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The effect of creation and simultaneous destruction of intravibrational and rotational phonons of methane molecules caused by thermal neutron collisions

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Abstract. Via the quantum mechanical description of thermal neutron scattering at the hydrogen atom nucleus of the methane molecule, a new effect is discovered showing that a neutron with insufficient energy for creating one intravibrational phonon creates the phonon by simultaneous destruction of another intravibrational phonon of the methane molecule. The effect occurs not only with combined inelastic thermal neutron collisions on the intravibrational phonons of the methane molecule but also with combined inelastic scattering at the rotational and intravibrational phonons. Evidently this effect exists theoretically for all incident thermal neutron energies $E_0 \in [1 \text{ meV}, 100 \text{ meV}]$ investigated.

1. Introduction

The interaction of thermal neutrons with atomic nuclei bound in molecules has been studied by many researchers. It is well known that the interaction can be successfully described by applying the Fermi (1936) pseudopotential interaction in Born's first approximation. Starting from this fact, Zemach and Glauber (1956) developed the general operator formalism of quantum mechanics for the description of the interaction of thermal neutrons with bound atoms. The incoherent approximation in the Zemach-Glauber formalism yields an expression for the microscopic thermal neutron kernel scattering on the hydrogen atom nucleus bound in the methane molecule (CH_4) in the following form (in the units system where Planck's constant is unity and the neutron mass $m_n = 1$):

$$\sigma(E_0 \rightarrow E, \nu) = \frac{d^2\sigma}{dE d\nu} = \frac{\sigma_b}{4\pi} \sqrt{\frac{E}{E_0}} \int_{-\infty}^{+\infty} \exp(i^* \epsilon t) \langle \chi(\kappa, t) \rangle_{\Omega} dt \quad (1)$$

where E_0 and E are the energies of the incident and scattered neutrons, respectively, ν is the cosine of the scattering angle in the laboratory system, $\sigma_b = 81.6 \times 10^{-28} \text{ m}^2$ is the bound effective microscopic cross section of the hydrogen atom nucleus for thermal neutron scattering, $\epsilon = E - E_0$ is the neutron energy exchange, i^* is the complex conjugate value of the imaginary unit, t is the collision time, κ is the neutron wavevector exchange, the symbol $\langle \dots \rangle_{\Omega}$ is the sign for averaging over all

space orientations of the methane molecule and $\chi(\kappa, t)$ is an intermediary scattering function including the influence of the translational, rotational and intravibrational motion of methane molecules on the thermal neutron scattering.

In order to obtain an analytical expression for the microscopic kernel scattering on the hydrogen atom nucleus of the methane molecule based on equation (1), it is necessary to adopt the dynamic model of the methane molecule, in order to perform the orientation averaging and the integration with respect to the collision time.

First, the general dynamic methane molecule model based on the work of Nelkin (1960) and Koppel and Young (1964) is employed. The translation motion is described by the well known free-gas model; the rotational motion is approximated by the isotropic quasi-torsional oscillator. The intravibrations are represented by statistically independent oscillators. In order that this general dynamic model might be applicable, a new microdynamic model for the rotational-intravibrational motion of the methane molecule will be adopted in the following section. This new microdynamic model precisely takes the influence of intravibrations of the methane molecule on the thermal neutron scattering into account. The orientation averaging of the intermediary scattering function $\chi(\kappa, t)$, will be approximately performed by the Krieger-Nelkin (1957) orientation averaging method, which is applicable to the most general case of the anisotropy of intramolecular vibrations according to Marković (1974). As in the work of Marković (1982), the exact collision time is taken for calculation. The integration over the collision time in (1) is performed exactly analytically. Taking the exact collision time into account, a unique expression is obtained in the whole thermal region for the microscopic scattering kernel ($\sigma(E_0 \rightarrow E, \nu)$), which physically describes the thermal neutron scattering on the hydrogen atom nucleus of the methane molecule more realistically. That is the expression obtained for each incident thermal neutron energy enables every single influence of translational, rotational and intravibrational motions of the methane molecule on the thermal neutron scattering to be established. It also involves the effects of quantum-inelastic combined scattering not only at the intravibrational phonons, but also at the rotational-intravibrational phonons of the methane molecule. This last characteristic of the expression obtained for the microscopic scattering kernel has enabled discovery of an effect that a neutron whose energy is not sufficient for creating one intravibrational phonon causes that creation by simultaneous destruction of another intravibrational phonon. The same effect occurs on quantum-inelastic combined thermal neutron scattering at rotational-intravibrational phonons of the methane molecule.

