UDC 539.183

A. V. Glushkov, P. A. Kondratenko, Yu. M. Lopatkin, V. V. Buyadzhi, A S. Kvasikova

[1]National Aviation University, 1, Komarov Ave, 03680 Kiev, Ukraine
[2]Odessa National Polytechnical University, 1, Shevchenko av., Odessa, Ukraine
[3]Odessa State Environmental University, 15, Lvovskaya str., Odessa, Ukraine
e-mail: dirac13@mail.ru

SPECTROSCOPY OF COOPERATIVE LASER ELECTRON-γ NUCLEAR PROCESSES IN MULTIATOMIC MOLECULES: OsO_4

The consistent quantum approach to calculating the electron-nuclear γ transition spectra (a set of the vibration-rotational satellites in a spectrum of molecule) of a nucleus in the multiatomic molecules is used to obtain the accurate data on the vibration-nuclear transition probabilities in a case of the emission and absorption spectrum of the nucleus ^{188}Os ($E^{(0)}_{\gamma}$= 155 ke V) in the molecule of OsO_4. Analysis shows that more sophisticated theoretical approach gives the higher values for the cited probabilities.

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 the transfer of part of a nuclear energy to atom or molecule under radiating (absorption) the **γ** quanta by a nucleus (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 photon. 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]. An appearance of the molecular nuclear lines represents a great interest as it opens a possibility of the changing **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 paper is going on our studying 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 multiatomic molecules has been earlier proposed [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 ^{191}Ir ($E^{(0)}_{\gamma}$= 82 keV) in the molecule of IrO_4 and other molecules were listed . Here we present the accurate data on the vibration-nuclear transition probabilities in a case of the emission and absorption spectrum of nucleus nucleus ^{188}Os ($E^{(0)}_{\gamma}$= 155 keV) in the molecule of OsO_4.

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

As the method of computing is earlier presented in details [14-16], here we consider only by the key topics The main purpose is calculating a structure of the gamma transitions (a probability of transition) or spectrum of the gamma satellites because of changing the electron-vibration-rotational states of the multi-atomic molecules under gamma quantum radiation (absorption). Here we are limited by a case of the five-atomic molecules (of XY_4 type; T_d). Hamiltonian of interaction of the gamma 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]:

$$H(r_n) = H(r_n^{'})\exp(-k_{\gamma}u) \tag{1}$$

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 in the space. The matrix element for transition from the initial state "a" to the final state "b" is presented as usually:

$$<\Psi_b^* \mid H \mid \Psi_a> \bullet <\Psi_b^* \mid e^{-ik_{\gamma}u} \mid \Psi_a> \tag{2}$$

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 a good approximation. The second multiplier is the matrix element of transition of the molecule from the initial state "a" to the final state "b":

$$\begin{aligned} M_{ba} &= <\Psi_b^*(r_e) \mid \Psi_a(r_e)> \bullet \\ &\bullet <\Psi_b^*(R_1,R_2) \mid e^{-ik_{\gamma}R_1} \mid \Psi_a(R_1,R_2)> \end{aligned} \tag{3}$$

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

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

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: $R_{om}= [(E_{\gamma}^{(o)}]^{2}/2Mc^{2}$. 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:

$$\begin{aligned} \overline{E}_{\text{vib}} &= \sum_{v=0}^{\infty} \hbar\omega\left(v+ {}^1\!/\!_2\right)\overline{P}(v,0) - \hbar\infty/2 = \\ &= \sum_{v=0}^{\infty} \hbar\omega\left(v+ {}^1\!/\!_2\right)P(v,0) - \hbar\omega/2 = \\ &= \sum_{v=0} \hbar\omega\left(v+ {}^1\!/\!_2\right)\frac{z^v}{v!}e^{-z} - \frac{\hbar\omega}{2} = \frac{1}{2}R\left(\frac{M-m}{m}\right), \end{aligned} \tag{5}$$

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 $\overline{E}_{\text{vib}}$ 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 momentum $L=k_v u \sin\vartheta$, which is transferred to a molecule by γ-quantum. In supposing that a nucleus is only in the single non-generated normal vibration and vibrational state of a molecule is not changed $v_a=v_b=0$, one could evaluate an averaged energy for excitation of the molecule rotations as follows:

$$\begin{aligned} \overline{E}_{\text{rot}} &= \left\langle \overline{BL^2} \right\rangle = Bk_{\gamma}^{\;2}\left\langle u^2 \right\rangle \overline{\sin^2\vartheta} = \\ &= {}^1\!/\!_2 R(B/\hbar\omega)[(M-m)/m] \end{aligned} \tag{6}$$

