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Spectroscopy of cooperative laser electron- γ -nuclear processes in polyatomic molecules

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Abstract. The consistent quantum approach to calculating the electron-nuclear γ transition spectra (a set of the vibration-rotational satellites in a molecule) of a nucleus in polyatomic molecules is used to determine accurate data of the vibration-nuclear transition probabilities. We present results for emission and absorption γ -spectra of nucleus ^{191}Ir ($E_{\gamma}^{(0)} = 82$ keV) in the molecule of IrO_4 .

1. Introduction

Any alteration of the molecular state must be manifested in the quantum transitions, for example, in a spectrum of the γ -radiation of a nucleus. It is well known that it is possible to transfer part of nuclear energy to atoms or molecules by emitting or absorbing γ quanta (c.f.[1-12]). A spectrum contains a set of the electron-vibration-rotation satellites, which are due to an alteration of the state of system interacting with photons. A mechanism of forming satellites in the molecule is connected with a shaking of the electron shell resulting from the interaction between a nucleus and γ quantum. [1, 3, 13-20]. Appearance of the molecular nuclear lines is of great interest as one can possibly modify the γ -radiation spectrum by means of changing the vibration-rotational state of a molecule by a laser light [1,12,16]. The well known example is the Szilard-Chalmers effect (molecular dissociation because of the recoil during radiating gamma quantum with large energy; c.f. [1, 2]).

This work addresses our study of the co-operative dynamical phenomena (c.f.[14-19]) due the interaction between atoms, ions, molecule electron shells and nuclei nucleons. We present an advanced approach to description of a new class of dynamical laser-electron-nuclear effects in molecular spectroscopy, in particular, we study the nuclear gamma-emission or absorption spectrum of a molecule. A consistent quantum-mechanical approach to calculation of the electron-nuclear γ transition spectra (set of vibration-rotational satellites in molecule) of a nucleus in the polyatomic molecules has been proposed earlier [14-16]. It generalizes the well known Letokhov-Minogin model [3]. Estimates of the vibration-nuclear transition probabilities in a case of the emission and absorption spectrum of nucleus ^{188}Os ($E_{\gamma}^{(0)} = 155$ keV) in the molecule of OsO_4 and other molecules were listed. Here we report accurate data on the vibration-nuclear transition probabilities in a case of the emission and absorption spectrum of nucleus ^{191}Ir ($E_{\gamma}^{(0)} = 82$ keV) in the molecule of IrO_4 .



2. The electron-nuclear γ transition spectra of nucleus in multi-atomic molecules

As the method of computing is presented in detail earlier [2,3,14-16], here we consider only the key aspects. The main purpose is calculating a structure of the gamma transitions (a probability of transition) or spectrum of the gamma satellites because of changes to the electron-vibration-rotational states of the polyatomic molecules under gamma quantum radiation (absorption). Let us note that obviously the γ -spectrum component intensities because of the electron transitions are very small (the smallness parameter m_e/M , where m_e is an electron mass, M is mass of a molecule). So, it is of an especial interest studying nuclear γ -spectra in molecules, because of the vibration-rotational transitions. Comparing the averaged energies for excitation of the molecule vibration and rotation, one could remind that for polyatomic molecules it is typical $B/\hbar\omega \sim 10^{-4}-10^{-2}$. If the γ -active nucleus is located in the centre of mass of the molecule, then it is possible to consider only the vibration transitions of the molecule and neglect of the rotational transitions [2,5]. This approximation is particularly important for those molecules in which the position of the γ -active nucleus in the centre of mass is due to the symmetry of the molecule and is not random. If the γ -active nucleus in a molecule is out of the centre of mass, then the situation becomes much more complicated. Formal foundations of the general theory of the γ -emission and absorption in the nuclei of symmetric polyatomic molecules have been presented in [2,3], where the authors considered the case of molecules of the XY_2 type.