It should be pointed out that the approximate description of the rotational motion of the methane molecule by an isotropic quasi-torsional oscillator can be eliminated on the basis of the studies of Griffing (1961), Sinha and Venkataraman (1966) and Hama and Miyagi (1973). However, an inadequate description of the rotation of the methane molecule in this paper does not influence the effect discovered at all. This effect occurs not only after thermal neutron scattering at a combination of rotational phonons and intravibrational phonons but also as a consequence of the exact description of the collision time for combined thermal neutron scattering at intravibrational phonons. Certainly, for adequate energy values of rotational phonons at low temperatures ($T \geq 0$ K), this model would give qualitatively the same results as those given by Kapulla and Gläser (1972). On the basis of this model it would be interesting to investigate the temperature dependence of this effect.

At the end of this introduction it should be pointed out that further calculations of the total scattering cross sections have recently been performed, but they will not

be represented here. From these calculations it has been established that the total scattering cross sections for small incident neutron energies $E_0 \in [1 \text{ meV}, 10 \text{ meV}]$ are of the correct order of magnitude, while for large incident neutron energies $E_0 = 1 \text{ eV}$ the total scattering cross sections are smaller by a half.

2. Inclusion of the influence of microdynamic methane molecule properties on thermal neutron scattering

The methane molecule consists of one carbon atom and four hydrogen atoms. The hydrogen atom nuclei are so arranged in the equilibrium configuration that they form the vertices of a tetrahedron of equal sides in the centre of which there is a carbon atom nucleus. On the basis of this tetrahedral equilibrium structure it is possible to obtain a valence angle α_0 , i.e. the angle between the valence direction C-H bonds, which is given by $\alpha_0 = 2 \tan^{-1}(\sqrt{2}) = 109^\circ 28'$. Also, the moment of inertia of the methane molecule for this equilibrium configuration is $I_0 = 8md_{CH}^2/3$ in relation to any of its main axes of inertia, where m is the hydrogen atom mass and d_{CH} is the valence length of the C-H bonds. The moment of inertia of the methane molecule $I_0 = 5.33 \times 10^{-47} \text{ kg m}^2$ is obtained by spectroscopic measurement (Childs 1936) of $B_0 = \hbar^2/2I_0 = 0.65 \text{ meV}$, the rotational molecule constant for the equilibrium configuration. This moment of inertia value for the valence length of the C-H bonds in the methane molecule yields $d_{CH} = 109.29 \text{ pm}$. As the methane molecule has five atoms ($N = 5$), then its number of degrees of freedom is 15 ($3N$), three of which are translational, three rotational and nine intravibrational. The number of different frequencies of the normal modes is, as a rule, equal to a number of intravibrational degrees of freedom. However, when intravibration degeneration exists, i.e. when frequencies of some normal modes are the same, then the number of different frequencies of normal modes is smaller than the number of intravibrational degrees of freedom. So the methane molecule has nine intravibrational normal modes, four of which are valence modes and five are deformation modes. The valence modes change the lengths of the C-H bonds, and the deformation modes change the angles between them. However, of these nine intravibrational normal modes the methane molecule has only four different frequencies. One frequency corresponds to the polysymmetrical non-degenerate pure-valence normal mode, and another frequency corresponds to twice the degenerate pure-deformation normal mode. The other two frequencies correspond to threefold-degenerate normal modes, which are hardly said to be pure-valence or pure-deformation modes, because they also change the values of the valence angles and vice versa by changing the valence bond lengths. However, the mutual influence of valence and deformation normal modes is relatively small owing to the high symmetry of the methane molecule; thus one threefold-degenerate normal mode can be approximately taken to be the valence mode and the other the deformation mode. Besides these intravibrational properties of the methane molecule, the rotational motion of the methane molecule will be approximated by an isotropic quasi-torsional oscillator of the same frequency, as stated in section 1. Now the way to include the effect of the previously described microdynamic rotational-intravibrational methane molecule properties in the thermal neutron scattering will be briefly given.

