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Multiphoton Selective Excitation and Analytical Control of Small Molecules in Intense Laser Fields: an Algebraic Model

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1. Introduction

The advent of high-power and short-pulse laser technology has led to the study of atomic and molecular multiphoton processes because of its importance in many areas, including photochemistry, fluorescence imaging, and photoionization (Denk et al., 1990; Blackwell et al., 1997; Constant et al., 1996; Paramonov, 2005; Windhorn et al., 2002; Oomens et al., 2004). The quantum control of problem by laser is also an active subject both in the theoretical and experimental area (Tannor & Rice, 1985; Brumer & Shapiro, 1986; Judson & Rabitz, 1992; Levis et al., 2001; Zou et al., 2006). The studies of the quantum control are motivated by fundamental interest in quantum properties of light, and the possible applications in physics, chemistry, and quantum computation, cryptology etc.. The comprehensive reviews could be found in Refs. (Gordon & Rice, 1997; Rice & Zhao, 2000; Brumer & Shapiro, 2003; Kral et al., 2007). One of the considerable attention in this field is multiphoton selective (vibrational) excitation and dissociation, which is important to control the chemical reaction and the quantum states on demand. Various approaches are suggested, for example, the pump-dump laser, optimal control theory, phase control etc.. Some theoretical and computational methods have been developed for improvements in experiment, such as the Coulter transformation method, the Floquet theory method, and dressed molecule picture etc. (Colgan et al., 2001; Chang et al., 1985; Chu & Telnov, 2004; Leforestier & Wyatt, 1985; 1983; Dibble & Shirts, 1991). Much of the work, however, has studied the problem by numerically solving the time-dependent Schrödinger equation (TDSE) as an initial value problem in Hilbert space. The development of generalized Floquet formalisms allows the reduction of the periodical or quasiperiodical time-dependent Schrödinger equation into a set of time-independent coupled equations or Floquet matrix eigenvalue problem. The Floquet method has also been employed to study the problem of molecules in intense laser fields, but it has usually been used to discuss the atomic problems (Leasure et al., 1981; Burke et al., 2000; Colgan et al., 2001; Chu & Telnov, 2004).

In this chapter we present the algebraic approach on a different background. In the algebraic framework we can obtain the explicit expression of the time-evolution operator directly, which avoiding the complex process of solving the time-dependent Schrödinger equation numerically. The time-evolution operator can be expressed a product of a finite number of exponential operators if the operators in Hamiltonian close under commutation. The parameters in time-evolution operator satisfy a set of differential equations. For the past

years the application of time-dependent problems of the dynamical Lie algebraic approach are advanced. Algebraic methods have been extensively used to study problems in nuclear physics, molecular physics and quantum optics etc., for example, the problems of vibrational excited states and potential energy surfaces for small polyatomic molecules have been solved successfully (Iachello, 1981; van Roosmalen et al., 1984; Benjamin et al., 1983; Ding & Zheng, 1999; Zheng & Ding, 2001; 1999; Benjamin et al., 1983; Lu & Kellman, 1995; 1997; Kellman, 1995; Zheng & Ding, 2001). This approach is recently used to study the interacting quantum system, the dynamical symmetries and its breaking in nanophysics, Tavis-Cummings problem and the decoherence in a general three-level system, the dynamical entanglement of vibrations etc.. (Batista & Ortiz, 2004; Kikoin et al., 2004; Vadeiko & Miroshnichenko, 2003; Rau & Zhao, 2005; Zheng, 2006; Hou et al., 2006; Dai et al., 2002; Feng et al., 2007a;b; Liu et al., 2008; Feng et al., 2010). In the present work, both the multiphoton selective excitation and analytical control of small molecules (diatomic and triatomic molecules) in intense laser fields are studied in the algebraic framework. The influences of laser pulse frequencies and shapes on control are discussed.

In sec. 2 we derived the time-evolution operator of the system by using algebraic method for the diatomic and triatomic molecules. The analytical expression of the transition probability is obtained via the time-evolution operator. In sec. 3 we discussed the selective vibrational excitation of OH and OD by shaped and chirped laser pulses and the multiphoton excitation problem. For triatomic molecules, we calculated the selective bond excitation of HCN and DCN molecules in the linear chirped laser pulses with different shapes. The problem of intramolecular vibrational redistribution (IVR) is also discussed in this section. The chapter ends with some concluding remarks in sec. 4.

2. Theoretical background

2.1 The general considerations

We consider the problem of small molecules (diatomic and triatomic molecules) in the laser field, the more ambitious case of including other environment are not pursued at this time. The Hamiltonian of the system considered here can be written as

$$\mathcal{H} = \mathcal{H}_{mol} + \mathcal{V}, \quad (1)$$

where \mathcal{H}_{mol} is the Hamiltonian of an unperturbed molecule, and \mathcal{V} is its interaction with a laser field. As usually, we here consider the interaction between the molecule and the laser field within the electric dipole approximation,

$$\mathcal{V} = -\boldsymbol{\mu} \cdot \mathcal{E}(t), \quad (2)$$

where $\mathcal{E}(t)$ is the external laser field, and $\boldsymbol{\mu}$ is the molecular dipole moment.

It is suitable to consider this problem in *the interaction representation*. The Hamiltonian (1) could be splitted into an \mathcal{H}_0 part and into an additional "interaction" \mathcal{H}' , namely, the Hamiltonian (1) can be rewritten as

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}'(t). \quad (3)$$

In the interaction representation, the system Hamiltonian is written as

$$\mathcal{H}_I(t) = e^{i\mathcal{H}_0 t/\hbar} \mathcal{H}'(t) e^{-i\mathcal{H}_0 t/\hbar}, \quad (4)$$

and the time-evolution operator is given by

$$i\hbar \frac{\partial}{\partial t} \mathcal{U}_I(t) = \mathcal{H}_I(t) \mathcal{U}_I(t), \quad (5)$$

with the initial condition $\mathcal{U}_I(t=0) = 1$. Once we know the evolution operator of the system, we can extract all the information of the system from the evolution operator. Hence, the influences of laser pulse frequencies and shapes can be investigated and take these quantum control on demand. Specially, here we are interested in the transition probability of the system from the initial state $|v_i\rangle$ to the final state $|v_f\rangle$,

$$\mathcal{P}_{v_i, v_f}(t) = \left| \langle v_f | \mathcal{U}_I(t) | v_i \rangle \right|^2. \quad (6)$$

