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Study on infrared multiphoton excitation of the linear triatomic molecule by the Lie-algebra approach

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Abstract. Infrared multiphoton vibrational excitation of the linear triatomic molecule has been studied using the quadratic anharmonic Lie-algebra model, unitary transformations, and Magnus approximation. An explicit Lie-algebra expression for the vibrational transition probability is obtained by using a Lie-algebra approach. The infrared multiphoton vibrational excitation of the DCN molecule is discussed as an example.

PACS. 33.80.Wz Other multiphoton processes – 02.20.Sv Lie algebras of Lie groups

1 Introduction

Multiphoton processes play an important role in many areas of science, including photochemistry [1], fluorescence imaging [2], and photoionization[3]. The study of atomic and molecular multiphoton processes has attracted considerable attention [4–6] in the past two decades because of the development of high-power and short-pulse laser technology. Some theoretical and computational methods have been developed for improvements in experiment, such as the Coulter transformation method, and the Floquet theory method [7–10]. In recent years, the Lie-algebra method has been proposed to solve the question of polyatomic molecular vibrations [11–15]. The multiphoton question in intense laser fields of a diatomic molecule has been successfully studied by the Lie-algebra method [16–20]. However, compared to the atomic case, the interaction of molecules with intense laser fields is considerably more complicated. Here, we introduce a theoretical method for studying the infrared multiphoton excitation of the linear triatomic molecule. The quadratic anharmonic algebra model [21] is used in this research. The explicit Lie-algebra expression for the time-evolution operator is obtained by using a Lie-algebraic approach, so that the problem of solving the Schrödinger equation is reduced to that of solving a set of ordinary nonlinear differential equations, and the computation time is greatly reduced.

The paper is organized as follows. In Section 2 we derive the Hamiltonian of the system in the interaction picture within the semiclassical approximation (wherein the molecule is treated quantum mechanically and the external field is assumed to be classical), while the solution of the time-evolution operator for the system, and the explicit Lie-algebra expression of the vibrational transition probability are also presented. In Section 3, we present an example and give a brief discussion. Conclusions are then given in Section 4.

2 Theory

2.1 The Hamiltonian of the system

The Hamiltonian of the system is

$$\mathcal{H} = \mathcal{H}_m + \mathcal{H}_\varepsilon, \tag{1}$$

where $\mathcal{H}_m$ denotes the Hamiltonian of a free linear triatomic molecule (ABC), which can be represented as two coupled quadratic anharmonic oscillators [21]:

$$\mathcal{H}_m = \hbar\omega_{0l}\left(\hat{A}_l^+\hat{A}_l^- + \frac{\hat{I}_{0l}}{2}\right) + \hbar\omega_{0r}\left(\hat{A}_r^+\hat{A}_r^- + \frac{\hat{I}_{0r}}{2}\right) - \lambda\left(\hat{A}_l^+\hat{A}_r^- + \hat{A}_r^+\hat{A}_l^-\right), \tag{2}$$

where the 'l' and 'r' subscripts denote the left (A-B) and right (B-C) bonds, respectively, ω_{0l} and ω_{0r} are the corresponding angular frequencies of the anharmonic oscillators, and λ is the coupling coefficient between the two chemical bonds and can be determined by the experimental values.

$\hat{A}_i^+$ and $\hat{A}_i^-$ $(i = l, r)$ have the commutation relations [21]

$$\left[\hat{A}_i^-, \hat{A}_i^+\right] = \hat{I}_{0i}, \quad \left[\hat{I}_{0i}, \hat{A}_i^\pm\right] = \mp 2x_{0i}\hat{A}_i^\pm, \tag{3}$$

in which $x_{0i} = a_i/(2\sqrt{2m_iD_i})$ is the anharmonicity parameter [21], where m_i, a_i and D_i $(i = l, r)$ denote the reduced mass, Morse parameters and the dissociation energy, respectively.

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We assume that the molecule is in line with the direction of the laser fields. $\mathcal{H}_\varepsilon$ represents the interaction between the molecule and the infrared laser field

$$\mathcal{H}_\varepsilon(\boldsymbol{R}_l, \boldsymbol{R}_r, t) = -\mu(\boldsymbol{R}_l, \boldsymbol{R}_r) E_0 \cos\omega_L t, \tag{4}$$

where E_0 and ω_L are the laser amplitude and angular frequency, respectively. The dipole moment $\mu(\boldsymbol{R}_l, \boldsymbol{R}_r)$ can be expanded to first order at the equilibrium nuclear configuration [22]

$$\begin{aligned} \mu(\boldsymbol{R}_l, \boldsymbol{R}_r) &\approx \mu_0 + \left(\frac{\partial\mu}{\partial\boldsymbol{R}_l}\right)_0 (\boldsymbol{R}_l - \boldsymbol{R}_l^0) \\ &+ \left(\frac{\partial\mu}{\partial\boldsymbol{R}_r}\right)_0 (\boldsymbol{R}_r - \boldsymbol{R}_r^0) = \mu_0 + \mu_l \hat{x}_l + \mu_r \hat{x}_r, \end{aligned} \tag{5}$$

where $\hat{x}_l = \boldsymbol{R}_l - \boldsymbol{R}_l^0$, $\hat{x}_r = \boldsymbol{R}_r - \boldsymbol{R}_r^0$, and $\boldsymbol{R}_l^0$ and $\boldsymbol{R}_r^0$ are the equilibrium nuclear separations of AB and BC.

Then the quadratic anharmonic Lie-algebra Hamiltonian of the system is

$$\begin{aligned} \mathcal{H}_m = &\hbar\omega_{0l}\left(\hat{A}_l^+\hat{A}_l^- + \frac{\hat{I}_{0l}}{2}\right) + \hbar\omega_{0r}\left(\hat{A}_r^+\hat{A}_r^- + \frac{\hat{I}_{0r}}{2}\right) \\ &- \lambda\left(\hat{A}_l^+\hat{A}_r^- + \hat{A}_r^+\hat{A}_l^-\right) - \frac{d_1}{\sqrt{2}}\left(\hat{A}_l^+ + \hat{A}_l^-\right) \\ &- \frac{d_2}{\sqrt{2}}\left(\hat{A}_r^+ + \hat{A}_r^-\right) - d_0, \end{aligned} \tag{6}$$

where

$$\begin{aligned} d_1 &= \frac{\mu_l}{a_l}\sqrt{\frac{\hbar\omega_{0l}}{2D_l}} E_0 \cos\omega_L t, \\ d_2 &= \frac{\mu_r}{a_r}\sqrt{\frac{\hbar\omega_{0r}}{2D_r}} E_0 \cos\omega_L t, \quad d_0 = \mu_0 E_0 \cos\omega_L t. \end{aligned} \tag{7}$$

