathbf{r}_3) \exp\{-i(\mathbf{p}_2 - \varkappa_1 - \mathbf{p}_2')\mathbf{r}_2\}\, \psi_{\mathbf{q}}(\boldsymbol{\rho})\, e^{i\varkappa'\rho}$$
$$\times \psi_0(\boldsymbol{\rho}, \mathbf{p}_2', \mathbf{r}_3)\, d\mathbf{p}_2\, d\mathbf{p}_2'\, d\boldsymbol{\rho}\, d\mathbf{r}_2\, d\mathbf{r}_3,$$
$$\varkappa_1 = \varkappa - \mathbf{k}_1,\ \ \mathbf{q} = \frac{m_2\mathbf{k}_1 - m_1\mathbf{p}_2}{m_1 + m_2}, \quad \varkappa' = \frac{m_2\varkappa - m_1\mathbf{p}_2'}{m_1 + m_2}. \tag{3}$$

Under quasiclassical conditions, which are valid for the collision of arbitrary sufficiently fast atoms, the function $\psi_{\mathrm{q}}(\rho)$, which describes the paired scattering, can be represented by some sum over the classical trajectory [5]:

$$\psi_{\mathbf{q}}(\boldsymbol{\rho}) = \sum_\lambda A_\lambda e^{iS_\lambda}, \tag{4}$$

where λ is a parameter that classifies the trajectory. The sum in (4) extends over all trajectories that arrive from infinity (in the direction of $\mathbf{q}$) at the point ρ:

$$S_\lambda = \int^{\rho} g_\lambda'\, dl, \quad g_\lambda' = [2\mu(E - V(\boldsymbol{\rho}'))]^{1/2},$$

$V(\rho)$—potential energy of the atoms 1 and 2; $E = q^2/2\mu$; $\mu = m_1m_2/(m_1 + m_2)$. The integration is carried out along a trajectory determined from the classical Hamilton-Jacobi equation

$$(\nabla S)^2 = 2\mu(E - V),$$
$$A_\lambda = [q\,/\,g_\lambda'(\boldsymbol{\rho})]^{1/2}[df_0\,/\,df_\lambda]^{1/2},$$

where df_0 and df_λ—cross sections of the bundle of trajectories at $\mathbf{q}\cdot\mathbf{p}' \to \infty$ and in the vicinity of the point ρ, respectively.

The main contribution of the integral

$$\sum_\lambda \int \psi_0(\boldsymbol{\rho}, \mathbf{p}_2', \mathbf{r}_3)\, e^{i\varkappa'\rho} e^{-iS_\lambda} d\boldsymbol{\rho} \tag{5}$$

is determined by values of ρ close to ρ_0, corresponding to spatial localization of particles 1 and 2 in the initial state of the molecule (the orientation of which is assumed, as usual, to be fixed if the energy transfer from the neutron to the molecule is large). The integration over ρ therefore selects, from among all the trajectories, only those close to ρ_0. The values of ρ_0 correspond to the minimum of the potential energy $V(\rho)$. For trajectories close to ρ_0 the phase integrals S_λ can be written in the form

$$S_\lambda = (\boldsymbol{\rho} - \boldsymbol{\rho}_0)\mathbf{g}_\lambda + B_\lambda(\boldsymbol{\rho}_0), \qquad \mathbf{g}_\lambda = \mathbf{g}_\lambda'(\boldsymbol{\rho}_0),$$
$$|\mathbf{g}_\lambda|^2 = q^2 + 2\mu D, \tag{6}$$

where D—value of the minimum of the potential curve $V(\rho)$ relative to $V(\infty)$ and $B_\lambda(\rho_0)$—large real quantity that depends strongly on λ.