On the basis of the general dynamic model of the methane molecule, described in section 1 of this paper, if we suppose that the translational, rotational and

intravibrational motion modes of the methane molecule are mutually independent, the following expression for the intermediary scattering function is obtained:

$$\chi(\kappa, t) = \exp \left(\frac{\kappa^2}{2M} (i^* t - t^2 E_T) + \frac{1}{2m} \sum_{q=1}^{12} (\kappa \cdot C_q)^2 f_q \right) \quad (2)$$

where $\kappa^2/2 = E_0 + E - 2\nu\sqrt{E_0 E} = \alpha$ is half the square of the neutron wavevector exchange, $M = 16$ is the mass of the methane molecule, $E_T = k_B T = 0.0253$ eV, $m = 1$ is the mass of the hydrogen atom, C_q is the amplitude vector of the q th normal mode and the magnitudes of f_q are of the form

$$f_q = (y_q + y_q^{-1}) / [2\omega_q \sinh(\omega_q/2E_T)] - 1 / [\omega_q \tanh(\omega_q/2E_T)] \quad (3)$$

$(q = 1, 2, \dots, 12)$

where ω_q is the energy of the q th rotational ($q = 1, 2, 3$) and intravibrational ($q = 4, 5, \dots, 12$) phonons, and y_q is given by the following expression:

$$y_q = \exp[-(i^* \omega_q t + \omega_q/2E_T)] \quad (q = 1, 2, \dots, 12). \quad (4)$$

In order that equation (2) is applicable, the amplitude vectors C_q ($q = 1, 2, \dots, 12$) of rotational ($q = 1, 2, 3$) and intravibrational ($q = 4, 5, \dots, 12$) normal modes should be determined. The amplitude vector directions C_q ($q = 1, 2, \dots, 12$) will be determined on the basis of the microdynamical structure of the rotational-intravibrational methane molecule motion previously presented qualitatively.

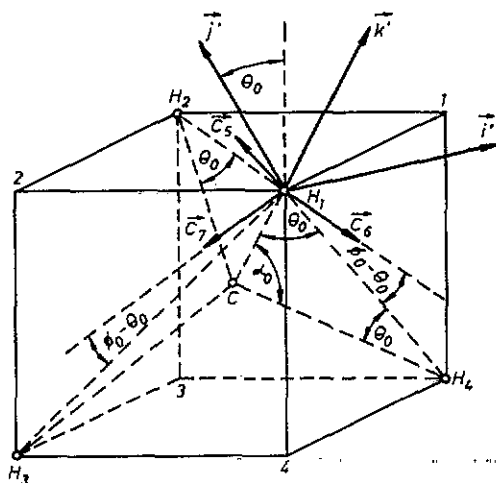


Figure 1. Illustration of the tetrahedral structure of the methane molecule and the way of tying the Cartesian moving coordinate system $O'x'y'z'$ to the H_1 hydrogen atom.

Let the moving Cartesian coordinate system $O'x'y'z'$ be tied to the equilibrium position of any hydrogen atom of the methane molecule, e.g. H_1 (figure 1), whose unit vectors i', j', k' are so oriented that the unit vector k' is in the direction and

orientation of the C-H₁ bond (see figure 1); j' lies in the plane H₁CH₂ and is perpendicular to the C-H₁ bond, and its orientation is as in figure 1.

Therefore, the unit vectors j' and k' lie in the H₁CH₂ plane. As the H₁1H₂ plane is perpendicular to the H₁CH₂ plane, then the unit vector i' must lie in the H₁1H₂ plane. The angle between the direction of the unit vector i' and the direction of H₁1 is $\pi/4$, and the orientation of i' is as in figure 1. When determining all the amplitude vectors of the rotational, i.e. quasi-torsional, oscillations we assume them to be of the same intensities, i.e. $|C_q| = C_r$ ($q = 1, 2, 3$). The same assumption is valid for the amplitude of the intravibrational vectors, i.e. $|C_q| = C_v$ ($q = 4, 5, \dots, 12$). Based on the above conclusions and figure 1, the following microdynamic model of the rotational-intravibrational methane molecule motion is adopted in this paper; this is defined by means of amplitude vectors C_q ($q = 1, 2, \dots, 12$) in the following way:

$$\begin{aligned}
 C_1 &= C_2 = C_3 = (\sqrt{3}/3) C_r (j' \sqrt{2} + k') \\
 C_4 &= C_v k' \\
 C_5 &= C_6 = C_7 = C_v k' \\
 C_8 &= C_v \left[(\sqrt{6}/3) j' - (\sqrt{3}/3) k' \right] \\
 C_9 &= C_v \left[(\sqrt{2}/2) i' - (\sqrt{6}/6) j' - (\sqrt{3}/3) k' \right] \\
 C_{10} &= -C_v \left[(\sqrt{2}/2) i' + (\sqrt{6}/6) j' + (\sqrt{3}/3) k' \right] \\
 C_{11} &= C_v j' \\
 C_{12} &= C_v i'
 \end{aligned} \tag{5}$$

where $C_1 = C_2 = C_3$ are amplitude vectors of the quasi-torsional oscillations, C_4 is the amplitude vector of the polysymmetrical non-degenerate pure-valence intravibrations, $C_5 = C_6 = C_7$ are amplitude vectors of the threefold-degenerate valence intravibrations, C_8, C_9 and C_{10} are amplitude vectors of the threefold-degenerate deformation intravibrations which form the orthogonal trihedron symmetrically placed with respect to the C-H₁ bond, and C_{11} and C_{12} are mutually orthogonal and represent amplitude vectors of the twofold degenerate deformation intravibrations. The first vector bends the C-H₁ bond, and the other turns the H₁1H₂ plane with respect to the H₃3H₄ plane (see figure 1) and the angle between the directions of the amplitude vectors C_8, C_9 and C_{10} and the direction of the C-H₁ bond is $\Phi_0 = \cos^{-1}(\sqrt{3}/3) = \sin^{-1}(\sqrt{6}/3) = 54.73561^\circ$.

Replacing the amplitude vector values C_q ($q = 1, 2, \dots, 12$), which are defined by equation (5), into equation (2), by applying the approximate Krieger-Nelkin (1957) orientation averaging, expanding into phonon series over the Bessel modified functions and performing the exact integration over time in equation (1), the following final expression for the microscopic thermal neutron kernel scattering on the hydrogen atom nucleus of the methane molecule is obtained:

$$\begin{aligned}
 \sigma(E_0 \rightarrow E, \nu) &= \frac{d^2 \sigma}{dE d\nu} = \frac{\sigma_b}{2^{v+2}} \sqrt{\frac{EM}{\pi \alpha E_0 E_T}} \\
 &\times \exp \left\{ - \left[\frac{\alpha}{4ME_T} + \frac{\epsilon}{2E_T} + \sum_{q=1}^4 b_q \cosh \left(\frac{\omega_q}{2E_T} \right) \right] \right\}
 \end{aligned}$$

$$\begin{aligned}
& - \sum_{n=1}^{16} \sum_{i=0}^{+\infty} \sum_{j=0}^{+\infty} \sum_{k=0}^{+\infty} \sum_{l=0}^{+\infty} I_i(b_1) - I_j(b_2) - I_k(b_3) - I_l(b_4) \\
& - \exp \left[- \frac{M}{4\alpha E_T} (\epsilon - B_n)^2 \right] \tag{6}
\end{aligned}$$

where $I_i(b_1)$, $I_j(b_2)$, $I_k(b_3)$ and $I_l(b_4)$ are the modified Bessel functions, while the quantities b_q ($q = 1, 2, 3, 4$) are given by the following relations:

$$\begin{aligned}
b_1 &= \alpha / [m_r \omega_1 \sinh(\omega_1 / 2E_T)] & b_3 &= \alpha / [m_v \omega_3 \sinh(\omega_3 / 2E_T)] \\
b_2 &= \alpha / [0.75 - m_v \omega_2 \sinh(\omega_2 / 2E_T)] & b_4 &= \alpha / [1.50 - m_v \omega_4 \sinh(\omega_4 / 2E_T)] \tag{7}
\end{aligned}$$

where $\omega_1 = 0.011$ eV is the energy of the rotational phonon, $\omega_2 = 0.3684$ eV is the energy of the polysymmetrical non-degenerate pure-valence and threefold-degenerate valence phonons (because of the closeness of the energies of these phonons and the simpler calculation, it is assumed that their energies are the same and equal to the arithmetical mean value of their actual energies), $\omega_3 = 0.1618$ eV is the energy of the threefold-degenerate phonon, $\omega_4 = 0.1896$ eV is the energy of the twofold-degenerate deformation phonon, and $m_r = mC_r^{-2} = 3.40$ and $m_v = mC_v^{-2} = 1.55$ are the effective rotational and vibrational hydrogen atom masses, respectively, linked to the methane molecule.