As for multi-atomic molecules it is typical $B/\hbar\omega \sim 10^{-4}\text{-}10^{-2}$, so one could miss the molecule rotations and consider γ-spectrum of a nucleus in the molecule mass centre as a spectrum of the vibration-nuclear transitions. A shift u of the γ-active nucleus can be expressed through the normal co-ordinates $Q_{s\sigma}$ of a molecule:

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

where m is a mass of the γ- active nucleus; components of the vector $b_{s\sigma}$ of nucleus shift due to the σ-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 eq.(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:

$$M(b,a) =$$

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

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

$$|\, v_s \rangle = \Phi_v (Q_s), \qquad (9)$$

for double degenerated vibration in the form:

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

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

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

$$\times \sum_{v\,\sigma_1, v\,\sigma_2, v\,\sigma_3} \Phi_{v_{s\sigma 1}}(Q_{s\sigma_1}) \Phi_{v_{s\sigma_2}}(Q_{s\sigma_2}) \Phi_{v_{s\sigma_3}}(Q_{s\sigma_3}) \qquad (11)$$

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 chosen in a form of the linear harmonic oscillator one. More exact calculating requires a numerical determination of these functions. Taking directly the wave functions $|v_s^a\rangle$ and $|v_s^b\rangle$, calculating the matrix element (8) is reduced to a definition of the matrix elements on each component γ of the normal vibration.

3. Results and conclusions

Below we present the accurate data on the vibration-nuclear transition probabilities in a case of the emission and absorption spectrum of nucleus ^{188}Os ($E^{(0)}_\gamma$ = 155 keV in the molecule OsO_4. Note that the main difficulty during calculating (8) 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 Eccart conditions, normalization one and data about the molecule symmetry. For several normal vibrations of the one symmetry type, a definition of $b_{s\gamma}$ requires solving the secular equation for molecule $|GF\text{-}\lambda E|=0$ [21-24]. We have used the results of advanced theoretical calculating electron structure of the studied system within an advanced relativistic scheme of the X_γ- scattered waves method (see details in Refs.[21,24]). In table 1 we present the results of calculating probabilities of the first several vibration-nuclear transitions in a case of the emission and absorption spectrum of nucleus ^{188}Os ($E^{(0)}_\gamma$= 155 keV in OsO_4 and compare with available data Ref.[2], where more simple approximation has been used. Analysis shows that more sophisticated calculation gives the higher values for probabilities.

Table 1

Probabilitites of the vibrational-nuclear transitions in spectrum of OsO_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	0.795	0.803
1,0 – 0,0	0.018	0.023
0,1 – 0,0	0.074	0.081
1,0 – 1,0	0.750	0.757
0,1 – 0,1	0.673	0.678

References

1. Letokhov V S, 1997 Laser Spectroscopy, Academic Press, N.-Y.
2. Letokhov V S, 1974 *Phys. Lett.A* 46 257.
3. Letokhov V S and Minogin V G, 1995 *JETP.* 69(5) 1568.
4. Ivanov L N, Letokhov V S 1995 *JETP* 68 1748;
5. Glushkov A V, Loboda A, Gurnitskaya E, Svinarenko A 2009 *Phys. Scripta* T135 014022
6. Glushkov A V, 2012 *J. of Phys.: Conf. Ser.*. 397 012011
7. Glushkov A V and Ivanov L N 1992*Phys. Lett.* A 170 33
8. Law J and Campbell J L,1992 *Phys. Rev.C* 25 514.
9. Amudsen P A and Barker P H, 1999 *Phys.Rev.C* 50 2466
10. L Wauters, N Vaeck, M Godefroid, H var der Hart and M Demeur, 1997 *J.Phys.B.* 30 4569.
11. Shahbaz A, Müller C, Staudt A, Bürvenich T J, Keitel C H 2007 *Phys.Rev. Lett.* 98 263901
12. Fedorov M V 1999 *Phys.-Uspekhi* 169 66
13. Ivanov L N and Letokhov V S *Com. Mod. Phys.* D 4 169
14. A V Glushkov, O Yu Khetselius, Malinovskaya S V, 2008 *Europ. Phys. Journ ST* 160 195.
15. A V Glushkov, O Yu Khetselius, Malinovskaya S V, 2008 *Molec.Phys.* 106 1257.
16. Glushkov A V, Khetselius O Yu, Malinovskaya S V, 2008 *Frontiers in Quantum Systems in Chem. and Phys., Ser. Progr. in Theor. Chem. and Phys.*, vol. 18. ed. S Wilson, P J Grout, J Maruani, G; Delgado-Barrio, P Piecuch (Berlin: Springer) p 523
17. Glushkov A V, Malinovskaya S V, Svinarenko A A and Chernyakova Yu G, 2004 *Int. J. Quant. Chem.* 99 889.
18. Glushkov A , Ivanov L, Ivanova E, 1996 *Autoionization Phenomena in Atoms*, Moscow Univ. Press, Moscow, p 58
19. Khetselius OYu, 2012 *J. of Phys.: Conf. Ser.* 397 012012;
20. Khetselius OYu, 2009 *Phys.Scr.* T.135 014023.
21. Glushkov A, 1993 Journ.Struct. Chem. 34(1) 3.
22. Glushkov A.V. and Malinovskaya S.V., 1998 *Russian J.Phys.Chem.* 62(1) 100.
23. G Simons and R G Parr, 2002 Quantum Chemistry, Academic Press, N-Y.
24. V N Gedasimov, A G Zelenkov, V M Kulakov, V A Pchelin, M V Sokolovskaya, A A Soldatov, L V Chistyakov, 1994 *JETP.* 86 1169; A A Soldatov, 1993 Preprint of I.V.Kurchatov Institute for Atomic Energy IAE-3916, Moscow