In the interaction picture,

$$\begin{aligned} \mathcal{H}_I(t) &= e^{i\mathcal{H}_0 t/\hbar}\mathcal{V}e^{-i\mathcal{H}_0 t/\hbar} \\ &= \left(-\frac{d_0}{2}\hat{T}_{0l} - \frac{d_1}{\sqrt{2}}B_l\hat{a}_{l+} - \frac{d_1}{\sqrt{2}}B_l\hat{a}_{l-}\right) \\ &\quad + \left(-\frac{d_0}{2}\hat{T}_{0r} - \frac{d_2}{\sqrt{2}}B_r\hat{a}_{r+} - \frac{d_2}{\sqrt{2}}B_r\hat{a}_{r-}\right) \\ &\quad - \lambda B_l B_r\left(\hat{a}_{l+}\hat{a}_{r-} + \hat{a}_{r+}\hat{a}_{l-}\right), \end{aligned} \tag{8}$$

where $\hat{T}_{0i}(i = l, r)$ is an identity operator and

$$\begin{aligned} \mathcal{H}_0 &= \hbar\omega_{0l}\left(\hat{A}_l^+\hat{A}_l^- + \frac{\hat{I}_{0l}}{2}\right) + \hbar\omega_{0r}\left(\hat{A}_r^+\hat{A}_r^- + \frac{\hat{I}_{0r}}{2}\right), \\ \mathcal{V} &= -\lambda B_l B_r\left(\hat{A}_l^+\hat{A}_r^- + \hat{A}_r^+\hat{A}_l^-\right) - \frac{d_1}{\sqrt{2}}\left(\hat{A}_l^+ + \hat{A}_l^-\right) \\ &\quad - \frac{d_2}{\sqrt{2}}\left(\hat{A}_r^+ + \hat{A}_r^-\right) - d_0, \end{aligned} \tag{9}$$

$$\begin{aligned} \hat{a}_{i+} &= e^{i\omega_{0i}t\hat{I}_{0i}}\hat{A}_i^+, \quad \hat{a}_{i-} = e^{-i\omega_{0i}t\hat{I}_{0i}}\hat{A}_i^-, \\ B_i &= e^{i\omega_{0i}x_{0i}t}, \qquad B_i^* = e^{-i\omega_{0i}x_{0i}t}. \end{aligned} \tag{10}$$

When the commutation relations are satisfied, i.e.

$$\begin{aligned} &[\hat{a}_{i+}, \hat{a}_{i-}] = -B_i^{*2}\hat{I}_{0i}, \quad \left[\hat{I}_{0i}, \hat{a}_{i\pm}\right] = \mp 2x_{0i}\hat{a}_{i\pm}, \\ &\left[\hat{T}_{0i}, \hat{a}_{i\pm}\right] = 0, \qquad\qquad \left[\hat{T}_{0i}, \hat{I}_{0i}\right] = 0, \end{aligned} \tag{11}$$

$(\hat{T}_{0l}, \hat{I}_{0l}, \hat{a}_{l+}, \hat{a}_{l-})$ or $(\hat{T}_{0r}, \hat{I}_{0r}, \hat{a}_{r+}, \hat{a}_{r-})$ can construct a four-dimensional dynamic Lie algebra.

2.2 Time-evolution operator

The time-evolution operator $\mathcal{U}_I$ satisfies the equation in the interaction picture

$$i\hbar\frac{\partial\mathcal{U}_I}{\partial t} = \mathcal{H}_I\mathcal{U}_I. \tag{12}$$

$\mathcal{H}_I$ can be divided into two parts:

$$\mathcal{H}_I = \mathcal{H}_1 + \mathcal{H}_2, \tag{13}$$

$$\begin{aligned} \mathcal{H}_1 &= \left(-\frac{d_0}{2}\hat{T}_{0l} - \frac{d_1}{\sqrt{2}}B_l\hat{a}_{l+} - \frac{d_1}{\sqrt{2}}B_l\hat{a}_{l-}\right) \\ &\quad + \left(-\frac{d_0}{2}\hat{T}_{0r} - \frac{d_2}{\sqrt{2}}B_r\hat{a}_{r+} - \frac{d_2}{\sqrt{2}}B_r\hat{a}_{r-}\right), \\ \mathcal{H}_2 &= -\lambda B_l B_r(\hat{a}_{l+}\hat{a}_{r-} + \hat{a}_{r+}\hat{a}_{l-}), \end{aligned} \tag{14}$$

and $\mathcal{U}_I$ is written as

$$\mathcal{U}_I = \mathcal{U}_1\mathcal{U}_2, \tag{15}$$

by putting equations (13) and (15) into equation (12),

$$i\hbar\frac{\partial\mathcal{U}_1}{\partial t} = \mathcal{H}_1\mathcal{U}_1, \tag{16}$$

$$i\hbar\frac{\partial\mathcal{U}_2}{\partial t} = \mathcal{H}_2'\mathcal{U}_2, \tag{17}$$

where

$$\mathcal{H}_2' = \mathcal{U}_1^{-1}\mathcal{H}_2\mathcal{U}_1. \tag{18}$$

2.2.1 Time-evolution operator $\mathcal{U}_1(t)$

According to the theory of Lie-algebra, the time-dependent evolution operator can be expressed by elements of Lie algebra [23,24]:

$$\mathcal{U}_1 = \mathcal{U}_{1l}\mathcal{U}_{1r} = \prod_{j=1}^{4} e^{\tilde{X}_j\hat{C}_{jl}} \prod_{k=1}^{4} e^{\tilde{Y}_k\hat{C}_{kr}}, \tag{19}$$

$$\begin{aligned} \mathcal{U}_{1l} &= e^{\tilde{X}_1\hat{T}_{0l}}e^{\tilde{X}_2\hat{I}_{0l}}e^{\tilde{X}_3\hat{a}_{l+}}e^{\tilde{X}_4\hat{a}_{l-}}, \\ \mathcal{U}_{1r} &= e^{\tilde{Y}_1\hat{T}_{0r}}e^{\tilde{Y}_2\hat{I}_{0r}}e^{\tilde{Y}_3\hat{a}_{r+}}e^{\tilde{Y}_4\hat{a}_{r-}}, \end{aligned} \tag{20}$$

where $\tilde{X}_j$ and $\tilde{Y}_k$ $(j, k = 1, 2, 3, 4)$ are the time-dependent complex coefficients.

$\mathcal{U}_{1l}$ and $\mathcal{U}_{1r}$ are obtained in the same way, and so we will only describe the solving process of $\mathcal{U}_{1l}$.