The energy value of the rotational phonon $\omega_1 = 0.011$ eV is established according to Griffing (1961), Sinha and Venkataraman (1966), Dasannacharya and Venkataraman (1967), Webb (1967) and Sears (1967). On the other hand a quantum mechanical expression for the rotator energy $E_l = l(l+1)B_0$ is well known, where $B_0 = \hbar^2/2I_0$ is the rotational constant of a molecule. According to this expression the value quoted for ω_1 is approximately equal to $E_8 - E_7 = 16B_0 \approx 0.011$ eV, because for the methane molecule $2B_0 = E_1 - E_0 = 1.3$ meV (Childs 1936). Besides this, the calculated effect will be more pronounced if occupation of the level E_7 is sufficiently strong, i.e. when the Gibbs factor is approximately of the order of unity ($\exp(-E_7/kT) \approx 1$). The calculated effect will increase exponentially with increasing temperature owing to the appropriate occupation number $\exp(-E_7/k_B T) / \sum_i \exp(-E_i/k_B T)$.

The quantities B_n ($n = 1, 2, \dots, 16$) in equation (6) are determined by the following expressions:

$$\begin{aligned}
B_1 &= i\omega_1 + j\omega_2 + k\omega_3 + l\omega_4 & B_9 &= -i\omega_1 + j\omega_2 + k\omega_3 + l\omega_4 \\
B_2 &= i\omega_1 + j\omega_2 + k\omega_3 - l\omega_4 & B_{10} &= -i\omega_1 + j\omega_2 + k\omega_3 - l\omega_4 \\
B_3 &= i\omega_1 + j\omega_2 - k\omega_3 + l\omega_4 & B_{11} &= -i\omega_1 + j\omega_2 - k\omega_3 + l\omega_4 \\
B_4 &= i\omega_1 + j\omega_2 - k\omega_3 - l\omega_4 & B_{12} &= -i\omega_1 + j\omega_2 - k\omega_3 - l\omega_4 \\
B_5 &= i\omega_1 - j\omega_2 + k\omega_3 + l\omega_4 & B_{13} &= -i\omega_1 - j\omega_2 + k\omega_3 + l\omega_4 \\
B_6 &= i\omega_1 - j\omega_2 + k\omega_3 - l\omega_4 & B_{14} &= -i\omega_1 - j\omega_2 + k\omega_3 - l\omega_4 \\
B_7 &= i\omega_1 - j\omega_2 - k\omega_3 + l\omega_4 & B_{15} &= -i\omega_1 - j\omega_2 - k\omega_3 + l\omega_4 \\
B_8 &= i\omega_1 - j\omega_2 - k\omega_3 - l\omega_4 & B_{16} &= -i\omega_1 - j\omega_2 - k\omega_3 - l\omega_4. \tag{8}
\end{aligned}$$

It should be pointed out that in equation (6) the primes in the summations over the indices i, j, k and l denote specific i.e. incomplete summations which take into account the fact that $\sigma(E_0 \rightarrow E, \nu)$ essentially consists of the sum of 16 partial microscopic scattering kernels. Therefore in equation (6) a specific unique way of summation over indices i, j, k and l exists. These unique summations as well as the values of the parameters ν in equation (6) are given in table 1.

