ANALYSIS OF THE INTERACTION BETWEEN MOLECULES
AND IR-LASER RADIATION USING ONE-DIMENSIONAL LIE
GROUPS

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The effect of IR-laser radiation on molecules has been examined by using one-dimensional Lie groups in a
triangular matrix to solve the equation for a nonuniform harmonic oscillator. The "red shift" effect is
explained.

The analysis and systematics of the energy levels of complex atomic and molecular systems on the basis of one-
dimensional Lie groups is currently undergoing intense development in a number of directions, initiated by the problems of
elementary particle physics. It is noteworthy that useful information can be obtained even without knowing in detail the nature
and rules of the interaction of elementary objects and the dynamics of their interaction. This development is based on the
expansion of Lie groups or the introduction of additional parameters, the quantum numbers [1].

In this paper, as in previous work [2, 3], a somewhat different approach is taken, based on the determination of
possible assignments of the one-dimensional Lie subgroups corresponding to the dynamic variables of the problem.

According to [4] the categories of representations for continuous one-dimensional Lie groups are defined as the general
form of exp in the group $g_{i}$ :

$$\{ f(x+y) = f(x)f(y) \rightarrow [f(g_{1},g_{2}) = f(g_{1})f(g_{2})] \} .$$

where $g_{1}$ and $g_{2}$ are group elements. In general the solution (1) is insufficient for the analysis of some arbitrary functions of
group $G$, for example for noncommutative groups. Additional solutions can be obtained by changing from scalar to matrix
functions, i.e., linear transformations. For a finite order representation in the basis $\{ i \}$ each operator of the representation $T(g)$
of the matrix $t_{ij}$ has the general form:

$$T(g)e_{i} = \sum_{i=1}^{n} t_{ij}g_{i}e_{j} \quad (T(g) = t_{ij}(g)).$$

For one-dimensional subgroups of group $G$ with representation $T(g)$ an infinitesimal operator $A$ is introduced:

$$A = \lim_{t \to 0} \frac{T(g(t)) - E}{t} \equiv \left( \frac{dT(g)}{dt} \right)_{t=0},$$

where $E$ is a unit matrix and the matrix tangent is

$$\{ e^{\text{tan}} = \left( \frac{dg(t)}{dt} \right)_{t=0} \} .$$

The linear extent of $a_{i}$ together with the commutator $[a_{i}, a_{j}] = a_{i}a_{j} - a_{j}a_{i}$ is described by Lie matrix algebra. In the finite
dimensional case the representation $T(g)$ for the one-parameter subgroup $g(t)$ is determined by solution of the infinitesimal
operator of the subgroup according to the formula

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In the given case the required Lie group is recovered during analysis of the most interesting effect in laser photochemistry, the frequency shift of the 0 → 1 transition during resonance interaction of powerful short impulse IR-laser irradiation. This effect was first observed in gaseous boron trichloride [5] and it has been confirmed in other materials and has been adequately analyzed [6]. Possible explanations have been based on the importance of electron-vibrational interactions in polyatomic molecules in which the electronic and vibrational levels converge [7]. Raising of degeneracy and Fermi resonance leads to the formation of a quasicontinuum of definite energy zones with allowed transitions. The conditions for interaction of laser radiation with such a zone requires a red shift of the 0 → 1 transition frequency.

Calculation of the frequency shift for a vibrational transition under the influence of laser radiation for an arbitrary polyatomic molecule leads to a two level approximation. For the vibrational mode $b_1^+$ we have [8]:

$$i\hbar\dot{b}_1^+ = -\hbar\omega b_1^+ - f\sqrt{2}g_0[\sigma_+D(f\sqrt{2}) - \sigma_-D^*(f\sqrt{2})]$$

and

$$i\hbar\dot{\alpha}_0 = 2g_0[\sigma_-^*D(f\sqrt{2}) - \sigma_-D(f\sqrt{2})]$$

$$i\hbar\dot{\alpha}_+ = -g_0\sigma_0D^*(f\sqrt{2})$$

$$i\hbar\dot{\alpha}_- = g_0\sigma_0D(f\sqrt{2})$$

where $f$ is the electron-vibrational interaction parameter, $I$ is the electron-photon interaction parameter, $\sigma_+$, $\sigma_-$, and $\sigma_0$ are Pauli operators, $D(\alpha) = \exp[-\alpha(b_1 - b_1^+)]$ is the mixing operator; and $g_0 = f\hbar\omega/\sqrt{2}(b_1^2 + b_2) - i\hbar\omega/\sqrt{2}(a^+ - a)$, where $a^+$ and $a$ are the photon field operators. Introducing an operator with the coordinates $q = (b_1^+ + b_1)q_0$, $q_0 = \sqrt{\hbar}2m\omega$ and the dimensionless parameter $\tilde{g}_0 = (g_0)/\hbar\omega$, we obtain

$$\ddot{q} + \omega^2q + 2\omega^2\tilde{g}_0[\sigma_+D(f\sqrt{2}) - \sigma_-D^*(f\sqrt{2})]q =$$

$$= 2\tilde{g}_0f\sqrt{2}[\sigma_-D(f\sqrt{2}) - \sigma_-^*D(f\sqrt{2})]q + 4\tilde{g}_0^2\sqrt{2}\sigma_0\omega^2$$

or more simply

$$\ddot{q} + \omega^2x^2q = \Lambda x^2\omega^2.$$  

(6)

Apparently systematization of the energy levels of the harmonic oscillator was first carried out on the basis of Heisenberg—Weil groups [9].

In this paper (6) is solved by using the operator $q$ in the form of one-parameter subgroups of the third order triangular matrix $G_3(t)$

$$g = \begin{pmatrix} 1 & a & b \\ 0 & c & d \\ 0 & 0 & 1 \end{pmatrix},$$

where $a$, $b$, $c$, and $d$ are real numbers with $c > 0$. The following were chosen as one-parameter subgroups:

$$g_+(t) = \begin{pmatrix} 1 & t & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad g_-(t) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & t \\ 0 & 0 & 1 \end{pmatrix},$$

$$Z(t) = \begin{pmatrix} 1 & 0 & t \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad e(t) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & e^t & 0 \\ 0 & 0 & 1 \end{pmatrix}.$$  

(8)