$\mathcal{U}_{1l}$ satisfies the dynamic equation

$$i\hbar\frac{\partial\mathcal{U}_{1l}(t,t_0)}{\partial t}\mathcal{U}_{1l}^{-1}(t,t_0)=\mathcal{H}_{1l}(t),\ \ \mathcal{U}_{1l}(t_0,t_0)=1. \tag{21}$$

By putting equation (20) into equation (21),

$$\begin{aligned} i\hbar(&\eta_0\hat{T}_{0l}+\eta_1\hat{I}_{0l}+\eta_2\hat{a}_{l+}+\eta_3\hat{a}_{l-}+\eta_4\hat{I}_{0l}\hat{a}_{l+}\\ &+\eta_5\hat{I}_{0l}\hat{a}_{l-}+\eta_6\hat{I}_{0l}^2+\eta_7\hat{a}_{l+}^2+\eta_8\hat{a}_{l+}\hat{a}_{l-}+\eta_9\hat{a}_{l-}^2)=\\ &-\frac{d_0}{2}\hat{T}_{0l}-\frac{d_1}{\sqrt{2}}B_l\hat{a}_{l+}-\frac{d_1}{\sqrt{2}}B_l\hat{a}_{l-}, \end{aligned} \tag{22}$$

where

$$\begin{aligned} \eta_0 &= \dot{\tilde{X}}_1,\\ \eta_1 &= \dot{\tilde{X}}_2-\tilde{X}_3\dot{\tilde{X}}_4B_l^{*2}-i\omega_{0l}x_{0l}^2\tilde{X}_3^2\tilde{X}_4^2B_l^{*4},\\ \eta_2 &= (\dot{\tilde{X}}_3-\dot{\tilde{X}}_4\dot{\tilde{X}}_3^2B_l^{*2}x_{0l}+4i\omega_{0l}x_{0l}^2\tilde{X}_3^2\tilde{X}_4B_l^{*2}\\ &\quad +2i\omega_{0l}x_{0l}^2\tilde{X}_3^3\tilde{X}_4^2B_l^{*4})e^{-2x_{0l}\tilde{X}_2},\\ \eta_3 &= (\dot{\tilde{X}}_4+i\omega_{0l}x_{0l}^2\tilde{X}_3\tilde{X}_4^2B_l^{*2})e^{2x_{0l}\tilde{X}_2},\\ \eta_4 &= (i\omega_{0l}\tilde{X}_3+3i\omega_{0l}x_{0l}\tilde{X}_3^2\tilde{X}_4B_l^{*2}\\ &\quad +2i\omega_{0l}x_{0l}^2\tilde{X}_3^3\tilde{X}_4^2B_l^{*4})e^{-2x_{0l}\tilde{X}_2},\\ \eta_5 &= (-i\omega_{0l}\tilde{X}_4-2i\omega_{0l}x_{0l}\tilde{X}_3\tilde{X}_4^2B_l^{*2})e^{2x_{0l}\tilde{X}_2},\\ \eta_6 &= i\omega_{0l}\tilde{X}_3\tilde{X}_4B_l^{*2}+i\omega_{0l}x_{0l}\tilde{X}_3^2\tilde{X}_4^2B_l^{*4},\\ \eta_7 &= (2i\omega_{0l}x_{0l}^2B_l^{*2}\tilde{X}_3^3\tilde{X}_4\\ &\quad +i\omega_{0l}x_{0l}^3\tilde{X}_3^4\tilde{X}_4^2B_l^{*4})e^{-4x_{0l}\tilde{X}_2},\\ \eta_8 &= -2i\omega_{0l}x_{0l}\tilde{X}_3\tilde{X}_4-2i\omega_{0l}x_{0l}^2\tilde{X}_3^2\tilde{X}_4^2B_l^{*2},\\ \eta_9 &= ix_{0l}\omega_{0l}\tilde{X}_4^2e^{4x_{0l}\tilde{X}_2}. \end{aligned} \tag{23}$$

$\hat{T}_{0l}$ is an identity operator, and so one can first get the following formula:

$$i\hbar\eta_0=-\frac{d_0}{2} \tag{24}$$

because the quadratic anharmonic creation and annihilation operators $\hat{A}_\pm$ have the relations [11]

$$\begin{aligned} \hat{I}_{0i}|N_i,v_i\rangle &= (1-2x_{0i}v_{0i})|N_i,v_i\rangle,\\ \hat{A}_i^+|N_i,v_i\rangle &= \sqrt{(1-x_{0i}v_i)(v_i+1)}|N_i,v_i+1\rangle,\\ \hat{A}_i^-|N_i,v_i\rangle &= \sqrt{[1-x_{0i}(v_i-1)]v_i}|N_i,v_i-1\rangle, \end{aligned} \tag{25}$$

where $i=l,r$. By calculating matrix elements of the two sides of equation (22) between $\langle v_l,0,v_r|$ and $|v_l,0,v_r\rangle$, and $\langle v_l+1,0,v_r|$ and $|v_l,0,v_r\rangle$, one obtains

$$\begin{aligned} i\hbar\eta_1(1-2x_{0l}v_l)&+i\hbar\eta_6(1-2x_{0l}v_l)^2\\ &+i\hbar\eta_8B_l^{*2}[1-x_{0l}(v_l-1)]v_l=0, \end{aligned} \tag{26}$$

$$i\hbar\eta_3+i\hbar\eta_5[1-x_{0l}(v_l-1)]=-\frac{d_1}{\sqrt{2}}B_l, \tag{27}$$

where $|v_l,0,v_r\rangle=|N_l,v_l\rangle|N_r,v_r\rangle$ [11], $N_l=1/x_{0l}$, and $N_r=1/x_{0r}$. Here v_l, v_r are the vibrational quantum numbers.

By putting equation (23) into equations (24), (26) and (27), the set of differential equations becomes

$$\begin{aligned} i\hbar\dot{\tilde{X}}_1 &= -\frac{d_0}{2},\\ i\hbar\dot{\tilde{X}}_2 &= -\frac{d_1}{\sqrt{2}}B_l\tilde{X}_3e^{-2x_{0l}\mathrm{Re}\tilde{X}_2}\\ &\qquad -2\omega_{01}x_{0l}\tilde{X}_3\tilde{X}_4\left\{\frac{[1-x_{0l}(v_l-1)]v_l}{1-2x_{0l}v_l}+1\right\}, \qquad (28)\\ i\hbar\dot{\tilde{X}}_4 &= -\frac{d_1}{\sqrt{2}}B_le^{-2x_{0l}\mathrm{Re}\tilde{X}_2}-2\omega_{01}x_{0l}[1-2x_{0l}(v_l-1)]\tilde{X}_4, \end{aligned}$$

with the initial condition from equation (21) that

$$\tilde{X}_j(t_0,t_0)=0\ \ (j=1,2,3,4). \tag{29}$$

At the same time, the unitary condition of the time-evolution operator must be satisfied, i.e.