Table 1. Summations over i, j, k, l and p , together with values of ν .

| ν | Σ_i | Σ_j | Σ_k | Σ_l | Σ_p |
|-------|------------|------------|------------|------------|------------|
| 4 | 0 | 0 | 0 | 0 | 1 |
| 3 | 1, ..., +∞ | 0 | 0 | 0 | 2 |
| 3 | 0 | 1, ..., +∞ | 0 | 0 | 3 |
| 3 | 0 | 0 | 1, ..., +∞ | 0 | 4 |
| 3 | 0 | 0 | 0 | 1, ..., +∞ | 5 |
| 2 | 1, ..., +∞ | 1, ..., +∞ | 0 | 0 | 6 |
| 2 | 1, ..., +∞ | 0 | 1, ..., +∞ | 0 | 7 |
| 2 | 1, ..., +∞ | 0 | 0 | 1, ..., +∞ | 8 |
| 2 | 0 | 1, ..., +∞ | 1, ..., +∞ | 0 | 9 |
| 2 | 0 | 1, ..., +∞ | 0 | 1, ..., +∞ | 10 |
| 2 | 0 | 0 | 1, ..., +∞ | 1, ..., +∞ | 11 |
| 1 | 1, ..., +∞ | 1, ..., +∞ | 1, ..., +∞ | 0 | 12 |
| 1 | 1, ..., +∞ | 1, ..., +∞ | 0 | 1, ..., +∞ | 13 |
| 1 | 1, ..., +∞ | 0 | 1, ..., +∞ | 1, ..., +∞ | 14 |
| 1 | 0 | 1, ..., +∞ | 1, ..., +∞ | 1, ..., +∞ | 15 |
| 0 | 1, ..., +∞ | 1, ..., +∞ | 1, ..., +∞ | 1, ..., +∞ | 16 |

In table 1 the index p determines 16 partial microscopic scattering kernels, i.e.

$$\sigma(E_0 \rightarrow E, \nu) = \sum_{p=1}^{16} \sigma_p(E_0 \rightarrow E, \nu) \quad (9)$$

where every partial microscopic scattering kernel $\sigma_p(E_0 \rightarrow E, \nu)$ ($p = 1, 2, \dots, 16$) is also defined by both equation (6) and table 1. The partial microscopic scattering kernels determine the influence of single modes of the methane molecule motions on the thermal neutron scattering. Thus, $\sigma_1(E_0 \rightarrow E, \nu)$ determines the influence of translations and elastic vibration transfers of the methane molecule on the thermal neutron scattering, i.e. it defines the quasi-elastic scattering. The other 15 partial microscopic kernels $\sigma_p(E_0 \rightarrow E, \nu)$ ($p = 2, 3, \dots, 16$) determine the effects of the inelastic thermal neutron scattering on the rotational and single intravibrational phonons ($p = 2, 3, 4, 5$), on the combination of the rotational phonons with single intravibrational phonons ($p = 6, 7, 8$), on the combination of single intravibrational phonons ($p = 9, 10, 11$), on the combination of rotational phonons with two intravibrational phonons ($p = 12, 13, 14$), on the combination of all three intravibrational phonons ($p = 15$) and on the combination of the rotational phonons with all three intravibrational phonons ($p = 16$).

3. The effect of creation and simultaneous destruction of intravibrational and rotational phonons

On the basis of equation (6) derived for the microscopic scattering kernel, the

complete analysis of the thermal neutron scattering on a hydrogen atom nucleus of the methane molecule will not be given, but only the effect showing that a neutron with an insufficient energy for the creation of one intravibrational phonon can cause this creation by the simultaneous destruction of another intravibrational phonon. The same effect occurs on combined inelastic thermal neutron scattering at rotational-intravibrational phonons of the methane molecule. The microscopic differential cross sections are calculated, for three incident thermal neutron energies ($E_0 = 1, 20$ and 100 meV), by integrating the microscopic scattering kernels (6) over all the possible values of the cosine angle of the scattered thermal neutrons which are defined by the following relation:

$$\sigma(E_0 \rightarrow E) = \int_{-1}^{+1} \sigma(E_0 \rightarrow E, \nu) d\nu. \quad (10)$$