Taking (6) into account, we now substitute (4) in the expression (3) for the matrix element and integrate in that formula with respect to the variable $\boldsymbol{\rho}$, obtaining

$$\sum_\lambda A_\lambda(\boldsymbol{\rho}_0)\exp[-iB_\lambda(\boldsymbol{\rho}_0)]\iiiint\varphi_b^*(\mathbf{p}_2,\mathbf{r}_3)$$
$$\times\exp\{-i(\mathbf{p}_2-\boldsymbol{\varkappa}_1-\mathbf{p}_2')\mathbf{r}_2\} \tag{7}$$
$$\times\psi_0(\mathbf{p}_1=\mathbf{g}_\lambda-\boldsymbol{\varkappa}',\mathbf{p}_2',\mathbf{r}_3)\,d\mathbf{p}_2\,d\mathbf{p}_2'\,d\mathbf{r}_2\,d\mathbf{r}_3.$$

The summation over λ is equivalent to integration over the directions of the vector $\mathbf{g}_\lambda$. It is easy to verify that when $|\mathbf{g}_\lambda|^2$ and $\kappa'^2 \gg p_1^2$, this corresponds in turn to integration with respect to $\mathbf{p}_1$ under the condition

$$g_\lambda^2=|\boldsymbol{\varkappa}'+\mathbf{p}_1|^2\approx\varkappa'^2+2\boldsymbol{\varkappa}'\mathbf{p}_1.$$

If we now substitute here the expressions for g_λ^2 and κ' from (3) and (6) and divide the result by 2μ, we find that integration with respect to $\mathbf{p}_1$ in (7) occurs at a fixed value

$$\varkappa^2/2m_1+\boldsymbol{\varkappa}\mathbf{p}_1/m_1=k_1^2/2m_1+\varkappa_1^2/2m_2+\boldsymbol{\varkappa}_1\mathbf{p}_2'/m_2+D, \tag{8}$$

which corresponds precisely to the impulse conditions for the transfer of energy from the neutron to the atom 1 ($\epsilon=\kappa^2/2m_2+\boldsymbol{\kappa}\cdot\mathbf{p}_1/m_1$) and from atom 1 to atom 2 ($\epsilon_b=\kappa_1^2/2m_2+\boldsymbol{\kappa}_1\cdot\mathbf{p}'_2/m_2$); we recall that the energy conservation law takes the form

$$\varepsilon=\varepsilon_b+k_1^2/2m_1+D. \tag{9}$$

If the excitation of the residue molecule is such that $|\epsilon_b-\kappa_1^2/2m_2|\ll\epsilon$ (this corresponds to the impulse approximation in its simplified treatment of the collision between the recoil atom and the molecular residue, when the law of paired interaction of atoms 1 and 2 can be regarded as independent of the initial momentum of the scattering atom 2 [3]), the condition (8) with

$$\varepsilon=\varkappa^2/2m_1+\boldsymbol{\varkappa}\mathbf{p}_1/m_1 \tag{9a}$$

corresponds simply to the law of energy conservation. Taking this circumstance (the fact that $\mathbf{p}_1$ is independent of $\mathbf{p}'_2$) into account, we can write down the result of integration (7) with respect to $\mathbf{p}_2$ and $\mathbf{p}'_2$ in the form

$$\sum_\lambda A_\lambda(\boldsymbol{\rho}_0)\exp[-iB_\lambda(\boldsymbol{\rho}_0)]\iint\varphi_b^*(\mathbf{r}_2,\mathbf{r}_3)\,e^{i\boldsymbol{\varkappa}_1\mathbf{r}_2}$$
$$\times\psi_0(\mathbf{p}_1=\mathbf{g}_\lambda-\boldsymbol{\varkappa}',\mathbf{r}_2,\mathbf{r}_3)\,d\mathbf{r}_2d\mathbf{r}_3.$$

In calculating the probability (the square of the modulus of the matrix element) it is necessary to retain in the sum $\Sigma_{\lambda\lambda'}$ only the terms that are diagonal in λ and λ', inasmuch as all the other terms oscillate rapidly when λ is varied, i.e., we have for the scattering cross section

$$d\sigma_1=a_1^2\frac{k}{k_0}\sum_\lambda|\langle\varphi_b(\mathbf{r}_2,\mathbf{r}_3)\,e^{i\varkappa_1\mathbf{r}_2}$$
$$\times\psi_0(\mathbf{p}_1=\mathbf{g}_\lambda-\boldsymbol{\varkappa}',\mathbf{r}_2,\mathbf{r}_3)\rangle|^2\,d\mathbf{k}_1\,do_n.$$