The corresponding long-time average probability is defined as

$$\langle \mathcal{P}_{v_i, v_f} \rangle = \lim_{T \rightarrow \infty} \left\{ \frac{1}{T} \int_0^T \mathcal{P}_{v_i, v_f}(t) dt \right\}. \quad (7)$$

Also, the long-time-averaged absorption energy spectra are obtained using

$$\langle \varepsilon \rangle = \sum_f \langle \mathcal{P}_{v_i, v_f} \rangle \varepsilon_{v_i, v_f}, \quad (8)$$

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

$$\langle n(t) \rangle = \sum_f \frac{\varepsilon_{v_i, v_f}}{\hbar \omega_L} \mathcal{P}_{v_i, v_f}(t), \quad (9)$$

where ε_{v_i, v_f} is the energy difference between the states $|v_f\rangle$ and $|v_i\rangle$, ω_L is the frequency of the external laser field. Here we neglect the obvious labels of the various aspects of the laser field in Eqs. (6) and (7). In the following sections we would obtain the obvious expressions of Eqs. (6) ~ (9) in the theoretical framework.

2.2 Theoretical framework for diatomic molecules

We consider here the one-dimensional vibrational motion of a diatomic molecule. For completeness, we brief review the well-known results for the $U(2)$ dynamical symmetry of diatomic molecules. The $U(2)$ algebra is successfully applied to describe vibrations of diatomic molecule, and $U(2)$ possesses two dynamical symmetry chains (Levine, 1983; 1982; Kellman, 1985; Cooper, 1997; 1998):

$$\begin{aligned} U(2) &\supset U(1), \\ U(2) &\supset O(2). \end{aligned} \quad (10)$$

The generators of $U(2)$ could be realized in terms of two boson creation and annihilation operators:

$$t^\dagger t, s^\dagger s, t^\dagger s, s^\dagger t. \quad (11)$$

The new four generators could be constructed using these four bilinear products, familiar with Schwinger,

$$\begin{aligned} \hat{Q} &= (1/2N)^{1/2} (t^\dagger s + s^\dagger t), \\ \hat{P} &= (1/2N)^{1/2} (t^\dagger s - s^\dagger t), \\ \hat{I}_0 &= N^{-1} (s^\dagger s - t^\dagger t), \\ \hat{E}_0 &= N^{-1} (t^\dagger t + s^\dagger s), \end{aligned} \quad (12)$$

which N is the eigenvalues of the number operator $\hat{N}(= t^\dagger t + s^\dagger s)$.

The Hamiltonian of a non-rotating Morse oscillator could be written as

$$\mathcal{H}_{mol} = \hbar\omega_0\left(\frac{\hat{p}^2}{2} + \frac{\hat{Q}^2}{2}\right), \quad (13)$$

where ω_0 is the frequency of the anharmonic oscillator, \hat{P} and \hat{Q} are the bilinear products constructed by the generators of $U(2)$ algebra in Eq. (12) (Levine, 1983; 1982).

An equivalent expression of Morse oscillator Hamiltonian Eq. (13) is

$$\mathcal{H}_{mol} = \hbar\omega_0\left(\hat{A}^\dagger \hat{A} + \frac{\hat{I}_0}{2}\right), \quad (14)$$

where the creation (A^\dagger) and annihilation (A) operators are defined as (Levine, 1983),

$$\hat{A}^\dagger = \frac{1}{\sqrt{2}}(\hat{Q} - i\hat{P}), \quad \hat{A} = \frac{1}{\sqrt{2}}(\hat{Q} + i\hat{P}), \quad (15)$$

which obey the commutation relations

$$[A, A^\dagger] = I_0, \quad [I_0, A] = -2\chi_0 A, \quad [I_0, A^\dagger] = 2\chi_0 A^\dagger. \quad (16)$$

It should be noted that I_0 is an operator and tends to the identity operator in the harmonic limits, and $\chi_0 \rightarrow 0$ in this limits. The anharmonic correction is given in order of $\chi_0 = \frac{1}{N}$.

In our numerical calculation we would take the replace of N by $N + 1$. According to Cooper *et al* (Cooper, 1997; 1998) this modification can lead to quite accurate fits to the vibrational energy levels of diatomic molecules, potential function including the zero-point energy and achieves the known value of dissociation energy.

The molecular dipole moment $\mu(x)$ could be expanded in a series at the equilibrium, and we keep the first order in our present work, as in Refs.(Hay & Dunning, 1976; Walker & Preston, 1977; Tung & Yuan, 1987; Chelkowski, 1995; Dai et al., 2002), i.e.

$$\mu(x) = \mu_0 x. \quad (17)$$

The interaction between the molecule and the laser field Eq. (2), by using Eq. (15), can be rewritten as

$$\mathcal{V} = -\frac{d(t)}{2} (A^\dagger + A), \quad (18)$$

where the time-dependent parameter $d(t)$ is

$$d(t) = \mu_0 \cdot \mathcal{E}(t) \frac{1}{\alpha} \sqrt{\frac{\hbar\omega_0}{D}}. \quad (19)$$

Here we choose $\mathcal{H}_0 = \mathcal{H}_{mol}$ and $\mathcal{H}' = \mathcal{V}$, the system Hamiltonian, in the interaction representation from Eq.(4), reads

$$\begin{aligned} \mathcal{H}_I(t) &= e^{i\mathcal{H}_0 t/\hbar} \mathcal{V} e^{-i\mathcal{H}_0 t/\hbar} \\ &= e^{i\omega_0 \chi_0 t \hat{A}^\dagger \hat{A}} e^{i\omega_0 t I_0/2} d(A^\dagger + A) e^{-i\omega_0 t I_0/2} e^{-i\omega_0 \chi_0 t A^\dagger A} \\ &= d e^{i\omega_0 \chi_0 t} e^{i\omega_0 I_0 t} A^\dagger d e^{i\omega_0 \chi_0 t} e^{-i\omega_0 I_0 t} A \\ &\equiv \gamma_+ A^\dagger + \gamma_- A. \end{aligned} \quad (20)$$

Here we have taken the operator I_0 as an identity approximately in the exponential term since the anharmonic correction is of order χ_0 . It is often of the order of 1% or less for realistic molecules (Levine, 1983).