$$\mathcal{U}_{1l}^{+}(t)=\mathcal{U}_{1l}^{-1}(t), \tag{30}$$

$$\begin{aligned} \mathcal{U}_{1l}^{+}(t) &= \exp\left(\tilde{X}_4^*{B_l}^2\hat{a}_{l+}\right)\exp\left(\tilde{X}_3^*{B_l}^2\hat{a}_{l-}\right)\\ &\quad\times\exp\left(\tilde{X}_2^*\hat{I}_{0l}\right)\exp\left(\tilde{X}_1^*\hat{T}_{0l}\right)\\ &= \exp\left\{\tilde{X}_1^*\hat{T}_{0l}+\left[\tilde{X}_2^*-\frac{1}{2}\left(1-\tilde{X}_2^*x_{0l}\right)\right.\right.\\ &\quad\left.\times\ \tilde{X}_3^*\tilde{X}_4^*B_l^{*2}\right]\hat{I}_{0l}+[\tilde{X}_4^*B_l^{*2}(1+\tilde{X}_2^*x_{0l})\\ &\quad-\frac{1}{3}\tilde{X}_3^*\tilde{X}_4^{*2}B_l^{*2}x_{0l}(1-\tilde{X}_2^*x_{0l})]\hat{a}_{l+}\\ &\quad\left.+[(1-\tilde{X}_2^*x_{0l})\tilde{X}_3^*B_l^{*2}]\hat{a}_{l-}\right\}, \end{aligned} \tag{31}$$

$$\begin{aligned} \mathcal{U}_{1l}^{-1}(t) &= \exp(-\tilde{X}_4\hat{a}_{l-})\exp(-\tilde{X}_3\hat{a}_{l+})\\ &\quad\times\exp(-\tilde{X}_2\hat{I}_{0l})\exp(-\tilde{X}_1\hat{T}_{0l})\\ &= \exp\left\{-\tilde{X}_1\hat{T}_{0l}+\left[\tilde{X}_2+\frac{1}{2}(\tilde{X}_2x_{0l}-1)\tilde{X}_3\tilde{X}_4B_l^{*2}\right]\hat{I}_{0l}\right.\\ &\quad+[\tilde{X}_3(\tilde{X}_2x_{0l}-1)]\hat{a}_{l+}+[-\tilde{X}_4(1+\tilde{X}_2x_{0l})\\ &\quad\left.-\frac{1}{3}\tilde{X}_3\tilde{X}_4^2B_l^{*2}x_{0l}(\tilde{X}_2x_{0l}-1)]\hat{a}_{l-}\right\}, \end{aligned} \tag{32}$$

equations (30), (31) and (32) lead to the following relations

$$\begin{aligned} \tilde{X}_3 &= \frac{L_1-iL_2}{F}{B_l}^2\tilde{X}_4^*,\\ \mathrm{Re}\tilde{X}_2 &= -\frac{|\tilde{X}_4|^2}{2+\frac{1}{3}x_{0l}|\tilde{X}_4|^2}, \end{aligned} \tag{33}$$

where

$$\begin{aligned} F &= [(1-x_{0l}\mathrm{Re}\tilde{X}_2)^2+(x_{0l}\mathrm{Im}\tilde{X}_2)^2]\left[1+\frac{1}{3}|\tilde{X}_4|^2x_{0l}\right]\\ L_1 &= \left[\frac{1}{3}x_{0l}\mathrm{Re}\tilde{X}_2-1\right][1-x_{0l}\mathrm{Re}\tilde{X}_2]-(x_{0l}\mathrm{Im}\tilde{X}_2)^2\\ L_2 &= \frac{2}{3}x_{0l}\mathrm{Re}\tilde{X}_2\mathrm{Im}\tilde{X}_2, \end{aligned} \tag{34}$$

in which $\tilde{X}_j$ can be obtained by solving equations (28), (29) and (33), and the time-evolution operator $\mathcal{U}_{1l}(t)$ is as follows:

$$\begin{aligned}\mathcal{U}_{1l}(t) = &\exp\left(-i\frac{d_0}{2\hbar}t\hat{T}_{0l}\right)\\ &\times\exp\left[\left(-\frac{|\tilde{X}_4|^2}{2+\frac{1}{3}x_{0l}|\tilde{X}_4|^2}+i\mathrm{Im}\tilde{X}_2\right)\hat{I}_{0l}\right]\\ &\times\exp\left[\left(\frac{L_1-iL_2}{F}\tilde{X}_4^*{B_l^*}^2\right)\hat{a}_{l+}\right]\exp(\tilde{X}_4\hat{a}_{l-}).\end{aligned}\tag{35}$$

Similarly,

$$\begin{aligned}\mathcal{U}_{1r}(t) = &\exp\left(-i\frac{d_0}{2\hbar}t\hat{T}_{0r}\right)\\ &\times\exp\left[\left(-\frac{|\tilde{Y}_4|^2}{2+\frac{1}{3}x_{0r}|\tilde{Y}_4|^2}+i\mathrm{Im}\tilde{Y}_2\right)\hat{I}_{0r}\right]\\ &\times\exp\left[\left(\frac{L_1^{'}-iL_2^{'}}{F^{'}}\tilde{Y}_4^*{B_r^*}^2\right)\hat{a}_{r+}\right]\exp(\tilde{Y}_4\hat{a}_{r-}),\end{aligned}\tag{36}$$

where

$$\begin{aligned}F^{'} &= [(1-x_{0r}\mathrm{Re}\tilde{Y}_2)^2+(x_{0r}\mathrm{Im}\tilde{Y}_2)^2]\left[1+\frac{1}{3}|\tilde{Y}_4|^2x_{0r}\right]\\ L_1^{'} &= \left[\frac{1}{3}x_{0r}\mathrm{Re}\tilde{Y}_2-1\right][1-x_{0r}\mathrm{Re}\tilde{Y}_2]-(x_{0r}\mathrm{Im}\tilde{Y}_2)^2\\ L_2^{'} &= \frac{2}{3}x_{0r}\mathrm{Re}\tilde{Y}_2\mathrm{Im}\tilde{Y}_2.\end{aligned}\tag{37}$$