As already mentioned, averaging over the trajectories that lie near ρ_0 corresponds to integration with respect to do_n, or to integration with respect to the variable $\mathbf{p}_1$ under the condition (9a). Taking all this into account, we arrive at the following expression for the cross section for knocking out atom 1 and producing a specified excitation of the molecular residue (for arbitrary molecules, $\mathbf{r}_3$ should be taken to mean the coordinates that make up together with $\mathbf{r}_2$ the entire set of coordinates of the molecular residue):

$$\frac{d^2\sigma_1}{d\varepsilon do_n}=a_1^2\frac{k}{k_0}\int|\langle\varphi_b(\mathbf{r}_2,\mathbf{r}_3)\,e^{i\varkappa_1\mathbf{r}_2}\,\psi_0(\mathbf{p}_1,\mathbf{r}_2,\mathbf{r}_3)\rangle|^2$$
$$\times W(\boldsymbol{\varkappa},\mathbf{k}_1,\boldsymbol{\rho}_0)\,\delta\left(\varepsilon-\frac{\varkappa^2}{2m_1}-\frac{\boldsymbol{\varkappa}\mathbf{p}_1}{m_1}\right)d\mathbf{p}_1\,do_{\mathbf{k}_1}\,do_\varkappa, \tag{10}$$

$$W(\boldsymbol{\varkappa},\mathbf{k}_1,\boldsymbol{\rho}_0)=\frac{1}{4\pi}\left[\frac{df_0}{df_{\lambda\,(\rho_0)}}\right]\left(\frac{m_1+m_2}{m_1}\right)^2\frac{k_1'}{\varkappa'}\frac{k_1}{\varkappa},$$

$$\mathbf{k}_1'=\frac{m_2\mathbf{k}_1-m_1\boldsymbol{\varkappa}_1}{m_1+m_2},\quad \boldsymbol{\varkappa}'=\frac{m_2}{m_1+m_2}\boldsymbol{\varkappa}. \tag{11}$$

Here df_0/df_λ—dimensionless quantity which coincides as $\rho\to\infty$ with $d\sigma/\rho^2d\Omega$, where $d\sigma/d\Omega$ is the classical differential cross section of the paired scattering of the atoms.

From the differential neutron scattering cross section (10) we can go over to the total cross section σ_{1f} of an inelastic process of a definite type which is of greatest interest. In finding this cross section we can always neglect the momentum spread of the atom A_1, as a result of which σ_{1f} takes the form

$$\sigma_{1f}=a_1^2\cdot4\pi\left(\frac{m_1}{m_1+m}\right)^2\int w_f(\varepsilon)\frac{d\varepsilon}{E_{max}},$$
$$w_f=\int W(\boldsymbol{\varkappa},\mathbf{k}_1,\boldsymbol{\rho}_0)\,P(\boldsymbol{\varkappa}_1)\,do_\varkappa\,do_{\mathbf{k}_1}. \tag{12}$$

where

$$P(\boldsymbol{\varkappa}_1)=\int\left|\int\varphi_b^*(\mathbf{r}_2,\mathbf{r}_3)\,e^{i\varkappa_1\mathbf{r}_2}\,\psi_0(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)\,d\mathbf{r}_2\,d\mathbf{r}_3\right|^2d\mathbf{r}_1,$$

$$\varepsilon=\frac{\varkappa^2}{2m_1},\ \boldsymbol{\varkappa}_1=\boldsymbol{\varkappa}-\mathbf{k}_1,\ E_{max}=4\frac{m_1m}{(m_1+m)^2}\frac{k_0^2}{2m}$$

(m—neutron mass). The quantity w_f, which has the meaning of the probability that the transfer of energy from the neutron to the atom A_1 will result in some transformation of the residue, is naturally the probability W of the transfer of momentum κ_1 from atom A_1 to A_2 multiplied by the probability P of some transition in the residue, both probabilities being averaged over the directions of $\mathbf{k}_1$ and $\boldsymbol{\kappa}$.