The partial microscopic differential cross sections $\sigma_{11}(E_0 \rightarrow E)$ and $\sigma_{14}(E_0 \rightarrow E)$ for the incident neutron energy $E_0 = 1$ meV are presented in figure 2. The partial microscopic differential cross section $\sigma_{11}(E_0 \rightarrow E)$ defines the thermal neutron scattering on the combination of the twofold-degenerate ($\omega_4 = 0.1896$ eV) and threefold-degenerate ($\omega_3 = 0.1618$ eV) deformation phonons, and $\sigma_{14}(E_0 \rightarrow E)$ defines the thermal neutron scattering on the combination of the rotational ($\omega_1 = 0.011$ eV), twofold-degenerate ($\omega_4 = 0.1896$ eV) and threefold-degenerate ($\omega_3 = 0.1618$ eV) deformation phonons. When the scattering neutron energy is $E \simeq E_0 + (\omega_4 - \omega_3) = 0.0288$ eV, then $\sigma_{11}(E_0 \rightarrow E)$ has the maximum value (see figure 2). This property shows that the neutron, although having a small incident energy ($E_0 = 1$ meV), is able to cause the creation of an intravibrational threefold-degenerate deformation phonon ($\omega_3 = 0.1618$ eV), enabling the creation of that phonon to take place by the simultaneous destruction of one intravibrational twofold-degenerate deformation phonon ($\omega_4 = 0.1896$ eV). The neutron obtains the energy difference ($\omega_4 - \omega_3 = 0.0278$ eV) and is scattered with energy $E \simeq E_0 + (\omega_4 - \omega_3) = 0.0288$ eV which is greater than its incident energy E_0 . The partial microscopic differential cross section $\sigma_{14}(E_0 \rightarrow E)$ shows a similar property in figure 2, because it has the maximal value when the scattered neutron energy is $E \simeq E_0 + (\omega_4 - \omega_3 + 2\omega_1) = 0.0498$ eV. This maximum shows the effect of neutron influence on the creation of one threefold-degenerate deformation intravibrational phonon ($\omega_3 = 0.1618$ eV) and a simultaneous destruction of one twofold-degenerate deformation phonon ($\omega_4 = 0.1896$ eV) and two rotational ($\omega_1 = 0.011$ eV) phonons.

In the same way as for $E_0 = 1$ meV the partial microscopic differential cross section $\sigma_{11}(E_0 \rightarrow E)$ for $E_0 = 20$ meV has a maximum when $E \simeq E_0 + (\omega_4 - \omega_3) = 0.0478$ eV (figure 3(a)) showing the same effect of neutron influence on the creation of one threefold-degenerate deformation intravibrational phonon (ω_3) and a simultaneous destruction of one twofold-degenerate deformation intravibrational phonon (ω_4). The partial microscopic differential cross section $\sigma_{14}(E_0 \rightarrow E)$ is shown in figure 3(b), because it also has a maximum for $E \simeq E_0 + (\omega_4 - \omega_3 - \omega_1) = 0.0368$ eV, indicating the existence of the effects of the neutron influence on the simultaneous creation of one threefold-degenerate deformation intravibrational phonon (ω_3) and one rotational phonon (ω_1) and the simultaneous destruction of one twofold-degenerate deformation intravibrational phonon (ω_4).

However, when the incident neutron energy is $E_0 = 100$ meV, then the curve $\sigma_{11}(E_0 \rightarrow E)$ has a maximum for $E \simeq E_0 - (\omega_4 - \omega_3) = 0.0722$ eV, showing a

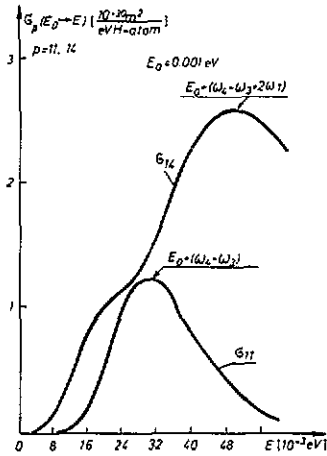


Figure 2. The curves of the partial differential cross section $\sigma_p(E_0 \rightarrow E)$ ($p = 11, 14$) for the incident neutron energy $E_0 = 0.001$ eV.