Since the generators A^\dagger , A and I_0 form a closed Lie algebra (see Eq. (16)), the evolution operator can be written as (Wei & Norman, 1964; Alhassid & Levine, 1978)

$$\begin{aligned} U_I &= e^{-\frac{i}{\hbar}\mu_0\hat{I}_0}e^{-\frac{i}{\hbar}\mu_+\hat{A}^\dagger}e^{-\frac{i}{\hbar}\mu_-\hat{A}} \\ &\equiv \prod_r U_r(t), \text{ and } U_r(t) = e^{-\frac{i}{\hbar}\mu_r(t)X_r}, \end{aligned} \tag{21}$$

where the coefficients $\mu_r(t)$ ($r = 0, +, -$) are known as the Lagrange parameters (Alhassid & Levine, 1978). The operators in Eq. (21) are $X_0 \equiv I_0$, $X_+ \equiv A^\dagger$ and $X_- \equiv A$. The equations of $U_r(t)$, from the Eqs.(5) and (20), are as

$$i\hbar \frac{\partial U_r(t)}{\partial t} = \frac{\partial \mu_r(t)}{\partial t} X_r U_r(t), \tag{22}$$

or the equations of the Lagrange parameters $\mu_r(t)$, by using the Baker-Hausdorff expansion, are given by

$$\begin{aligned} \dot{\mu}_0 &= -\frac{i}{\hbar}\mu_+\gamma_-e^{\frac{i}{\hbar}2\chi_0\mu_0}, \\ \dot{\mu}_+ &= \gamma_+e^{-\frac{i}{\hbar}2\chi_0\mu_0} - \frac{\chi_0}{\hbar^2}\mu_+^2\gamma_-e^{\frac{i}{\hbar}2\chi_0\mu_0}, \\ \dot{\mu}_- &= \gamma_-e^{\frac{i}{\hbar}2\chi_0\mu_0}, \end{aligned} \tag{23}$$

with the initial conditions

$$\mu_r(t=0) = 0, \quad (r = 0, \pm). \tag{24}$$

The probability from the initial state $|v_i\rangle$ to the final state $|v_f\rangle$, from Eq. (6), is

$$\mathcal{P}_{v_i,v_f}(t) = |\langle v_f|U_I(t)|v_i\rangle|^2 = |\lambda(t)|^2\delta_{v_f,v_i-m+n}, \tag{25}$$

where

$$\begin{aligned} \lambda(t) &= \exp\left\{-\frac{i}{\hbar}\mu_0[1-2\chi_0(v_i-m+n)]\right\} \\ &\times \sum_{n=0}^{\infty} \frac{1}{n!} \left(-\frac{i}{\hbar}\mu_+\right)^n \sqrt{\prod_{n'=0}^n [1-\chi_0(v_i-m'+n')](v_i-m'+n')} \\ &\times \sum_{m=0}^{\infty} \frac{1}{m!} \left(-\frac{i}{\hbar}\mu_-\right)^m \sqrt{\prod_{m'=0}^m [1-\chi_0(v_i-m')(v_i-m'+1)]}. \end{aligned} \tag{26}$$

The analytic expression of vibrational transition probability is obtained and we can tackle many concrete examples using this expression.

2.3 Theoretical framework for triatomic molecules

For the case of triatomic molecule, its dynamical symmetry group is

$$U_1(2) \otimes U_2(2). \tag{27}$$

For stretching vibrations in a triatomic molecule, by using Eq. (15), the algebraic Hamiltonian reads

$$\mathcal{H}_m = \hbar\omega_{01}(\hat{A}_1^\dagger \hat{A}_1 + \frac{\hat{I}_{01}}{2}) + \hbar\omega_{02}(\hat{A}_2^\dagger \hat{A}_2 + \frac{\hat{I}_{02}}{2}) - \lambda(\hat{A}_1^\dagger \hat{A}_2 + \hat{A}_2^\dagger \hat{A}_1), \quad (28)$$

where ω_{01} and ω_{02} are the angular frequencies of the bond 1 and bond 2 of triatomic molecule, respectively. λ is the coupling coefficient, we include here the kinetic and potential linear couple terms.

The interaction between the triatomic molecule and the laser field can be expressed, similar to the diatomic molecule, as:

$$\mathcal{V} = d_1(\hat{A}_1^\dagger + \hat{A}_1) + d_2(\hat{A}_2^\dagger + \hat{A}_2), \quad (29)$$

where

$$d_1 = -\frac{1}{2\alpha_1} \sqrt{\frac{\hbar\omega_{01}}{D_1}} \boldsymbol{\mu}_1 \cdot \mathcal{E}(t), \quad \text{and} \quad d_2 = -\frac{1}{2\alpha_2} \sqrt{\frac{\hbar\omega_{02}}{D_2}} \boldsymbol{\mu}_2 \cdot \mathcal{E}(t). \quad (30)$$

Here the linear dipole moment form of $\boldsymbol{\mu}_1$ and $\boldsymbol{\mu}_2$ (Umeda et al., 1994) is employed for the molecule.