The energy conservation law takes after these simplifications the form

$$\varepsilon = \frac{\varkappa^2}{2m_1} = D + \frac{k_1^2}{2m_1} + \frac{\varkappa_1^2}{2(m_2+m_3)} + \varepsilon_b', \tag{13}$$

where ϵ'_b—value of the internal energy transfer to the molecular residue. The final momentum $\mathbf{k}_1$ of the atom A_1 is determined uniquely from its initial value $\boldsymbol{\kappa}$ by using the law of paired interaction of the atoms A_1 and A_2, after which the integration limits in (4) are determined from the roots of (13).

The quantity $P = \int |M|^2 d\mathbf{r}_1$ can be simplified by introducing the internal (Jacobian) coordinates of the molecule. As applied to a triatomic molecule these coordinates ρ_1 and ρ' are expressed in terms of $\mathbf{r}_1$, $\mathbf{r}_2$, and $\mathbf{r}_3$ as follows:

$$\boldsymbol{\rho}_1 = \mathbf{r}_2 - \mathbf{r}_3, \qquad \boldsymbol{\rho}' = \mathbf{r}_1 - \mathbf{R},$$

$$\mathbf{R} = (m_2\mathbf{r}_2 + m_3\mathbf{r}_3) / (m_2 + m_3).$$

The result of the transformation can be written in the form

$$P = \int |M|^2 d\boldsymbol{\rho}',$$

$$M = \int \varphi_b^* (\boldsymbol{\rho}_1) \exp\left(-i\frac{m_3}{m_3+m_2}\boldsymbol{\varkappa}_1\boldsymbol{\rho}_1\right) \psi_0(\boldsymbol{\rho}_1, \boldsymbol{\rho}')\, d\boldsymbol{\rho}_1. \tag{14}$$

If the coordinates ρ_1 and ρ' coincide approximately with the normal coordinates (this is the situation, for example, if $m_3 \gg m_1$ or m_2—the case of H_2O), then $\psi_0 = \varphi_0(\rho_1)\varphi'_0(\rho')$, and consequently

$$P = \left|\int \varphi_b^* (\boldsymbol{\rho}_1) \exp\left(-i\frac{m_3}{m_3+m_2}\boldsymbol{\varkappa}_1\boldsymbol{\rho}_1\right) \varphi_0(\boldsymbol{\rho}_1) d\boldsymbol{\rho}_1\right|^2. \tag{15}$$

Let us consider in greater detail the case of large energies $\epsilon \gg D$. Under this condition we can separate two essentially different processes:

1. The neutron transfers to the atom A_1 a momentum κ in a direction such that A_1 does not come in close contact with the other atoms of the molecule. The role of "collision" is played in this case simply by the motion of A_1 in the field of the atom with which it has a valence bond (we shall denote it by A_2).

At the high energies in question, we can assume the trajectory of A_1 to be a straight line, i.e., $\mathbf{k}_1$ coincides with κ in direction. The momentum $\kappa_1 \ll \kappa$, transferred to A_2 in a direction parallel to $\mathbf{k}_1$, can be approximately determined by neglecting in (13) the term $\kappa_1^2/2(m_2 + m_3)$, i.e.,

$$\varkappa_1^{\parallel} = \varkappa - k_1 = \varkappa - \sqrt{\varkappa^2 - \varkappa_0^2} \approx \varkappa_0^2 / 2\varkappa,$$

$$\varkappa_0^2 = 2m_1(D + \varepsilon_b'). \tag{15a}$$

The component $\kappa_1^{\perp}$ perpendicular to $\mathbf{k}_1$ is given by the general formula for the transfer of momentum at high energies of the colliding particles (see [6], p. 74):

$$\varkappa_1^{\perp} = \frac{m_1\delta}{\varkappa}\int\limits_{\rho_0}^{\infty} \frac{dV(\rho)}{d\rho}\frac{dl}{\rho}, \tag{16}$$

where $V(\rho)$—potential of interaction of particles 1 and 2, δ—an impact parameter uniquely connected with the direction of κ, and l—coordinate reckoned along the trajectory of particle A_1.

