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CONSISTENT QUANTUM APPROACH TO NEW LASER-ELECTRON-NUCLEAR PHOTO PHENOMENA IN MOLECULES: XY_4

Consistent, quantum approach to calculation of the electron-nuclear γ transition spectra (set of vibration-rotational satellites in molecule) of nucleus in multiatomic molecules is used for estimates of the vibration-rotation-nuclear transition probabilities in a case of the emission and absorption spectrum of nucleus ^{191}Ir($E^{(0)}_\gamma = 82$ keV) linked with molecular system IrO_4.

Our paper is devoted to studying co-operative dynamical phenomena due to interaction between photons, nuclei and electron shells in the atomic and molecular systems (c. f. [1—15]). In this direction the following problems would be considered [1, 2, 8, 9]: i). Studying the mixed optical quantum photo transitions and laser-electron-nuclear photo phenomena in molecular systems; ii). A spectroscopy of resonance's and creation of additional satellites and narrow resonance's inside the Doppler contour of radiation line; iii). a governing by the intensity of the complicated transitions due to change of the atomic and molecular excited states population in a laser field. We will consider a consistent approach to description of a new class dynamical laser-electron-nuclear effects in molecular systems, in particular, the nuclear emission or absorption spectrum of the diatomic molecule. This spectrum contains a set of electron-vibration-rotation satellites, which are due to an alteration of the state of system under interaction with photon. [2, 12]. A mechanism of forming satellites in the molecule is connected with a shaking of the electron shell resulting from the interaction between the nucleus and γ quantum. The well known example is the Szilard-Chalmers effect (molecular dissociation because of the recoil during radiating gamma quantum with large energy) [1]. It is well known that in a case of an atom the corresponding satellite spectrum is much enriched and transitions between the fine structure components, 0-0 transitions and transitions, which do not involve a change in the electron configuration, can be considered. In the molecules, especially multi-atomic ones, a spectrum is naturally more complicated in comparison with an atom. Under nuclear γ-quantum emission or absorption there is a change of the electron (vibration-rotation) states. In ref. [8—16] we have developed new consistent, quantum-mechanical and quantum-electrodynamical approaches to calculation of the electron-nuclear γ transition spectra (set purely electron satellites in atoms, ions and of vibration-rotational-electron satellites in diatomic molecules) of nucleus in atoms, ions and diatomics. In ref. [8, 16] we developed a consistent, quantum approach to

calculation of the electron-nuclear γ transition spectra (set of vibration-rotational satellites in molecule) of nucleus in multiatomic molecules. This approach generelizes the well known Letokhov-Minogin model [2]. In ref. [16] the estimates were made for vibration-rotation-nuclear transition probabilities in a case of the emission and absorption spectrum of nucleus ^{188}Os ($E^{(0)}_\gamma = 155$ keV) linked with molecule OsO_4. Here we consider vibration-rotation-nuclear transition probabilities in a case of the emission and absorption spectrum of nucleus ^{191}Ir ($E^{(0)}_\gamma = 82$ keV) linked with molecule IrO_4.

Let us present the key moments of theory [8, 16]. The main purpose is calculation of a structure of the gamma transitions (probability of transition) or spectrum of the gamma satellites because of the changing the electron-vibration-rotational states of multiatomic molecules under the gamma quantum radiation (absorption). Further we are limited by a case of the five-atomic molecules (of the XY_4 type; T_d). Hamiltonian of interaction of the gamma radiation with system of nucleons for the first nucleus can be expressed through the co-ordinates of nucleons rn' in a system of the mass centre of the one nucleus [1, 5, 6]:

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

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

$$< \Psi_b^* \mid H \mid \Psi_a > \cdot < \Psi_b^* \mid e^{-ik_\gamma u} \mid \Psi_a >, \qquad (2)$$

where a and b is a set of quantum numbers, defining vibrational and rotational states before and after interaction (with gamma- quantum). The first multiplier in (1) is defined by the gamma transition of nucleus and is not dependent upon an internal structure of molecule in a good approximation. The second multiplier is a matrix element of transition of the molecule from initial state "a" to final state "b":

$$M_{ba} = < \Psi_b^*(r_e) \mid \Psi_a(r_e) > \cdot$$

$$\cdot < \Psi_b^*(R_1, R_2) \,|\, e^{-ik_\gamma R_1} \,|\, \Psi_a(R_1, R_2) > . \qquad (3)$$

