

Consistent quantum approach to new laser-electron-nuclear effects in diatomic molecules

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Abstract. We present a consistent, quantum approach to the calculation of electron-nuclear γ spectra (set of vibrational and rotational satellites) for nuclei in diatomic molecules. The approach generalizes the well known Letokhov-Minogin model and is based on the Dunham model potential approximation for potential curves of diatomic molecules. The method is applied to the calculation of probabilities of the vibration-rotation-nuclear transitions in a case of emission and absorption spectrum for the nucleus ^{127}I ($E_{\gamma}^{(0)} = 203$ keV) linked with the molecule H^{127}I .

1. Introduction

In the last two decades the photo excitation and ionization of molecules, semiconductors and solids by neutrons, photons, electrons has attracted widespread research (c.f. [1–8]). It is stimulated by appearance of the intensive neutron pencils, powerful laser sources etc. Our paper is devoted to studying co-operative dynamical phenomena due to the interaction between photons, nuclei and electron shells in atomic and molecular systems. In this direction one could consider the following problems: i) the exploration of mixed optical quantum photo transitions and laser-electron-nuclear photo processes in molecular systems, ii) spectroscopy of resonances and the creation of additional satellites and narrow resonances inside the Doppler contour of radiation line, and iii) governing by intensity of the complicated transitions due to the change of atomic and molecular excited state populations in a laser field. We present a consistent approach to the description of new class of the dynamical laser-electron-nuclear effects in molecular systems. In particular, the exploration of nuclear emission or absorption spectrum for diatomic molecules is a subject of our interest. This spectrum contains a set of electron-vibration-rotational satellites, which are arisen due to the alteration of the corresponding molecular states during the interaction of molecules with intense laser (gaser)fields [1, 4–8]. The mechanism of forming the corresponding satellites in molecular spectra is connected with the shaking of electron shells resulting from the interaction between nuclei and γ quanta. 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 atoms the corresponding satellite spectrum is much enriched. One could consider the transitions between the fine structure components, 0–0 transitions and transitions, which do not involve the change in electron configurations. Compared to atoms, molecules represent a more complex

class of system to their multicenter nature which introduces additional and rotational degrees of freedom. Compared to atomic spectra, molecular spectrum is naturally more complicated. Due to the nuclear γ -quantum emission or absorption the electron, vibrational and rotational states are changed. Here we present a consistent, quantum-mechanical approach to the calculation of electron-nuclear γ spectra (set of vibrational and rotational satellites) for nuclei in diatomic molecules. It generalizes the well-known Letokhov-Minogin model [1] and is based on using the Durham model potential approximation for the potential curves of diatomic molecules [9, 10]. The method is applied to the calculation of probabilities of the vibration-rotation-nuclear transitions in a case of emission and absorption spectrum for the nucleus ^{127}I ($E_\gamma^{(0)} = 203$ keV) linked with the molecule H^{127}I .

2. Quantum approach to the calculation of electron-nuclear γ transition spectra in diatomics

Our purpose is the exploration of spectrum of the gamma satellites because of the changing electron-vibration-rotational states of diatomic molecules due to gamma quantum radiation (absorption). In adiabatic approximation the wave function of molecule can be represented as the multiplying electronic wave function and wave function of nuclei: $\psi(r_e)\psi(R_1, R_2)$. Hamiltonian of the interaction of gamma radiation with system of nucleons for the first nucleus can be expressed in the co-ordinates of nucleons r'_n in a system of mass centre for the first nucleus [1, 5, 6]:

$$H(r_n) = H(r_n) \exp(-ik_\gamma R_1), \quad (1)$$

k_γ being the wave vector of gamma quantum. The matrix element of transition from the initial state 'a' to the final state 'b' is presented as follows:

$$\langle \Psi_b^*(r_n) | H(r_n) | \Psi_a(r_n) \rangle \langle \Psi_b^*(r_e) Psi_b^*(R_1, R_2) | e^{-ik_\gamma R_1} | \Psi_a(r_e) Psi_a(R_1, R_2) \rangle. \quad (2)$$

The first multiplier in Eq. (1) is defined by gamma transition of nucleus. It is not dependent upon the internal structure of molecule in good approximation. The second multiplier is the matrix element for the transition of molecule from the initial state 'a' to the final state 'b':