For approximately linear trajectories we can assume also in the definition of W(11) that

$$\left(\frac{m_1+m_2}{m_1}\right)^2 \frac{df}{df_\lambda} = \delta(o_\varkappa - o_{\mathbf{k}_1})$$

and average in (12) only over the directions of $\boldsymbol{\kappa}$ and $\mathbf{k}_1$; this is equivalent to averaging over the orientations of the molecule (symbol $\langle\,\rangle_\Omega$) at specified directions of the neutron before and after scattering, i.e.,

$$w_f = \langle P(\boldsymbol{\varkappa}_1)\rangle_\Omega, \tag{17}$$

where $P(\kappa)$ is defined in (14) and (15), and the components of κ_1 are given in (16) and (15a). The limits of integration with respect to ϵ in (12) can be taken to be

$$\frac{m_1+m_2+m_3}{m_2+m_3}(D+\varepsilon_b') \text{ and } E_{max}.$$

The effective region of variational coordinate in (16) is of the order of magnitude of the molecular vibrations $1/\sqrt{\mu_{23}\omega}$. Therefore at very high neutron energies, such that

$$\frac{m_3}{m_2+m_3}\frac{\varkappa_0^2}{\varkappa\sqrt{\mu_{23}\omega}} \ll 1 \qquad \left(\mu_{23} = \frac{m_2m_3}{m_2+m_3}\right)$$

we can assume in (14) that the exponential is equal to unity and M coincides with the reduced width

$$\Theta(\boldsymbol{\rho}') = \int \varphi_b^*(\boldsymbol{\rho}_1)\psi_0(\boldsymbol{\rho}_1, \boldsymbol{\rho}')\, d\boldsymbol{\rho}_1.$$

In other words, the quantity w_f does not depend on this case on the energy and can be written in the form

$$w_f = \frac{\Omega}{4\pi}\int |\Theta|^2 d\boldsymbol{\rho}', \tag{18}$$

where Ω—effective region of the angles in which the foregoing treatment is applicable.

Formula (18) coincides, apart from a factor $\Omega/4\pi$, with the analogous result of Matthies and Neudachin [1] which, consequently, can be regarded as correct to a certain degree only in the limiting case of very high energies (of the order of keV), satisfying the condition

$$\frac{m_3}{m_2+m_3}\frac{\varkappa_0^2}{\varkappa\sqrt{\mu_{23}\omega}} \ll 1, \quad \text{i.e.,} \quad \varepsilon \gg \frac{m_3}{m_3+m_2}\frac{m_1}{m_2}\frac{D^2}{\omega}.$$

To the contrary, formulas (12) and (17) are valid also in the intermediate region $\epsilon \lesssim D^2/\omega$, where the probabilities $w_f(\epsilon)$ have maxima.

2. A different type of process takes place if the neutron transfers to the atom A_1 a momentum in a direction such that the atom comes in close contact with the atom A_2. In this case, as can be readily seen, the quantity W is expressed simply in terms of the cross section $\sigma(\kappa', k'_1)$ of the paired elastic scattering of atoms A_1 and A_2, i.e.,

$$W = \left(\frac{m_1+m_2}{m_1}\right)^2 \frac{\sigma(\varkappa', \mathbf{k}_1')}{4\pi\rho_0^2}. \tag{19}$$

The probability $w_f(\epsilon)$ is given by the formula

$$w_f(\varepsilon) = \frac{k_1}{\varkappa}\int \left(\frac{m_1+m_2}{m_1}\right)^2 \frac{\sigma(\varkappa', \mathbf{k}_1')}{4\pi\rho_0^2} P(\varkappa_1)\, do_{\mathbf{k}_1}. \tag{20}$$

The probabilities determined in this manner can usually be regarded as small compared with those given by (17), in a ratio on the order of $\sigma/4\pi\rho_0^2$. Therefore formula (17) yields in many cases what is essentially the total probability of the process. An exception in this respect is the case when the coordinates ρ and ρ' are close to normal, and the value of P at sufficiently high energies is equal to zero for collisions in which the momentum of the atom A_1 changes little. In this case nearly-frontal collisions connected with large transfer of momentum from A_1 to A_2, can become significant. Such a situation obtains, as already noted, when the neutrons knock out H atoms from H_2O molecules. We note also that if P is of the form (15), the probability (20) coincides simply with the probability of transformation of the residue as a result of collisions between the atom A_1, which is incident from infinity, with A_2.