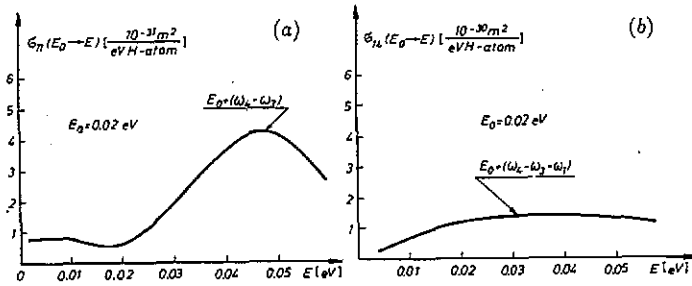


Figure 3. The curves of the partial differential cross sections (a) $\sigma_{11}(E_0 \rightarrow E)$ and (b) $\sigma_{14}(E_0 \rightarrow E)$ for the incident neutron energy $E_0 = 0.02$ eV.

completely inverse effect compared with the effects described for $E_0 = 1$ meV and $E_0 = 20$ meV. That is, this maximum (figure 4(a)) on the curve $\sigma_{11}(E_0 \rightarrow E)$, shows the effect of the neutron influence on the creation of one twofold-degenerate deformation intravibrational phonon (ω_4) and the simultaneous destruction of one threefold-degenerate deformation intravibrational phonon (ω_3). The curve $\sigma_{14}(E_0 \rightarrow E)$ is presented in figure 4(b) for the same incident neutron energy $E_0 = 100$ meV. It has a maximum for $E \simeq E_0 - (\omega_4 - \omega_3 + 3\omega_1) = 0.0392$ eV, showing a partially inverse effect, obtained for $E_0 = 1$ meV and $E_0 = 20$ meV. This maximum shows the existence of the effect of neutron influence on the simultaneous creation of one twofold-degenerate deformation intravibrational phonon (ω_4) and on the creation of three rotational phonons (ω_1) and on the simultaneous destruction of one threefold-degenerate deformation intravibrational phonon (ω_3).

4. Conclusion

In this paper, a new effect of the methane molecule is discovered, namely that a neutron with insufficient energy is able to cause the creation of an intravibrational phonon by the simultaneous destruction of another intravibrational phonon. The

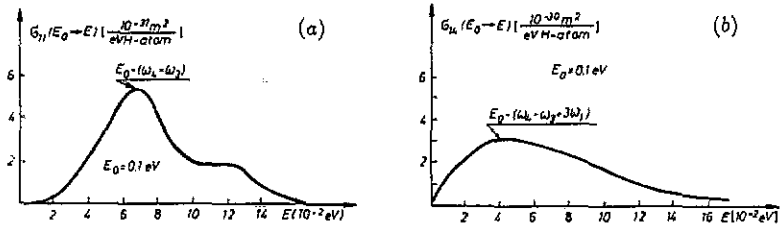


Figure 4. The curves of the partial differential cross sections (a) $\sigma_{11}(E_0 \rightarrow E)$ and (b) $\sigma_{14}(E_0 \rightarrow E)$ for the incident neutron energy $E_0 = 0.1$ eV.

same effect exists not only with intravibrational phonons but also with combinations of rotational and intravibrational phonons. Evidently, this effect exists theoretically for the thermal neutron energies $E_0 \in [1 \text{ meV}, 100 \text{ meV}]$.

References

- Childs W H J 1936 *Proc. R. Soc.* **153** 555
- Dasannacharya B A and Venkataraman G 1967 *Phys. Rev.* **156** 196–203
- Fermi E 1936 *Ricerca Sci.* **7** 2–13
- Griffing G W 1961 *Phys. Rev.* **124** 1489–93
- Hama J and Miyagi H 1973 *Prog. Theor. Phys.* **50** 1142–55
- Herzberg G 1966 *Molecular Spectra and Molecular Structure* vol III *Electronic Spectra and Electronic Structure of Polyatomic Molecules* (New York: Van Nostrand) p 527
- Kapulla H and Gläser W 1972 *Neutron Inelastic Scattering* (Vienna: IAEA) pp 841–9
- Koppel J U and Young J A 1964 *Nucl. Sci. Eng.* **19** 412–17
- Krieger T J and Nelkin M S 1957 *Phys. Rev.* **106** 290–5
- Marković M I 1974 *J. Phys. B: At. Mol. Phys.* **7** L305–9
- 1982 *Nucl. Instrum. Methods* **195** 597–606
- Nelkin M S 1960 *Phys. Rev.* **119** 741–6
- Sears V F 1967 *Can. J. Phys.* **45** 237–54
- Sinha S K and Venkataraman G 1966 *Phys. Rev.* **149** 1–13
- Webb F J 1967 *Proc. Phys. Soc.* **92** 912–20
- Zemach A C and Glauber R J 1956 *Phys. Rev.* **101** 118–29, 129–36