Then the time-evolution operator $\mathcal{U}_1(t)$ can be expressed as

$$\begin{aligned}\mathcal{U}_1(t) = &\exp\left(-i\frac{d_0}{2\hbar}t\hat{T}_{0l}\right)\\ &\times\exp\left[\left(-\frac{|\tilde{X}_4|^2}{2+\frac{1}{3}x_{0l}|\tilde{X}_4|^2}+i\mathrm{Im}\tilde{X}_2\right)\hat{I}_{0l}\right]\\ &\times\exp\left[\left(\frac{L_1-iL_2}{F}\tilde{X}_4^*{B_l^*}^2\right)\hat{a}_{l+}\right]\exp(\tilde{X}_4\hat{a}_{l-})\\ &\times\exp\left(-i\frac{d_0}{2\hbar}t\hat{T}_{0r}\right)\\ &\times\exp\left[\left(-\frac{|\tilde{Y}_4|^2}{2+\frac{1}{3}x_{0r}|\tilde{Y}_4|^2}+i\mathrm{Im}\tilde{Y}_2\right)\hat{I}_{0r}\right]\\ &\times\exp\left[\left(\frac{L_1^{'}-iL_2^{'}}{F^{'}}\tilde{Y}_4^*{B_r^*}^2\right)\hat{a}_{r+}\right]\exp(\tilde{Y}_4\hat{a}_{r-}).\end{aligned}\tag{38}$$

2.2.2 Time-evolution operator $\mathcal{U}_2$(t)

By putting equation (38) into equation (18) and using the rotating wave approximation,

$$\begin{aligned}\mathcal{H}_2^{'} &= \mathcal{U}_{1r}^{-1}\mathcal{U}_{1l}^{-1}\mathcal{H}_2\mathcal{U}_{1l}\mathcal{U}_{1r}\\ &= \hat{I}_{0r}(\beta_1\hat{I}_{0l}+\beta_2\hat{a}_{l+}+\beta_3\hat{a}_{l-})\\ &\quad+\hat{I}_{0l}(\beta_4\hat{I}_{0r}+\beta_5\hat{a}_{r+}+\beta_6\hat{a}_{r-})\\ &\quad+\beta_7\hat{a}_{l+}\hat{a}_{r-}+\beta_8\hat{a}_{r+}\hat{a}_{l-},\end{aligned}\tag{39}$$

where $\beta_k(t)$ $(k = 1, 2, ...8)$ are

$$\begin{aligned}\beta_1 &= \lambda B_l^*B_r^*\{\exp[2(x_{0l}\tilde{X}_2-x_{0r}\tilde{Y}_2)]\\ &\quad\times\tilde{X}_4\tilde{Y}_3(1+\tilde{Y}_3\tilde{Y}_4{B_r^*}^2x_{0r})\}\\ \beta_2 &= \lambda B_l^*B_r^*\{\exp[2(x_{0r}\tilde{Y}_2-x_{0l}\tilde{X}_2)]\\ &\quad\times\tilde{X}_3\tilde{Y}_4(1+\tilde{X}_3\tilde{X}_4{B_l^*}^2x_{0l})\}\\ \beta_3 &= -\lambda B_l^*B_r\{\exp[2(x_{0l}\tilde{X}_2-x_{0r}\tilde{Y}_2)]\tilde{X}_4\tilde{Y}_3^2{B_r^*}^2x_{0r}\\ &\quad+\exp[2(x_{0r}\tilde{Y}_2-x_{0l}\tilde{X}_2)]\tilde{X}_3(1+\tilde{X}_3\tilde{X}_4{B_l^*}^2x_{0l})\}\\ \beta_4 &= \lambda B_l^*B_r\{\exp[2(x_{0l}\tilde{X}_2-x_{0r}\tilde{Y}_2)]\tilde{X}_4(1+\tilde{Y}_3\tilde{Y}_4{B_r^*}^2x_{0r})^2\\ &\quad+\exp[2(x_{0r}\tilde{Y}_2-x_{0l}\tilde{X}_2)]\\ &\quad\times\tilde{X}_3\tilde{Y}_4^2{B_r^*}^2x_{0r}(1+\tilde{X}_3\tilde{X}_4{B_l^*}^2x_{0l})\}\\ \beta_5 &= -\lambda B_lB_r^*\{\exp[2(x_{0l}\tilde{X}_2-x_{0r}\tilde{Y}_2)]\tilde{Y}_3(1+\tilde{Y}_3\tilde{Y}_4{B_r^*}^2x_{0r})\\ &\quad+\exp[2(x_{0r}\tilde{Y}_2-x_{0l}\tilde{X}_2)]\tilde{X}_3^2\tilde{Y}_4{B_l^*}^2x_{0l}\}\\ \beta_6 &= \lambda B_lB_r^*\{\exp[2(x_{0l}\tilde{X}_2-x_{0r}\tilde{Y}_2)]\\ &\quad\times\tilde{X}_4^2\tilde{Y}_3{B_l^*}^2x_{0l}(1+\tilde{Y}_3\tilde{Y}_4{B_r^*}^2x_{0r})\\ &\quad+\exp[2(x_{0r}\tilde{Y}_2-x_{0l}\tilde{X}_2)]\tilde{Y}_4(1+\tilde{X}_3\tilde{X}_4{B_l^*}^2x_{0l})^2\}\\ \beta_7 &= -\lambda B_lB_r\{\exp[2(x_{0l}\tilde{X}_2-x_{0r}\tilde{Y}_2)](1+\tilde{Y}_3\tilde{Y}_4{B_r^*}^2x_{0r})^2\\ &\quad+\exp[2(x_{0r}\tilde{Y}_2-x_{0l}\tilde{X}_2)]\tilde{X}_3^2\tilde{Y}_4^2{B_l^*}^2{B_r^*}^2x_{0l}x_{0r}\}\\ \beta_8 &= -\lambda B_lB_r\{\exp[2(x_{0l}\tilde{X}_2-x_{0r}\tilde{Y}_2)]\tilde{X}_4^2\tilde{Y}_3^2{B_l^*}^2{B_r^*}^2x_{0l}x_{0r}\\ &\quad+\exp[2(x_{0r}\tilde{Y}_2-x_{0l}\tilde{X}_2)](1+\tilde{X}_3\tilde{X}_4{B_l^*}^2x_{0l})^2\}.\end{aligned}\tag{40}$$

$\mathcal{U}_2(t)$ is so much smaller than $\mathcal{U}_1(t)$ that we can obtain $\mathcal{U}_2(t)$ by using the Magnus approximation [25]

$$\mathcal{U}_2(t) = \exp[\Omega(t)],\tag{41}$$

where Ω is an infinite series

$$\Omega(t) = \sum_{m=1}^{\infty}\Omega_m(t),\tag{42}$$

in which Ω_m denotes the integrals of m-fold multiple commutators [26]. The first two terms of the Magnus operator are

$$\Omega_1(t) = -\frac{i}{\hbar}\int_0^t dt_1\hat{H}_2^{'}(t_1),\tag{43}$$

$$\Omega_2(t) = \frac{1}{2\hbar^2}\int_0^t dt_2\int_0^{t_2}dt_1[\hat{H}_2^{'}(t_1),\hat{H}_2^{'}(t_2)].\tag{44}$$