The expression (3) gives a general formula for calculation of the probability of changing internal state of molecule under absorption or emitting gamma quantum by nucleus of the molecule and defines an amplitude of the corresponding gamma satellites. Their positions are fully determined by the energy and pulse conserving laws as follows [2]:

$$E_\gamma = E_\gamma^0 \pm R + \hbar k_\gamma \upsilon \pm (E_b - E_a). \qquad (4)$$

Here R_{om} is an energy of recoil: $R_{om} = [(E_\gamma^{(o)})^2/2Mc^2$, 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 molecule before and after interaction, E_γ is an energy of nuclear transition. An averaged energies for excitations of roations and vibrations under absorption or emitting gamma quantum by nucleus of the molecule can be easily evaluated as follows [2]. One can suppose that only single non-generated normal vabration (vibration quantum $\hbar\omega$) is excited and initially a moleculae 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 single normal vibration is as follows:

$$\bar{E}_{vib} = \sum_{v=0}^{\infty} \hbar\omega\left(v + \tfrac{1}{2}\right)\overline{P}(v,0) - \hbar\infty/2 =$$

$$= \sum_{v=0}^{\infty} \hbar\omega\left(v + \tfrac{1}{2}\right)P(v,0) - \hbar\omega/2 =$$

$$= \sum_{v=0} \hbar\omega\left(v + \tfrac{1}{2}\right)\frac{z^v}{v!}e^{-z} - \frac{\hbar\omega}{2} = \frac{1}{2}R\left(\frac{M-m}{m}\right) \qquad (5)$$

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

To estimate an averaged energy for excitation of the molecule rotation one must not miss the molecule vibrations as they provide non-zeroth transfered to molecule momentum of γ-quantum $L = k_v u\sin\vartheta$. In supposition that a nucleus takes participance only in single non-generated normal vibration and vibrational state of molecule is not changed $v_a = v_b = 0$, one could evaluate an averaged energy for excitation of the molecule rotation as [2]:

$$\bar{E}_{rot} = \left\langle \overline{BL^2} \right\rangle = Bk_\gamma^2 \left\langle u^2 \right\rangle \overline{\sin^2\vartheta} =$$

$$= \tfrac{1}{2}R(B/\hbar\omega)[(M-m)/m]. \qquad (6)$$

So, it is ture a relation $\bar{E}_{rot}/\bar{E}_{vib} \sim B/\hbar\omega$, i. e. under absorption or emitting gamma quantum by nucleus of the molecule a relationship between averaged energies for excitation of the molecule rotations and vibrations coincides on order of value with relationship between energies of rotational and vibrational quantums. As for multiatomic molecules it is typical $B/\hbar\omega$ $\sim10^{-4}-10^{-2}$, so one could miss the molecule rotations and consider γ-spectrum of nucleus in molecule nass centre as spectrum of vibration-nuclear transitions.

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

$$u = \frac{1}{\sqrt{m}}\sum_{s\sigma} b_{s\sigma}Q_{s\sigma}, \qquad (7)$$

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

$$M(b,a) = \prod_s \left\langle v_s^b \,\bigg|\, \prod_\sigma \exp(-ik_\gamma b_{s\sigma}Q_{s\sigma}/\sqrt{m})\, v_s^a \right\rangle. \qquad (8)$$

It is obvious that missing molecular rotations means missing rotations, connected with degenerated vibrations. Usually wave functions of molecule can be written for non-degenerated vibration as:

$$|v_s\rangle = \Phi_{vs}(Q_s), \qquad (9)$$

for double degenerated vibration in the form:

$$|v_s\rangle = (v_s + 1)^{-\frac{1}{2}} \sum_{vs\sigma_1, vs\sigma_2, vs\sigma_3} \Phi_{v_{s\sigma_1}}(Q_{s\sigma_1})\Phi_{v_{s\sigma_2}}(Q_{s\sigma_2}), \qquad (10)$$