In the interaction picture, the Hamiltonian of the system for the triatomic molecule is

$$\begin{aligned} \mathcal{H}_I(t) &= e^{i\mathcal{H}_0 t/\hbar} \mathcal{H}' e^{-i\mathcal{H}_0 t/\hbar} \\ &= d_1(\kappa_{1+} \hat{A}_1^\dagger + \kappa_{1-} \hat{A}_1) + d_2(\kappa_{2+} \hat{A}_2^\dagger + \kappa_{2-} \hat{A}_2) \\ &\quad - \lambda(\kappa_{1+} \kappa_{2-} A_1^\dagger A_2 + \kappa_{1-} \kappa_{2+} A_2^\dagger A_1), \end{aligned} \quad (31)$$

where

$$\kappa_{j+} = e^{i\omega_{0j}(x_{0j} + I_{0j})t}, \quad \text{and} \quad \kappa_{j-} = e^{i\omega_{0j}(x_{0j} - I_{0j})t}, \quad (j = 1, 2). \quad (32)$$

In the calculation we have chosen

$$\begin{aligned} \mathcal{H}_0 &= \hbar\omega_{01}(\hat{A}_1^\dagger \hat{A}_1 + \frac{\hat{I}_{01}}{2}) + \hbar\omega_{02}(\hat{A}_2^\dagger \hat{A}_2 + \frac{\hat{I}_{02}}{2}), \\ \mathcal{H}' &= -\lambda(\hat{A}_1^\dagger \hat{A}_2 + \hat{A}_2^\dagger \hat{A}_1) + d_1(\hat{A}_1^\dagger + \hat{A}_1) + d_2(\hat{A}_2^\dagger + \hat{A}_2). \end{aligned} \quad (33)$$

We partition the Hamiltonian (31), for the simplicity of algebraic structure and calculation, into two parts

$$\mathcal{H}_I(t) = \mathcal{H}_I^{(0)} + \mathcal{H}_I^{(1)}(t), \quad (34)$$

where we let

$$\mathcal{H}_I^{(0)} = d_1(\kappa_{1+} \hat{A}_1^\dagger + \kappa_{1-} \hat{A}_1) + d_2(\kappa_{2+} \hat{A}_2^\dagger + \kappa_{2-} \hat{A}_2), \quad (35)$$

and

$$\mathcal{H}_I^{(1)}(t) = -\lambda(\kappa_{1+} \kappa_{2-} A_1^\dagger A_2 + \kappa_{1-} \kappa_{2+} A_2^\dagger A_1). \quad (36)$$

The evolution equation of the evolution operator $\mathcal{U}_I^{(0)}(t)$, corresponding to Hamiltonian (35), is as follows (Landau & Lifshitz, 1977)

$$i\hbar \frac{\partial}{\partial t} \mathcal{U}_I^{(0)}(t) = \mathcal{H}_I^{(0)} \mathcal{U}_I^{(0)}(t), \quad (37)$$

with the initial condition $\mathcal{U}_I^{(0)}(0) = 1$.

If $\mathcal{U}_I^{(0)}(t)$ is known, the evolution operator $\mathcal{U}_I(t)$ describing the whole system (corresponding total Hamiltonian (31)) will be given by (Landau & Lifshitz, 1977)

$$\mathcal{U}_I(t) = \mathcal{U}_I^{(0)}(t) \cdot \mathcal{U}_I^{(1)}(t), \tag{38}$$

where $\mathcal{U}_I^{(1)}(t)$ satisfies the evolution equation

$$i\hbar \frac{\partial}{\partial t} \mathcal{U}_I^{(1)}(t) = \mathcal{H}_I^{(1)'}(t) \mathcal{U}_I^{(1)}(t), \tag{39}$$

with the initial condition $\mathcal{U}_I^{(1)}(0) = 1$. The Hamiltonian $\mathcal{H}_I^{(1)'}(t)$ is given by

$$\mathcal{H}_I^{(1)'}(t) = \mathcal{U}_I^{(0)\dagger}(t) \mathcal{H}_I^{(1)}(t) \mathcal{U}_I^{(0)}(t). \tag{40}$$

It is easily to show that the operator in Hamiltonian (35) close under the commutation (16). The time-evolution operator $\mathcal{U}_I^{(0)}$ can be written as (Wei & Norman, 1964; Alhassid & Levine, 1978)

$$\mathcal{U}_I^{(0)} = e^{-\frac{i}{\hbar} \mu_{01} I_{01}} e^{-\frac{i}{\hbar} \mu_{1+} A_1^\dagger} e^{-\frac{i}{\hbar} \mu_{1-} A_1} e^{-\frac{i}{\hbar} \mu_{02} I_{02}} e^{-\frac{i}{\hbar} \mu_{2+} A_2^\dagger} e^{-\frac{i}{\hbar} \mu_{2-} A_2}. \tag{41}$$

Following the similar procedures to those carried out in preceding section, the Lagrange parameters are satisfied

$$\begin{aligned} \dot{\mu}_{01} &= -\frac{i}{\hbar} d_1 \kappa_{1-} \mu_{1+} e^{\frac{i}{\hbar} 2\chi_{01} \mu_{01}}, \\ \dot{\mu}_{1+} &= d_1 \kappa_{1+} e^{-\frac{i}{\hbar} 2\chi_{01} \mu_{01}} - \frac{\chi_{01}}{\hbar^2} d_1 \kappa_{1-} \mu_{1+}^2 e^{\frac{i}{\hbar} 2\chi_{01} \mu_{01}}, \\ \dot{\mu}_{1-} &= d_1 \kappa_{1-} e^{\frac{i}{\hbar} 2\chi_{01} \mu_{01}}, \\ \dot{\mu}_{02} &= -\frac{i}{\hbar} d_2 \kappa_{2-} \mu_{2+} e^{\frac{i}{\hbar} 2\chi_{02} \mu_{02}}, \\ \dot{\mu}_{2+} &= d_2 \kappa_{2+} e^{-\frac{i}{\hbar} 2\chi_{02} \mu_{02}} - \frac{\chi_{02}}{\hbar^2} d_2 \kappa_{2-} \mu_{2+}^2 e^{\frac{i}{\hbar} 2\chi_{02} \mu_{02}}, \\ \dot{\mu}_{2-} &= d_2 \kappa_{2-} e^{\frac{i}{\hbar} 2\chi_{02} \mu_{02}} \end{aligned} \tag{42}$$

The initial conditions are $\mu_r(t = 0) = 0$, ($r = 01, 1\pm, 02, 2\pm$).