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

The expression (3) gives general formula for the calculation of probabilities of the changing internal molecular states due to absorption or emitting gamma quantum by nuclei in diatomic molecules and defines the amplitude of gamma satellites. Their positions are fully determined by the energy and pulse conservation laws as follows [1]:

$$\pm E_\gamma + E_a + (1/2)Mv_0^2 = \pm E_\gamma^{(0)} + E_b + (1/2)Mv^2, \quad (4)$$

$$Mv_0 \pm \hbar k_\gamma = Mv. \quad (5)$$

Here M is the mass of molecule, v_0 and v are the velocities of molecule before and after the interaction of nucleus with γ quantum, E_a and E_b are the energies of molecule before and after the interaction, E_γ is the energy of nuclear transition. Then the energy of γ satellite is as follows from Eq. (4):

$$E_\gamma = E_\gamma^{(0)} + \hbar k_\gamma v_0 \pm R_{om} \pm (E_b - E_a). \quad (6)$$

Here R_{om} is the energy of recoil: $R_{om} = [(E_\gamma^{(0)})^2/2Mc^2]$. It is well known (c.f. [3, 6]) that only the transitions between vibrational and rotational levels of ground electron state including transitions into continuum with the further molecular dissociation have a practical interest. The matrix element of the transition for these transitions is as follows:

$$M_{ba} = \langle \Psi_b^*(R_1, R_2) | e^{ik_\gamma R_1} | \Psi_a(R_1, R_2) \rangle. \quad (7)$$

The values of energy accepted by vibrational and rotational degrees of freedom of the molecule are as follows:

$$\begin{aligned}\epsilon_{vib} &\approx v\hbar\omega = R_{om}(m_2/m_1), \\ \epsilon_{rot} &\approx BJ^2 = R_{om}(m_2/m_1).\end{aligned}\quad (8)$$

The simple adequate model for the definition of rotational motion is known as the rigid rotator approximation. In this approximation the wave functions with definite values of quantum numbers J, K are the eigen functions of the angle momentum operator, i.e.:

$$\psi(R_1, R_2) = Y_{J,K}(\theta, \varphi). \quad (9)$$

In a case of vibration motion the wave functions with definite values of vibrational quantum numbers are numerically found by solving the corresponding Schrödinger equation with some. Below this potential is chosen by us in the Dunham-like form (c.f. [9, 10]):

$$E(R) = B_0[(R - R_e)/R]^2 \left\{ 1 + \sum_{n=1}^{\infty} b_n[(R - R_e)/R]^2 \right\}. \quad (10)$$

Compared to the well known harmonic oscillator model, this approximation is surely more consistent. The harmonic oscillator wave functions were used for the calculation of matrix elements for the vibration-transitions in ref. [3]. The matrix element of vibration-rotation-nuclear transition can be written as follows:

$$\begin{aligned}M_{J_b, K_b; J_a, K_a}^{v_b, v_a} &= (4\pi)^{1/2} [(2J_a + 1)(2J_b + 1)]^{1/2} (-1)^{K_b} \times \\ &\times \sum_{l=|J_b - J_a|}^{J_a + J_b} i^l (2l + 1)^{1/2} \langle \Psi_{v_b} | (\pi/2a)^{1/2} J_{l+1/2}(a) | \Psi_{v_a} \rangle \begin{pmatrix} J_a & J_b & l \\ 0 & 0 & 0 \end{pmatrix} \sum_{m=-l}^{+l} Y_{lm}^* \begin{pmatrix} J_a & J_b & l \\ k_a & -k_b & m \end{pmatrix},\end{aligned}\quad (11)$$

$$a = \frac{E_\gamma^{(0)}}{\hbar c} \frac{m_2}{M} R^* \left(1 + \frac{Q}{\sqrt{mR^*}} \right).$$

Here $Q = (R - R_0)m^{1/2}$, $m = m_1 m_2 / M$ is the reduced mass of molecule, m_1 and m_2 are the masses of nuclei. The co-ordinate of mass centre for the first nucleus relatively the molecule mass centre is defined by the following expression:

$$R_1 = -\frac{m_2}{M} R = -\frac{m_2}{M} \left(R_0 + \frac{Q}{\sqrt{m}} \right) = -\frac{m_2}{M} R_0 - \sqrt{\frac{m_2}{m_1 M}} Q.$$