Using equations (10) and (39), the first term is the following expression

$$\begin{aligned}\Omega_1(t) = {} & [\alpha_1(t)+\alpha_4(t)]\hat{I}_{0l}\hat{I}_{0r} + \alpha_2(t)\hat{I}_{0r}\hat{A}_l^+ + \alpha_3(t)\hat{I}_{0r}\hat{A}_l^- \\ & +\alpha_5(t)\hat{I}_{0l}\hat{A}_r^+ + \alpha_6(t)\hat{I}_{0l}\hat{A}_r^- + \alpha_7(t)\hat{A}_l^+\hat{A}_r^- \\ & +\alpha_8(t)\hat{A}_r^+\hat{A}_l^-, \end{aligned} \qquad (45)$$

where the α_k $(k = 1, 2, ...8)$ are

$$\begin{aligned}
\alpha_1(t) &= -\frac{i}{\hbar}\int_0^t \beta_1(t_1)dt_1 \\
\alpha_2(t) &= -\frac{i}{\hbar}\int_0^t \beta_2(t_1)\exp(i\omega_{0l}\hat{I}_{0l}t_1)dt_1 \\
\alpha_3(t) &= -\frac{i}{\hbar}\int_0^t \beta_3(t_1)\exp(-i\omega_{0l}\hat{I}_{0l}t_1)dt_1 \\
\alpha_4(t) &= -\frac{i}{\hbar}\int_0^t \beta_4(t_1)dt_1 \\
\alpha_5(t) &= -\frac{i}{\hbar}\int_0^t \beta_5(t_1)\exp(i\omega_{0r}\hat{I}_{0r}t_1)dt_1 \\
\alpha_6(t) &= -\frac{i}{\hbar}\int_0^t \beta_6(t_1)\exp(-i\omega_{0r}\hat{I}_{0r}t_1)dt_1 \\
\alpha_7(t) &= -\frac{i}{\hbar}\int_0^t \beta_7(t_1)\exp[i(\omega_{0l}\hat{I}_{0l}-\omega_{0r}\hat{I}_{0r})t_1]dt_1 \\
\alpha_8(t) &= -\frac{i}{\hbar}\int_0^t \beta_8(t_1)\exp[i(\omega_{0r}\hat{I}_{0r}-\omega_{0l}\hat{I}_{0l})t_1]dt_1. \qquad (46)
\end{aligned}$$

The total time-dependent evolution operator can be represented by using equations (38) and (43)

$$\begin{aligned}
\mathcal{U}_I(t) &= \mathcal{U}_1(t)\mathcal{U}_2(t) \\
&= \exp\left(-i\frac{d_0}{2\hbar}t\hat{T}_{0l}\right) \\
&\quad\times\exp\left[\left(-\frac{|\tilde{X}_4|^2}{2+\frac{1}{3}x_{0l}|\tilde{X}_4|^2}+i\mathrm{Im}\tilde{X}_2\right)\hat{I}_{0l}\right] \\
&\quad\times\exp\left[\left(\frac{L_1-iL_2}{F}\tilde{X}_4^*B_l^{*2}\right)\hat{a}_{l+}\right]\exp\left(\tilde{X}_4\hat{a}_{l-}\right) \\
&\quad\times\exp\left(-i\frac{d_0}{2\hbar}t\hat{T}_{0r}\right) \\
&\quad\times\exp\left[\left(-\frac{|\tilde{Y}_4|^2}{2+\frac{1}{3}x_{0r}|\tilde{Y}_4|^2}+i\mathrm{Im}\tilde{Y}_2\right)\hat{I}_{0r}\right] \\
&\quad\times\exp\left[\left(\frac{L_1^{'}-iL_2^{'}}{F^{'}}\tilde{Y}_4^*B_r^{*2}\right)\hat{a}_{r+}\right]\exp\left(\tilde{Y}_4\hat{a}_{r-}\right) \\
&\quad\times\exp\left\{\Omega_1(t)\right\}. \qquad (47)
\end{aligned}$$

2.3 Transition probability

The probability of transition from state $|v_{0l}, 0, v_{0r}\rangle$ to state $|v_{fl}, 0, v_{fr}\rangle$ is

$$P_{0f}(t) = |\langle v_{fl}, 0, v_{fr}|\mathcal{U}_I(t)|v_{0l}, 0, v_{0r}\rangle|^2, \qquad (48)$$

$$\begin{aligned}
\mathcal{U}_1(t)|v_l,0,v_r\rangle = {} & \sum_{m_l=0}^{\infty}\tilde{X}_4^{m_l}\frac{\sqrt{[1-x_{0l}(v_l-1)]v_l\cdots[1-x_{0l}(v_l-m_l)](v_l-m_l+1)}}{m_l!} \\
& \times\sum_{n_l=0}^{\infty}(\tilde{X}_4^*)^{n_l}\frac{\sqrt{[1-x_{0l}(v_l-m_l)](v_l-m_l+1)\cdots[1-x_{0l}(v_l-m_l+n_l-1)](v_l-m_l+n_l)}}{n_l!} \\
& \times\left(\frac{L_1-iL_2}{F}B_l^{*2}\right)^{n_l}\exp\left\{-i\omega_{0l}m_l[1-2x_{0l}(v_l-m_l)]t\right\} \\
& \times\exp\left\{\left[-\frac{|\tilde{X}_4|^2}{2+\frac{1}{3}x_{0l}|\tilde{X}_4|^2}+i(\mathrm{Im}\tilde{X}_2+\omega_{0l}n_lt)\right][1-2x_{0l}(v_l-m_l+n_l)]\right\} \\
& \times\sum_{m_r=0}^{\infty}\tilde{Y}_4^{m_r}\frac{\sqrt{[1-x_{0r}(v_r-1)]v_r\cdots[1-x_{0r}(v_r-m_r)](v_r-m_r+1)}}{m_r!} \\
& \times\sum_{n_r=0}^{\infty}(\tilde{Y}_4^*)^{n_r}\frac{\sqrt{[1-x_{0r}(v_r-m_r)](v_r-m_r+1)\cdots[1-x_{0r}(v_r-m_r+n_r-1)](v_r-m_r+n_r)}}{n_r!} \\
& \times\left(\frac{L_1^{'}-iL_2^{'}}{F^{'}}B_r^{*2}\right)^{n_r}\exp\left\{-i\omega_{0r}m_r[1-2x_{0r}(v_r-m_r)]t\right\} \\
& \times\exp\left\{\left[-\frac{|\tilde{Y}_4|^2}{2+\frac{1}{3}x_{0r}|\tilde{Y}_4|^2}+i(\mathrm{Im}\tilde{Y}_2+\omega_{0r}n_rt)][1-2x_{0r}(v_r-m_r+n_r)\right]\right\} \\
& \times\exp\left(-i\frac{d_0}{\hbar}t\right)|v_l-m_l+n_l,0,v_r-m_r+n_r\rangle \qquad (49)
\end{aligned}$$