The Hamiltonian $\mathcal{H}_I^{(1)'}(t)$ can be then expressed as

$$\begin{aligned} \mathcal{H}_I^{(1)'}(t) &= (\mathcal{U}_I^{(0)})^{-1} \mathcal{H}_I^{(1)}(t) \mathcal{U}_I^{(0)} \\ &= \beta_0 \hat{I}_{01} \hat{I}_{02} + \beta_1 \hat{I}_{02} \hat{A}_1^\dagger + \beta_2 \hat{I}_{02} \hat{A}_1 + \beta_3 \hat{I}_{01} \hat{A}_2^\dagger + \beta_4 \hat{I}_{01} \hat{A}_2 \\ &\quad + \beta_5 \hat{A}_1^\dagger \hat{A}_2 + \beta_6 \hat{A}_1 \hat{A}_2^\dagger + \beta_7 \hat{A}_1^\dagger \hat{A}_2^\dagger + \beta_8 \hat{A}_1 \hat{A}_2, \end{aligned} \tag{43}$$

where

$$\begin{aligned} \beta_0 &= \frac{\eta_1}{\hbar^2} \mu_{1-} \mu_{2+} (1 - \frac{\chi_{02}}{\hbar^2} \mu_{2+} \mu_{2-}) + \frac{\eta_2}{\hbar^2} \mu_{1+} \mu_{2-} (1 - \frac{\chi_{01}}{\hbar^2} \mu_{1+} \mu_{1-}), \\ \beta_1 &= -\frac{i}{\hbar} \eta_1 \mu_{2+} (1 - \frac{\chi_{02}}{\hbar^2} \mu_{2+} \mu_{2-}) + \frac{i}{\hbar^3} \eta_2 \chi_{01} \mu_{1+}^2 \mu_{2-}, \\ \beta_2 &= -\frac{i}{\hbar^3} \eta_1 \chi_{01} \mu_{1-}^2 \mu_{2+} (1 - \frac{\chi_{02}}{\hbar^2} \mu_{2+} \mu_{2-}) + \frac{i}{\hbar} \eta_2 \mu_{2-} (1 - \frac{\chi_{01}}{\hbar^2} \mu_{1+} \mu_{1-})^2, \end{aligned}$$

$$\begin{aligned}
\beta_3 &= \frac{i}{\hbar^3} \eta_1 \chi_{02} \mu_{1-} \mu_{2+}^2 - \frac{i}{\hbar} \eta_2 \mu_{1+} \left(1 - \frac{\chi_{01}}{\hbar^2} \mu_{1+} \mu_{1-}\right), \\
\beta_4 &= \frac{i}{\hbar} \eta_1 \mu_{1-} \left(1 - \frac{\chi_{02}}{\hbar^2} \mu_{2+} \mu_{2-}\right)^2 - \frac{i}{\hbar^3} \eta_2 \chi_{02} \mu_{1+} \mu_{2-}^2 \left(1 - \frac{\chi_{01}}{\hbar^2} \mu_{1+} \mu_{1-}\right), \\
\beta_5 &= \eta_1 \left(1 - \frac{\chi_{02}}{\hbar^2} \mu_{2+} \mu_{2-}\right)^2 + \frac{\eta_2}{\hbar^4} \chi_{01} \chi_{02} \mu_{1+}^2 \mu_{2-}^2, \\
\beta_6 &= \frac{\eta_1}{\hbar^4} \chi_{01} \chi_{02} \mu_{1-}^2 \mu_{2+}^2 + \eta_2 \left(1 - \frac{\chi_{01}}{\hbar^2} \mu_{1+} \mu_{1-}\right)^2, \\
\beta_7 &= \eta_1 \frac{\chi_{02}}{\hbar^2} \mu_{2+}^2 + \eta_2 \frac{\chi_{01}}{\hbar^2} \mu_{1+}^2, \\
\beta_8 &= \eta_1 \frac{\chi_{01}}{\hbar^2} \mu_{1-}^2 \left(1 - \frac{\chi_{02}}{\hbar^2} \mu_{2+} \mu_{2-}\right)^2 + \eta_2 \frac{\chi_{02}}{\hbar^2} \mu_{2-}^2 \left(1 - \frac{\chi_{01}}{\hbar^2} \mu_{1+} \mu_{1-}\right)^2.
\end{aligned} \tag{44}$$

The coefficients η_1 and η_2 , in Eq. (44), are defined as

$$\eta_1 = -\lambda \kappa_{1+} \kappa_{2-} e^{\frac{i}{\hbar} 2(\chi_{02} \mu_{02} - \chi_{01} \mu_{01})}, \quad \text{and} \quad \eta_2 = -\lambda \kappa_{1-} \kappa_{2+} e^{\frac{i}{\hbar} 2(\chi_{01} \mu_{01} - \chi_{02} \mu_{02})}. \tag{45}$$

The time-evolution operator $\mathcal{U}_I^{(1)}(t)$, corresponding to Hamiltonian Eq. (43), can be obtained, by using the Magnus approximation,

$$\mathcal{U}_I^{(1)}(t) = \exp \left[\sum_{m=1}^{\infty} \Gamma_m(t) \right], \tag{46}$$

where Γ_m denotes the integrals of m -fold multiple commutators (Klarsfeld & Oteo, 1989).

The total time-evolution operator $\mathcal{U}_I(t)$ of the whole system can be calculated by using Eq. (38). The transition probability from state $|v_{1i}, v_{2i}\rangle$ to state $|v_{1f}, v_{2f}\rangle$ is

$$P_{if}(t) = |\langle v_{1f}, v_{2f} | \mathcal{U}_I(t) | v_{1i}, v_{2i} \rangle|^2, \tag{47}$$

The explicit expression of the vibrational transition probability can be found in Ref. (Feng et al., 2007a). The average energy in bonds can also be obtained by using the time-evolution operator.