The corresponding probability can be written as follows:

$$P_{J_b J_a}^{v_b v_a} = (2J_b + 1) \sum_{l=|J_a - J_b|}^{J_a + J_b} (2l + 1) \langle \Psi_{v_b} | \sqrt{(\pi/2a)} J_{l+1/2}(a) | \Psi_{v_a} \rangle^2 \begin{pmatrix} J_a & J_b & l \\ 0 & 0 & 0 \end{pmatrix}^2. \quad (12)$$

3. Numerical results and discussion

We have carried out the calculation of probabilities for vibration-rotation-nuclear transitions from the initial states with $v_a = 0$, $J_a = 0$ and $v_a = 1$, $J_a = 0$ in emission and absorption spectrum of the nucleus ^{127}I ($E_\gamma^{(0)} = 203$ keV) linked with the molecule H^{127}I in the ground electron state $X^1\Sigma$ (molecular parameters: $R_0 = 1.61\text{\AA}$, $\nu_e = 2309$ cm^{-1} , $B = 6.55$ cm^{-1}). The

recoil energy for this molecule is 0.172 eV. The parameters $a_0 = 1.30$ and $\epsilon_0 5.29 \cdot 10^{-2}$ define the excitation of vibrations and rotations in this molecule due to the recoil. It should be also noted that the width of gamma lines is corresponding to temperature $T = 300$ K. We present the calculated spectra of emission and adsorption of the nucleus ^{127}I in H^{127}I in Figure 1 (Fig. 1a is corresponding to the initial state of molecule: $v_a = 0, J_a = 0$; Fig. 1b $\sim v_a = 1, J_a = 0$). It should be noted that our values of probabilities (calculated within the Dunham model potential approximation for potential curve [9, 10]) differ from the corresponding ones, calculated within the harmonic oscillator approximation [1]. This difference is 5–20% in average. It is obvious that the direct experimental observation of laser-electron-nuclear effects is of a great theoretical interest and significance in science and technology.

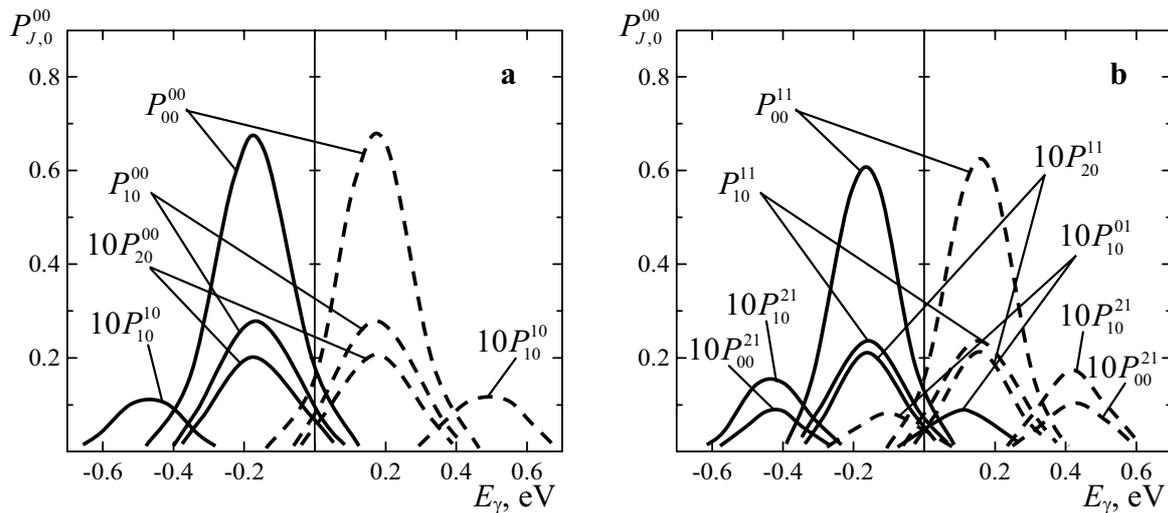


Figure 1. Calculated emission (solid curve) and absorption spectrum of nucleus ^{127}I ($E_\gamma^{(0)} = 203$ keV) linked with molecule H^{127}I . Initial state of molecule: a) above $v_a = 0, J_a = 0$ and b) below $v_a = 1, J_a = 0$.

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