$$\begin{aligned}\Omega_1(t)|v_{0l},0,v_{0r}\rangle = {} & [\alpha_1(t)+\alpha_4(t)](1-2x_{0l}v_{0l})(1-2x_{0r}v_{0r})|v_{0l},0,v_{0r}\rangle \\ & + \alpha_2(t)(1-2x_{0r}v_{0r})\sqrt{(1-x_{0l}v_{0l})(v_{0l}+1)}|v_{0l}+1,0,v_{0r}\rangle \\ & + \alpha_3(t)(1-2x_{0r}v_{0r})\sqrt{[1-x_{0l}(v_{0l}-1)]v_{0l}}|v_{0l}-1,0,v_{0r}\rangle \\ & + \alpha_5(t)(1-2x_{0l}v_{0l})\sqrt{(1-x_{0r}v_{0r})(v_{0r}+1)}|v_{0l},0,v_{0r}+1\rangle \\ & + \alpha_6(t)(1-2x_{0l}v_{0l})\sqrt{[1-x_{0r}(v_{0r}-1)]v_{0r}}|v_{0l},0,v_{0r}-1\rangle \\ & + \alpha_7(t)\sqrt{(1-x_{0l}v_{0l})(v_{0l}+1)}\sqrt{[1-x_{0r}(v_{0r}-1)]v_{0r}}|v_{0l}+1,0,v_{0r}-1\rangle \\ & + \alpha_8(t)\sqrt{[1-x_{0l}(v_{0l}-1)]v_{0l}}\sqrt{(1-x_{0r}v_{0r})(v_{0r}+1)}|v_{0l}-1,0,v_{0r}+1\rangle. \qquad (50)\end{aligned}$$

During the computation, we found that the values of α_k $(k = 1, 2, ...8)$ were all smaller than $\tilde{X}_j$, $\tilde{Y}_k$ $(j, k = 1, 2, 3, 4)$, and so we expand $e^{\Omega_1(t)}$ and take the first two terms in order to save computational time

$$e^{\Omega_1(t)} = 1 + \Omega_1(t) + \frac{1}{2}[\Omega_1(t)]^2 + \frac{1}{3}[\Omega_1(t)]^3 + \cdots, \qquad (51)$$

and then the explicit Lie-algebra expression for the vibrational transition probability can be obtained by using the following derivation

$$\begin{aligned}\mathcal{U}_I(t)|v_{0l},0,v_{0r}\rangle &= \mathcal{U}_1(t)\mathcal{U}_2(t)|v_{0l},0,v_{0r}\rangle \\ &= \mathcal{U}_1(t)[1+\Omega_1(t)]|v_{0l},0,v_{0r}\rangle. \qquad (52)\end{aligned}$$

Firstly, by applying the relations of equations (10), (25) and the well-known formula

$$\exp(\tilde{X}_i\hat{a}_i) = \sum_{m=0}^{\infty}\frac{\tilde{X}_i^m\hat{a}_i^m}{m!}, \qquad (53)$$

the explicit expression of any state function acted on by the time-evolution operator $\mathcal{U}_1(t)$ can be given by equation (49).

Secondly, the following formula (Eq. (50)) is easily obtained by using equations (25) and (45).

Then putting equations (49), (50) and (52) into equation (48), the explicit Lie-algebra expression of the vibrational transition probability is obtained.

The long-time-averaged transition probability is defined by

$$\overline{P}_{0f} = \lim_{T\to\infty}\frac{1}{T}\int_0^T P_{0f}(t)dt, \qquad (54)$$

while the long-time-averaged absorption energy spectra are obtained using

$$\langle\overline{\varepsilon}(\omega)\rangle = \sum_f \overline{P}_{0f}(\omega)\varepsilon_f, \qquad (55)$$

and the averaged number of photons absorbed by the molecule can be calculated by

$$\langle n(t)\rangle = \sum_f \frac{\varepsilon_f}{\hbar\omega_L}P_{0f}(t). \qquad (56)$$

Table 1. Values of parameters for the DCN molecule in a.u.

bonds	DC	CN
m	3144	11780
ω_0	0.01201	0.00889
x_0	0.00824	0.00509
D	0.3644	0.4366
a	0.7889	1.0325
λ	0.000472	

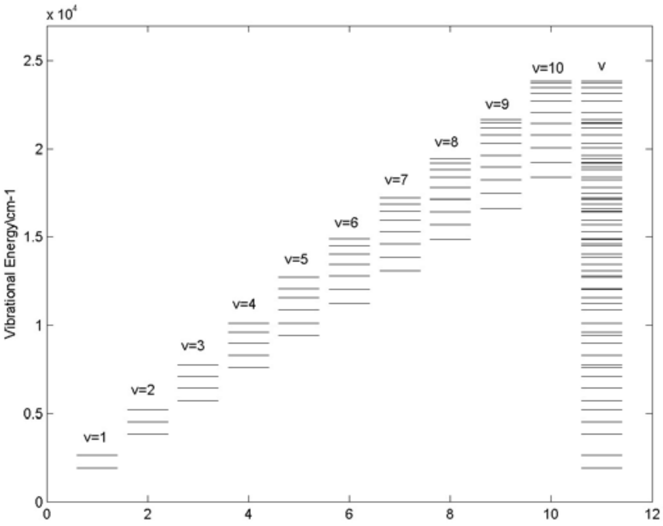

Fig. 1. The stretch-vibration energy diagrams for the first ten excited states of DCN ($v = v_l + v_r$).

3 Example

3.1 The stretch-vibration spectrum of the DCN molecule

The relevant spectral analysis [27] shows that the stretch-vibration of DCN is more important than the bending motion in its molecular vibrations. So only the stretch-vibration spectrum is calculated here. The values of parameters suitable for DCN are given in Table 1 [28].