3. Numerical results and discussions

3.1 Diatomic molecules

In this section, we study the multiphoton transition of the diatomic molecules. For the comparison with previous study, we use the local OH and OD bond of the HOD molecule in our numerical model. These models are extensively studied in previous study (Korolkov & Paramonov, 1997; Stranges et al., 2002; Amstrup & Henriksen, 1992; Elghobashi et al., 2003). All parameters, in atomic units, are taken from Refs. (Korolkov & Paramonov, 1997; Stranges et al., 2002; Amstrup & Henriksen, 1992; Elghobashi et al., 2003), namely, $\omega_0=0.01664$ a.u., $\chi_0=0.02323$ a.u., $D=0.1614$ a.u., $\alpha=1.156$ a.u. for OH and $\omega_0=0.0122$ a.u., $\chi_0=0.01645$ a.u., $D=0.1636$ a.u., $\alpha=1.142$ a.u. for OD, respectively.

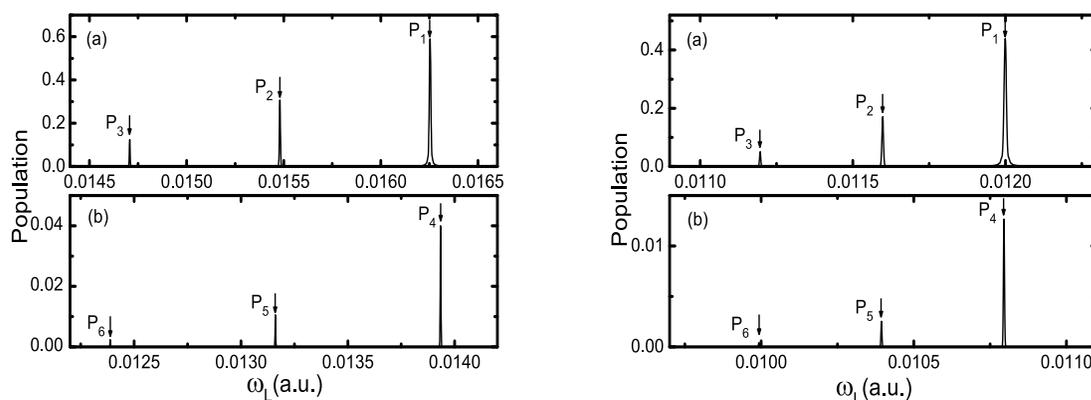


Fig. 1. The long-time average populations of OH (left) and OD (right) as a function of laser frequency. (a) The transitions from the ground state to the first, second and third excited states. (b) The transitions from the ground state to the fourth, fifth and sixth excited states. P_i ($i = 1, 2, 3, 4, 5, 6$) denotes the transition $|v = 0\rangle \rightarrow |v = i\rangle$.

3.1.1 The multiphoton vibrational transition

We first consider the simple case: *the multiphoton vibrational transition*. In this case we choose the laser field as follows

$$\mathcal{E}(t) = \mathcal{E}_0 \sin(\omega_L t). \quad (48)$$

The initial state of the molecules are set on the ground state. The frequency of the infrared laser field is adjusted to the resonant frequency ω_r .

Multiphoton transitions are said to be in resonance if the energy of one or several quanta of laser photons is close to the transition energy from the initial to an intermediate excited state or to that between the intermediate. The transition is called multiphoton resonance transition, if the following condition is satisfied (Jakubetz et al., 1990; Lin & Fujimura, 1984)

$$\omega_r \cong \omega_n = \frac{\varepsilon_f - \varepsilon_0}{n\hbar}, \quad (49)$$

where $\varepsilon_f - \varepsilon_0$ is the energy gap between the ground state and the f th excited state.

Fig. 1 gives the long-time average probabilities of OH and OD from the ground state to the different excited states using Eq. (7). We can obtain the resonance frequency ω_r corresponding to the peaks of the transition probabilities. The orders of multiphoton transitions could be determined using *the formal intensity law* (Lin & Fujimura, 1984),

$$\ln P_n = n \ln I + C, \quad (50)$$

where I is the intensity of the laser field, P_n is the transition probability for the n -photon process, and C is a constant.

If no saturation occurs, one can determine the order of the multiphoton transition from the slope of Eq. (50). The order of the n -photon transition could be known from the slopes of the log-log plots of the transition probabilities as a function of laser intensity I , which correspond to the transition probabilities in Fig. 1, respectively. The detail results are summarized in Tab. 1.

As shown in Tab. 1, the transitions from the ground state to the first, second, third, fourth, fifth and sixth excited states belong to one-, two-, three-, four-, five-, six-photon transitions. However, the deviation between the transition frequency ω_r and the frequency

		P_1	P_2	P_3	P_4	P_5	P_6
OH	ω_r	0.016253	0.015480	0.014707	0.013934	0.013161	0.012388
	n	1.002	2.004	3.005	4.008	5.003	5.984
	ω_n	0.016221	0.015835	0.015453	0.015063	0.014698	0.014359
	$ \omega_n - \omega_r $	0.000032	0.000355	0.000746	0.001129	0.001537	0.001971
OD	ω_r	0.011999	0.011598	0.011197	0.010796	0.010395	0.009994
	n	1.003	2.014	3.011	3.973	5.002	6.020
	ω_n	0.011963	0.011716	0.011556	0.011474	0.011192	0.010959
	$ \omega_n - \omega_r $	0.000036	0.000118	0.000359	0.000678	0.000797	0.000965

*All parameters are atomic units except n .

Table 1. Multiphoton vibrational transitions of OH and OD

ω_n is becoming larger. This deviation denotes the non-resonant transition appears when the vibrational state goes up.

From Fig. 1, we can also see that the resonance frequencies ω_r are little shifting when the system reaches the different final states. Namely, the resonant transition frequency decreases with the final energy level going up. And because of the higher dissociation energy in deuterated case the transition probabilities of OD are all smaller than those of OH at the same laser intensity. The value of the interval between the transition peaks in left panel of Fig. 1 (for OH) is about $0.000773a.u.$, and the value in right of Fig. 1 (for OD) is about $0.000401a.u.$. However, the interval of the transition peaks of OH is larger than that of OD. We attribute it to the difference of anharmonicity parameter of OH and OD, i.e., the anharmonic parameters of OH is larger than that of OD. The ratio of the anharmonicity parameters of OH and OD is 1.4 which is comparable to the ratio of 0.000773 to 0.000401 (≈ 1.9). Furthermore, we can see the deviations ($\omega_r - \omega_n$) of OH are also larger than those of OD in Tab. 1. These results show the anharmonicity of molecular vibrations has an important influence on the resonant transition frequency.