(where $v_{s\sigma_1} + v_{s\sigma_2} = v_s$) and for triple degenerated one as:

$$|v_s\rangle = \left(\frac{2}{(v_s+1)(v_s+2)}\right)^{\frac{1}{2}} \times$$

$$\times \sum_{vs\sigma_1, vs\sigma_2, vs\sigma_3} \Phi_{v_{s\sigma_1}}(Q_{s\sigma_1})\Phi_{v_{s\sigma_2}}(Q_{s\sigma_2})\Phi_{v_{s\sigma_3}}(Q_{s\sigma_3}), \qquad (11)$$

(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 choosen in a form of linear harmonic oscillator one. More exact calculating requires numerical determintion of these functions. Giving directly wave

functions $\left|v_s^a\right\rangle$ and $\left|v_s^b\right\rangle$, calculating of matrix element (8) is reduced to definition of the matrix elements on each component Φ of normal vibration.

Below we present the results of calculation for the vibration-nuclear transition probabilities in a case of the emission and absorption spectrum of nucleus ^{191}Ir $(E^{(0)}_\gamma = 82$ keV) linked with molecular system IrO_4. Note that the main difficulty under calculating (8) is connected with definition of values $b_{s\sigma-}$ of the normalized shifts of the γ-active decay. It is known B 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 Eccart conditions, normalization one and data about molecule symmetry. For several normal vibrations of the one symmetry type, a definition of $b_{s\sigma}$ requires solution of the secular equation for molecule $|GF - \lambda E| = 0$ [2]. We have used the results of theoretical calculating electron structure of studied system within relativistic scheme of the $X_\forall$- scattered waves method (see details in ref.[17]). In table 1 we present the results of calculating probabilitites of the first several vibrational-nuclear transitions for molecule IrO_4. Obviously a direct experimental observation of the considered gamma-electron-nuclear phenomena ts is of a great interest.

Table 1

Probabilitites of vibrational-nuclear transitions for molecule IrO_4

Vibration transition $v_3^a, v_4^a - v_3^b, v_4^b$	$\overline{P}$ $(v_3^a, v_4^a - v_3^b, v_4^b)$ present paper
0,0 — 0,0	0.863
1,0 — 0,0	0.025
0,1 — 0,0	0.097
1,0 — 1,0	0.812
0,1 — 0,1	0.731

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ПОСЛЕДОВАТЕЛЬНЫЙ КВАНТОВЫЙ ПОДХОД К ОПИСАНИЮ НОВЫХ ЛАЗЕРНЫХ ЭЛЕКТРОН-ЯДЕРНЫХ ФОТОЯВЛЕНИЙ В МОЛЕКУЛАХ: XY$_4$

Последовательный квантовый подход к описанию спектра лазерных электрон-ядерных γ переходов (система колебательно-вращательно-ядерных сателлитов) в много-атомных молекулах использован в расчете вероятностей колебательно-ядерных переходов в спектре излучения и поглощения ядра ^{191}Ir ($E^{(0)}_\gamma = 82$ keV) в системе IrO_4.

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ПОСЛІДОВНИЙ КВАНТОВИЙ ПІДХІД ДО ОПИСУ НОВИХ ЛАЗЕРНИХ ЕЛЕКТРОН-ЯДЕРНИХ ФОТОЯВИЩ У МОЛЕКУЛАХ: XY$_4$

Послідовний квантовий підхід до опису спектру лазерних електрон-ядерних γ переходів (система коливно-обертально-ядерних сателітів) у багатоатомних молекулах використано у розрахунку імовірностей коливно-ядерних переходів у спектрі випромінювання та поглинення ядра ^{191}Ir ($E^{(0)}_\gamma = 82$ keV) у системі IrO_4.