This article has been received within 2014

UDC 539.183

A. V. Glushkov, P. A. Kondratenko, Yu. M. Lopatkin, V. V. Buyadzhi, A. S. Kvasikova

SPECTROSCOPY OF COOPERATIVE LASER ELECTRON-G-NUCLEAR PROCESSES IN MULTIATOMIC MOLECULES: OsO_4

Abstract

The consistent quantum approach to calculating the electron-nuclear g transition spectra (a set of the vibration-rotational satellites in a spectrum of molecule) of a nucleus in the multiatomic molecules is used to obtain the accurate data on the vibration-nuclear transition probabilities in a case of the emission and absorption spectrum of the nucleus ^{188}Os ($E^{(0)}_g$= 155 keV) in the molecule of OsO_4. Analysis shows that more sophisticated theoretical approach gives the higher values for the cited probabilities.

Key words: electron-g-nuclear transition spectrum, multiatomic molecules

УДК 539.183

А. В. Глушков, П. А. Кондратенко, Ю. М. Лопаткин, В. В. Буяджи, А. С. Квасикова

СПЕКТРОСКОПИЯ КООПЕРАТИВНЫХ ЛАЗЕРНЫХ ЭЛЕКТРОННО-ГАММА-ЯДЕРНЫХ -ПРОЦЕССОВ В МНОГОАТОМНЫХ МОЛЕКУЛАХ: OsO_4

Резюме

Последовательный квантовый подход к расчету электронно-гамма-ядерного спектра (система колебательно-вращательных спутников в спектре молекуле) в многоатомных молекулах используется, чтобы получить уточненные данные о вероятностях колебательно-ядерных переходов в молекуле OsO_4 в случае испускания и поглощения гамма-кванта ядром ^{188}Os ($E^{(0)}_g$= 155 kэВ). Анализ показывает, что более последовательный теоретический подход дает более высокие значения искомых вероятностей.

Ключевые слова: спектр электрон- g -ядерных переходов, многоатомные молекулы

УДК 539.183

О. В. Глушков, П. О. Кондратенко, Ю. М. Лопаткін, В. В. Буяджи, Г. С. Квасикова

СПЕКТРОСКОПІЯ КООПЕРАТИВНИХ ЛАЗЕРНИХ ЕЛЕКТРОННО-ГАММА-ЯДЕРНИХ ПРОЦЕСІВ У БАГАТОАТОМНИХ МОЛЕКУЛАХ: OsO_4

Резюме

Послідовний квантовий підхід до розрахунку електронно-гамма-ядерного спектру (система коливально-обертальних супутників у спектрі молекули) в багатоатомних молекулах використовується, щоб отримати уточнені дані по ймовірностям коливально-ядерних переходів в молекулі OsO_4 у випадку випромінювання та поглинання гамма-кванта ядром ^{188}Os ($E^{(0)}_g$= 155 кеВ). Аналіз показує, що більш послідовний теоретичний підхід дає більш високі значення шуканих ймовірностей.

Ключові слова: спектр електрон- g -ядерних переходів, багатоатомні молекули