3.1.2 The multiphoton selective vibrational transition of OH

Sugimori *et al.* investigated the multiphoton absorption of OH using numerical simulation (Sugimori *et al.*, 2005). For the comparison, we here take the same molecular parameters as in Ref. (Sugimori *et al.*, 2005). We make compare all the calculations for the 1-photon, 2-photon, 3-photon and 4-photon absorption in Ref. (Sugimori *et al.*, 2005) with our analytical results. In this comparison, the laser field is employed with the same with Ref. (Sugimori *et al.*, 2005)

$$\begin{aligned}\mathcal{E}(t) &= \mathcal{E}_0 g(t) \sin(\omega_L t), \\ g(t) &= \exp[-(t - T_0)^2 / \sigma^2],\end{aligned}\quad (51)$$

where $T_0 = 100ps$, $\sigma = 30ps$ and the laser frequency ω_L is calculated using Eq.(49). In Tab. 2 a comparison is made between numerical results in Ref. (Sugimori *et al.*, 2005) and our analytical results. A good agreement is found, indicating that our algebraic model is excellent approximation.

Since there exist a little deviation of the resonant transition frequency for the different transition(see details in Tab. 1), we then take the calculated frequency as the laser frequency and study the multiphoton selective vibrational transition of OH. We find the selectivity of the vibrational transition will decrease as the value of the laser frequency has a little change (about $0.000001a.u. \approx 6.6 \times 10^3 MHz$). The little changes on the laser frequency may cause

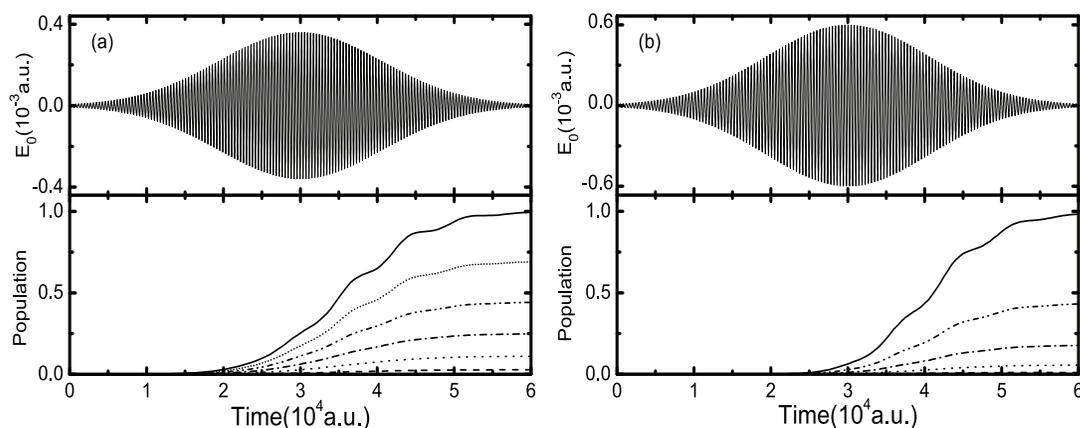


Fig. 2. Time dependence of the laser field and population: (a)1-photon transition between $|0\rangle$ and $|1\rangle$, (b)2-photon transition between $|0\rangle$ and $|2\rangle$, by changing the laser intensity with the fixed pulse duration at the non-chirped laser pulse. The laser parameters are given in sec.3.1.3.

	OH^*		OH	
	I_{max}	Ratio(%)	I_{max}	Ratio(%)
1-photon	4.74×10^6	100.00	4.68×10^6	99.99
2-photon	5.68×10^8	99.98	5.57×10^8	99.49
3-photon	5.58×10^9	99.88	5.42×10^9	99.23
4-photon	2.36×10^{10}	94.51	2.27×10^{10}	95.31

* Numerical solution from Ref. (Sugimori et al., 2005).

Table 2. The comparison of maximum laser intensity $I_{max}(W/cm^2)$ and the ratio of the target state population

the less selectivity and the stronger laser intensity. This denotes the selectivity of vibrational transition is sensitive to the laser frequency.

3.1.3 Control of vibrational excitation of OH and OD molecules

We have obtained the resonant transition frequencies of OH and OD from the ground state to some excited states in previous section. In this section, we use these resonant frequencies to study the selective vibrational transition of OH and OD. The laser field is

$$\begin{aligned} \mathcal{E}(t) &= \mathcal{E}_0 f(t) \cos \Phi(t), \\ f(t) &= \exp[-(t - \tau/2)^2 / (\tau/4)^2], \end{aligned} \tag{52}$$

where the Gaussian pulse envelop $f(t)$ is employed since it is extensively employed in experiment (Zou et al., 2006). The frequency of the field is chosen constant for non-chirped laser pulses(ω_r), or time-dependent for chirped ones, namely, the phase of the laser field pulse is chosen as

$$\Phi(t) = \begin{cases} \omega_r t, & \text{(non-chirped pulse)} \\ \Phi_0 + \int_0^t [\Delta\omega_1 + \Delta\omega_2 e^{-(t'/\tau)^2}] dt' & \text{(chirped pulse)} \end{cases}$$