This illustrative picture, as can be seen from the foregoing, is valid only under the special assumption that ρ_1 and ρ' coincide with the normal coordinates. It does hold, however, for water molecules. Therefore the cross section for the process whereby the neutron knocks out a hydrogen atom from the water molecule, accompanied by vibrational excitation of OH, can be directly determined from the general formulas [7] for the probabilities of vibrational excitation of diatomic molecules by sufficiently fast atoms, assuming that the excitation of the OH is produced by a hydrogen atom incident from infinity.

3. COLLISIONS WITH LINEAR TRIATOMIC MOLECULE

We shall investigate the more complicated laws that apply when the 'reduced widths'

$$\Theta = \int \varphi_b(\boldsymbol{\rho}_1)\,\psi_0(\boldsymbol{\rho}_1, \boldsymbol{\rho}')\,d\boldsymbol{\rho}_1$$

differ from zero, using as an example the collision between a neutron and the atom A_1 of a linear triatomic molecule, accompanied by vibrational excitation of its diatomic residue A_2A_3. The probability $w_n(\epsilon)$ of such a vibrational excitation of A_2A_3 is given by formula (17), which now must be written in the form

$$P = \int \left| \int \varphi_b^*(\rho_1)\, e^{-i\tilde{\varkappa}\rho_1}\, \psi_0(\rho_1, \rho')\, d\rho_1 \right|^2 d\rho',$$

$$\tilde{\varkappa} = \frac{m_3}{m_3+m_2}\left(\varkappa_1^{\parallel}\cos\vartheta + \varkappa_1^{\perp}\sin\vartheta\right), \tag{21}$$

where, in accord with (15) and (16), ϑ is the angle between the direction of emission of A_1 and the molecule axis.

Using the oscillator wave functions for the initial state of the molecule $A_1A_2A_3$

$$\psi_0 = \left(\frac{\omega_1}{\pi}\right)^{1/4} \exp\left(-\frac{\omega_1 u_1^2}{2}\right)\left(\frac{\omega_2}{\pi}\right)^{1/4} \exp\left(-\frac{\omega_2 u_2^2}{2}\right)$$

(u_1u_2—generalized coordinates), of the final state of A_2A_3

$$\varphi_b = (2^n n!)^{-1/2}\left(\frac{\gamma}{\pi}\right)^{1/4}$$
$$\times \exp\left[-\frac{1}{2}\gamma(\rho_1-\rho_{10})^2\right] H_n\left(\sqrt{\gamma}(\rho_1-\rho_{10})\right),$$

$$\gamma = \mu_{23}\omega_b$$

and known tabulated integrals [8], we reduce (21) to the form

$$w_n(\varepsilon) = A\,\frac{(\beta^2-1)^n}{2^n n!}\int_{-\infty} \exp\left\{-p\tilde{\varkappa}^2 - (qt - r\tilde{\varkappa})^2\right\}$$
$$\times H_n\left(\frac{\beta}{2\sqrt{\beta^2-1}}\,t\right)dt \tag{22}$$

where

$A = (2/\pi)^{1/2}\sqrt{\gamma/\gamma'}\,|\alpha_1\beta_2 - \alpha_2\beta_1|/c,$

$\gamma' = \nu + 2a - c^2/2b, \qquad a = {}^1\!/_2(\alpha_1^2 + \alpha_2^2),$

$b = {}^1\!/_2(\beta_1^2 + \beta_2^2),$

$c = |\alpha_1\beta_1 + \alpha_2\beta_2|, \quad \beta = \sqrt{2\gamma/\gamma'}, \quad p = 1/(\gamma + 2a),$

$q = [b(\gamma+2a)/c^2]^{1/2}, \quad r = [2\gamma' b/(\gamma+2a)c^2]^{1/2},$

$$\alpha_{1,2} = -\frac{1}{\eta}c_{1,2}^{(2,1)}\frac{m_3}{m_2+m_3},$$

$$\beta_{1,2} = \frac{1}{\eta}\frac{m_2+m_3}{m_1+m_2+m_3}\left(\frac{m_3}{m_3+m_2}c_{1,2}^{(2,1)} \pm c_{2,1}^{(2,1)}\right),$$

$$\eta = c_1^{(1)}c_2^{(2)} + c_1^{(2)}c_2^{(1)},$$

$c_{1,2}^{(1,2)}$ are the components of the amplitude vectors of the molecule $A_1A_2A_3$.