Here we limit our study to five-atomic molecules (of XY_4 type; T_d). The interaction Hamiltonian of the γ -radiation with a system of nucleons for the first nucleus can be expressed through the co-ordinates of nucleons r_n in a system of the mass centre of the one nucleus [3,15]:

$$H(r_n) = H(r_n') \exp(-ik_\gamma u) \quad (1)$$

where k_γ is a wave vector of the γ -quantum; u is the shift vector from equality state (coinciding with molecule mass centre) in system of co-ordinates in the space. The matrix element for transition from the initial state "a" to the final state "b" is presented as usual:

$$\langle \Psi_b^* | H | \Psi_a \rangle \bullet \langle \Psi_b^* | e^{-ik_\gamma u} | \Psi_a \rangle \quad (2)$$

where a and b is a set of quantum numbers, which define the vibrational and rotational states before and after interaction (with gamma- quantum). The first multiplier in Eq. (2) is defined by the gamma transition of nucleus and is not dependent upon the internal structure of molecule in good approximation. The second multiplier is the matrix element of transition of the molecule from the initial state "a" to the final state "b":

$$M_{ba} = \langle \Psi_b^*(r_e) | \Psi_a(r_e) \rangle \bullet \langle \Psi_b^*(R_1, R_2) | e^{-ik_\gamma R_1} | \Psi_a(R_1, R_2) \rangle \quad (3)$$

The expression in Equation (3) gives a general formula for calculating the probability of changing the internal state of molecule during absorption or emission of a γ -quantum by a nucleus. It determines an intensity of the corresponding γ -satellites. Their positions are fully determined as follows:

$$E_\gamma = E_\gamma^0 \pm R + \hbar k_\gamma v \pm (E_b - E_a) \quad (4)$$

Here, M is the molecule mass, v is a velocity of molecule before interaction of nucleus with γ quantum; E_a and E_b are the energies of the molecule before and after interaction; E_γ is an energy of nuclear transition; R_{om} is an energy of recoil: $R_{om} = [(E_\gamma^{(0)})^2 / 2Mc^2]$.

Further we miss the molecule rotations and consider γ -spectrum of a nucleus in the molecule mass centre as a spectrum of the vibration-nuclear transitions (see details in Refs. [3,15]). Further one can suppose that only single non-generated normal vibration (vibration quantum $\hbar\omega$) is excited and initially a molecule is on the vibrational level $v_a = 0$. If we denote a probability of the corresponding excitation as $P(v_b, v_a)$ and use expression for shift u of the γ -active nucleus through the normal co-ordinates, then an averaged energy for excitation of the single normal vibration is as follows [3]:

$$\begin{aligned} \bar{E}_{\text{vib}} &= \sum_{v=0}^{\infty} \hbar\omega(v + 1/2) \bar{P}(v,0) - \hbar\omega / 2 = \sum_{v=0}^{\infty} \hbar\omega(v + 1/2) P(v,0) - \hbar\omega / 2 = \\ &= \sum_{v=0}^{\infty} \hbar\omega(v + 1/2) \frac{z^v}{v!} e^{-z} - \frac{\hbar\omega}{2} = \frac{1}{2} R \left(\frac{M - m}{m} \right), \end{aligned} \quad (5)$$

where

$$z = (R / \hbar\omega) [M - m / m] \cos^2 \vartheta,$$

and m is the mass of γ -active nucleus, ϑ is an angle between nucleus shift vector and wave vector of γ -quantum and line in \bar{E}_{vib} means averaging on orientations of molecule (or on angles ϑ).