The first ten stretch-vibration energy levels have been calculated in order to check the Hamiltonian (Eq. (2)) that was used. Figure 1 shows a structure similar to other theoretical results [27]. The theoretical model of the linear

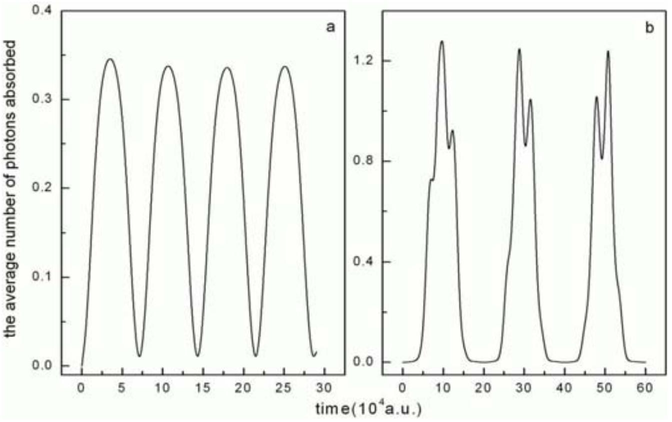

Fig. 2. Time-dependent average number of photons absorbed by DCN with $E_0 = 0.003$ a.u. ($I \approx 0.32$ TW/cm^2). (a) $\omega_L = 0.012$ a.u. (2630 cm^{-1}), the single-photon resonance, (b) $\omega_L = 0.01075$ a.u. (2360 cm^{-1}), the seven-photon resonance.

triatomic molecule introduced here is appropriate, and so it can be used to study the infrared multiphoton vibrational excitation of DCN.

3.2 Resonant excitation probability

The dipole moment of the DCN molecule is given by using equation (5) with experimental values [29]. We assume the molecule to be in the ground state at $t = 0$. Figure 3 gives the long-time-averaged absorption energy spectra plots of the DCN molecule at different laser intensities. Figure 2 gives the averaged number of photons absorbed by the molecule.

Based upon

$$N = (\epsilon_N - \epsilon_0)/\hbar\omega_N, \tag{57}$$

where ω_N is the resonant frequency, $\epsilon_N - \epsilon_0$ is the energy gap between the ground state and the Nth excited state [30], N-photons transition resonant frequencies can be found in the curves of the long-time-averaged vibrational transition probabilities as a function of external field frequency. The frequency of the infrared laser field is adjusted to obtain the highest peaks of the transition probabilities from $\omega_L = 0.007$ a.u. to $\omega_L = 0.013$ a.u.

Figure 3 shows that as the laser intensity increases, both the long-time-averaged absorption energies and the resonant excitation peaks increase. Calculation of the vibrational transition probabilities shows that the highest resonance excitation peak at different laser intensities belongs to different multiphoton resonances, which means an efficient multiphoton resonance can be achieved only under certain laser intensity. This result is consistent with experiment [31].

The average number of photons absorbed by the molecule have been studied at $E_0 = 0.003$ a.u. At $\omega_L = 0.012$ a.u. and $\omega_L = 0.01075$ a.u., the single-photon and the seven-photon resonant excitations are found; the corresponding curves of the time-dependent average number of photons absorbed by the molecule are given in Figure 2. The curve follows a simple Rabi oscillation, and the vibrational period is about 150 optical cycles (about 464 a.u. of one optical cycle) in the single-photon resonance. Within the Rabi cycle, the single-photon curves have cone-shaped peaks and broad bases. However, the average number of photons absorbed of the seven-photon resonance has split peaks and a long periodical behavior, the period being about 430 optical cycles. The multiphoton resonant excitation is a long-time process. This result also agrees with other relevant research [32].

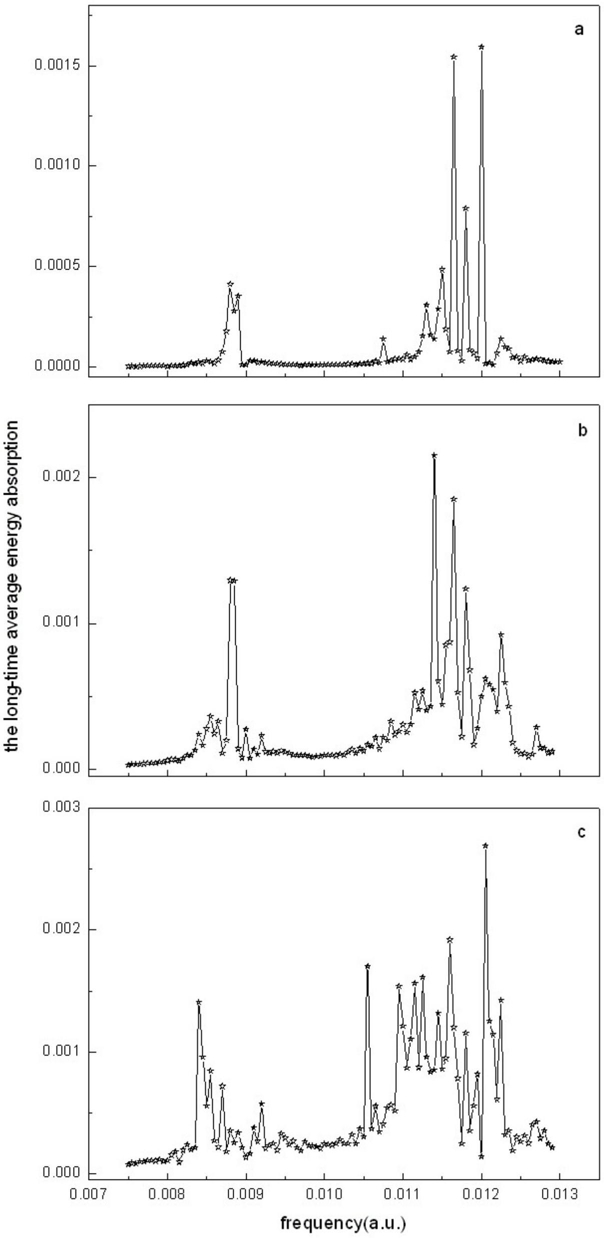

Fig. 3. Long-time-averaged energy absorption spectra of DCN at different laser intensities. (a) $E_0 = 0.003$ a.u. ($I \approx 0.32$ TW/cm^2), (b) $E_0 = 0.009$ a.u. ($I \approx 2.84$ TW/cm^2), (c) $E_0 = 0.015$ a.u. ($I \approx 7.89$ TW/cm^2).

4 Conlusion

In this study, the Lie-algebra approach of studying the infrared multiphoton vibrational excitation of the linear triatomic molecule has been introduced. The explicit Lie-algebra expressions for the time-evolution operator and vibrational transition probabilities make the computation clearer and easier. A simple example is given to explain the method, and the results agree well with experiments and other theoretical studies. Using this method to study the infrared multiphoton excitation of the linear triatomic molecules is effective. Many concrete examples can be tackled as using this method, such as control of the multiphoton vibrational excitation and dissociation in chirped pulses. This is a current work in progress. A much richer information of multiphoton processes is to be expected if bend-mode excitation and rotations are taken into account. These considerations will be addressed in future work.

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