In the limiting case when $\tilde{\kappa} = 0$ (when the generalized formula (18) is valid) we get from (22)

$$w_n = A\left[\frac{\beta^2-1}{1-s^2/2}\right]^{1/2}\frac{s}{\beta}\frac{\Gamma(n+1/2)}{n!}$$

$$\times(\beta^2-1)^n(s^2-1)^n F\left(\frac{1}{2}, \frac{1}{2}, -n+\frac{1}{2}, \frac{1}{2-s^2}\right),$$

$$s^2 = \frac{1}{2q^2}\frac{\beta^2}{\beta^2-1}, \tag{23}$$

with[1] $1 < s^2$ and $\beta^2 < 2$.

According to (23) w_n decreases monotonically with increasing n. As $n \to \infty$ we obtain from (23)

$$w_n = A\left[\frac{\beta^2-1}{1-s^2/2}\right]^{1/2}\frac{s}{\beta}\frac{1}{\sqrt{n}}(\beta^2-1)^n(s^2-1)^n. \tag{24}$$

In the general case when $\tilde{\kappa} \neq 0$ the energy dependence of $w_n(\epsilon)$ takes the form

$$w_n(\varepsilon) = A\frac{(\beta^2-1)^n}{2^n n!}\frac{\sqrt{\pi}}{q}\sum 2^k k!\binom{n}{k}^2\left(1-\frac{s^2}{2}\right)^{n-k}$$

$$\times\left\langle e^{-p\tilde{\varkappa}^2}H_{2n-2k}\left(\frac{sr\tilde{\varkappa}}{\sqrt{2-s^2}}\right)\right\rangle_\Omega. \tag{25}$$

In the approximate calculations we can replace the averaging over the molecule orientation in (15) by introducing a quantity $\tilde{\kappa}_{av} = \langle\tilde{\kappa}^2\rangle_\Omega^{1/2}$ averaged over the orientation. As can be readily shown, the probability w_n defined in this manner decreases exponentially when $\epsilon < D^2/\omega$, tends to a constant quantity given by (23) as $\epsilon \to \infty$, and has a maximum in the interval about $\epsilon \sim D^2/\omega$. In the particular case when n = 1 this dependence is given by the formula

$$w_n \sim \exp(-p\tilde{\varkappa}_{av}^2)(r^2\tilde{\varkappa}_{av}^2 + 1/2),$$

which has a maximum at $\tilde{\kappa}_{av} = (r^2/2p)^{1/2}$.

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[1] For the CO_2 molecule the numerical values of the parameters are $s^2 = 1.57$, $\beta^2 = 1.475$, $p = 0.196\ (m\cdot eV)^{-1}$, and $r = 0.44\ (m\cdot eV)^{-1/2}$.

[1] S. Matthies and V. G. Neudachin, JETP **45**, 131 (1963), Soviet Phys. JETP **18**, 95 (1964).

[2] L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Fizmatgiz, 1963, pp. 666 and 677.

[3] G. K. Ivanov and Yu. S. Sayasov, JETP **45**, 1456 (1963), Soviet Phys. JETP **18**, 1006 (1964).

[4] G. F. Chew, Phys. Rev. **80**, 190 (1950).

[5] I. I. Gol'dman and A. B. Migdal, JETP **28**, 394 (1955), Soviet Phys. JETP **1**, 304 (1955).

[6] L. D. Landau and E. M. Lifshitz, Mekhanika (Mechanics), Fizmatgiz, 1958.

[7] G. K. Ivanov and Yu. S. Sayasov, DAN SSSR **154**, 1314 (1964), Soviet Phys. Doklady **9**, 171 (1964).

[8] I. S. Gradshteĭn and I. M. Ryzhik, Tablitsy integralov (Tables of Integrals), Fizmatgiz, 1963.

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