A shift u of the γ -active nucleus can be expressed through the normal co-ordinates $Q_{s\sigma}$ of a molecule:

$$u = \frac{1}{\sqrt{m}} \sum_{s\sigma} b_{s\sigma} Q_{s\sigma} \quad (6)$$

where m is a mass of the γ -active nucleus; components of the vector $b_{s\sigma}$ of nucleus shift due to the Φ -component of “ s ” normal vibration of a molecule are the elements of matrix b [2]. It realizes the orthogonal transformation of the normal co-ordinates matrix Q to matrix of masses of the weighted Cartesian components of the molecule nuclei shifts q . According to Equation (1), the matrix element can be written as multiplying the matrix elements on molecule normal vibration, which takes contribution to a shift of the γ -active nucleus:

$$M(b, a) = \prod_s \left\langle v_s^b \mid \prod_{\sigma} \exp(-ik_{\gamma} b_{s\sigma} Q_{s\sigma} / \sqrt{m}) v_s^a \right\rangle. \quad (7)$$

It is obvious that missing molecular rotations means missing the rotations which are connected with the degenerate vibrations. The wave functions of a molecule should be written for non-degenerate vibration as: $|v_s\rangle = \Phi_{v_s}(Q_s)$, for double degenerate vibration in the form:

$$|v_s\rangle = (v_s + 1)^{-1/2} \sum_{v_{s\sigma_1}, v_{s\sigma_2}, v_{s\sigma_3}} \Phi_{v_{s\sigma_1}}(Q_{s\sigma_1}) \Phi_{v_{s\sigma_2}}(Q_{s\sigma_2}) \quad (8)$$

(where $v_{s\sigma_1} + v_{s\sigma_2} = v_s$) and for triple degenerate vibration as follows:

$$|v_s\rangle = \left(\frac{2}{(v_s + 1)(v_s + 2)} \right)^{1/2} \sum_{v_{s\sigma_1}, v_{s\sigma_2}, v_{s\sigma_3}} \Phi_{v_{s\sigma_1}}(Q_{s\sigma_1}) \Phi_{v_{s\sigma_2}}(Q_{s\sigma_2}) \Phi_{v_{s\sigma_3}}(Q_{s\sigma_3}) \quad (9)$$

where

$$v_{s\sigma_1} + v_{s\sigma_2} + v_{s\sigma_3} = v_s.$$

In the simple approximation function $\Phi_{v_{s\sigma}}(Q_{s\sigma})$ can be chosen in a form of the linear harmonic oscillator one. Taking into account the explicit expressions for the wave functions, the calculation of the matrix element (7) is reduced to the computation of the matrix elements for each component of the normal vibration.

3. Results and conclusions

Below we present the advanced data on the vibration-nuclear transition probabilities in a case of the emission and absorption γ -spectrum of nucleus ^{191}Ir ($E_{\gamma}^{(0)} = 82$ keV) in the molecule IrO_4 . The main difficulty during calculating (7) is connected with definition of the values $b_{s\sigma}$ of the normalized shifts of γ -active decay. It is known that if a molecule has the only normal vibration of the given symmetry

type, then the corresponding values of $b_{s\sigma}$ can be found from the well-known Eckart conditions. For several normal vibrations of one symmetry type, a definition of $b_{s\sigma}$ requires solving the secular equation for molecule $|GF-\lambda E|=0$ [21-24]. We have used the results of advanced theoretical calculation of electron structure of the studied system within an advanced scheme of the X_v -scattered waves method [22,24]. Other details can be found in Refs. [3,15]. In Table 1 we list the results of calculating probabilities of the first several vibration-nuclear transitions in a case of the emission and absorption γ -spectrum of nucleus ^{191}Ir ($E_{\gamma}^{(0)}=82$ keV) in IrO_4 and compare with available data in Ref.[2,15], where more simple approximation has been used. Analysis shows that more sophisticated calculation gives higher values for probabilities.

Table 1. Probabilities of the vibrational-nuclear transitions in spectrum of IrO_4 .

Vibration transition $v_3^a, v_4^a - v_3^b, v_4^b$	$\overline{P}(v_3^a, v_4^a - v_3^b, v_4^b)$ [15]	$\overline{P}(v_3^a, v_4^a - v_3^b, v_4^b)$ This work
0,0 – 0,0	0.863	0.953
1,0 – 0,0	0.025	0.032
0,1 – 0,0	0.097	0.104
1,0 – 1,0	0.812	0.901
0,1 – 0,1	0.731	0.813

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