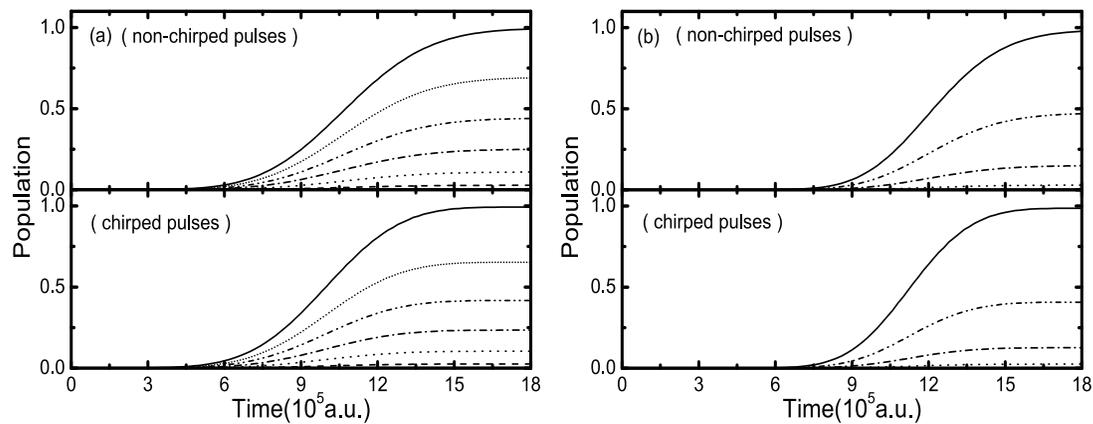


Fig. 3. Comparison of populations between the chirped by changing the laser intensity with the fixed pulse duration for OH molecule. (a) 1-photon transition between $|0\rangle$ and $|1\rangle$, the value of the laser field strength from the lowest curve to the highest one in the non-chirped pulse is from $\mathcal{E}_0 = 0.000001a.u.$ to $\mathcal{E}_0 = 0.000011a.u.$ ($\approx 4.25 \times 10^6 W/cm^2$), and in the chirped pulse is from $\mathcal{E}_0 = 0.0000017a.u.$ to $\mathcal{E}_0 = 0.0000102a.u.$ ($\approx 3.59 \times 10^6 W/cm^2$). (b) 2-photon transition between $|0\rangle$ and $|2\rangle$, the value of the laser field strength from the lowest curve to the highest one in the non-chirped pulse is from $\mathcal{E}_0 = 0.00004a.u.$ to $\mathcal{E}_0 = 0.00012a.u.$ ($\approx 5.05 \times 10^8 W/cm^2$), and in the chirped pulse is from $\mathcal{E}_0 = 0.00003a.u.$ to $\mathcal{E}_0 = 0.00011a.u.$ ($\approx 4.29 \times 10^8 W/cm^2$).

That is, the two types of the laser pulse are used to study the selective vibrational transition: *the non-chirped laser pulse* and *the chirped laser pulse*.

The selective excitations of 1-photon and 2-photon of OH are studied by the non-chirped laser pulse. The pulsewidth is set to be $\tau = 60000a.u.$ ($\approx 1.4ps$) as in Refs. (Just et al., 1992; Korolkov et al., 1996). We adjust the laser intensity to get maximum populations from $\mathcal{E}_0 = 0.000055a.u.$ to $\mathcal{E}_0 = 0.00033a.u.$ ($\approx 0.4 \times 10^{10} W/cm^2$) for 1-photon, and from $\mathcal{E}_0 = 0.00013a.u.$ to $\mathcal{E}_0 = 0.00065a.u.$ ($\approx 0.15 \times 10^{11} W/cm^2$) for 2-photon. In Fig. 2 we plot the time dependence of the laser field and population with different intensity. From Fig. 2 we see that the oscillations appear in the population with the laser intensity increases under the condition of the laser pulse duration is relatively small. To avoid the oscillations, we adjust the laser pulse duration and we find the oscillations disappear completely at the pulsewidth $\tau = 18 \times 10^5 a.u.$ ($\approx 40ps$). This value is consentient to the pulsewidth in Ref. (Sugimori et al., 2005). Then we study the changes of the transition probabilities when the non-chirped laser pulse is turned to the chirped one. The transitions from the ground state to the first and the

	OH			OD		
	$\Delta\omega_1(a.u.)$	$\Delta\omega_2(a.u.)$	$I_{max}(W/cm^2)$	$\Delta\omega_1(a.u.)$	$\Delta\omega_2(a.u.)$	$I_{max}(W/cm^2)$
1-photon	0.016253	0.0000042	3.59×10^6	0.011999	0.0000040	4.80×10^6
2-photon	0.015480	0.0000040	4.29×10^8	0.011598	0.0000033	5.61×10^8
3-photon	0.014707	0.0000038	4.25×10^9	0.011197	0.0000025	5.05×10^9
4-photon	0.013934	0.0000036	1.76×10^{10}	0.010796	0.0000018	1.91×10^{10}

Table 3. The optimal laser parameters of multiphoton selective vibrational excitation of OH and OD

second excited states at the different laser intensity are given in Fig. 3. The laser intensity begin to increase from a relative small value till the populations get the maximum values at the end of the non-chirped or chirped pulse (see Fig. 3 caption). For the chirped pulse, we take the frequency $\Delta\omega_1$ as the resonant frequency, namely, we let $\Delta\omega_1 = \omega_r$. The chirped term $\Delta\omega_2$ is adjusted to get a good selectivity at a certain laser intensity. From Fig. 3, we can see that the populations begin to increase and get the maximum earlier by the chirped pulse than the non-chirped one. It shows the chirped pulse gives a better control of vibrational excitation of molecules. Finally, we give the selective excitation of 1-photon, 2-photon, 3-photon and 4-photon of OH and OD. We can obtain complete selective vibrational excitation by a Gaussian shaped and chirped laser pulse. The optimal parameters of the laser pulse are given in Tab. 3. We find the values of the chirped term of OH are larger than those of OD which could be also relevant to the anharmonicity of molecular vibrations. In the same laser duration, the laser frequency of OH decrease faster than that of OD, but the laser intensity of OD is higher than that of OH.

From the above discussion, we see that the optimal laser frequency must decrease as a function of time in order to achieve a good selectivity of excitation. Furthermore, the chirping of OH is faster than that of OD.

3.2 Triatomic molecules

In this section we study the control of triatomic molecules. As for triatomic molecules, there exist richer information of selective laser excitation, for example, there is the phenomenon of intramolecular vibrational redistribution (IVR). The IVR can interfere with selective excitation. Here we investigate the direct resonant excitation of the intramolecular bond for linear triatomic molecule HCN and DCN.

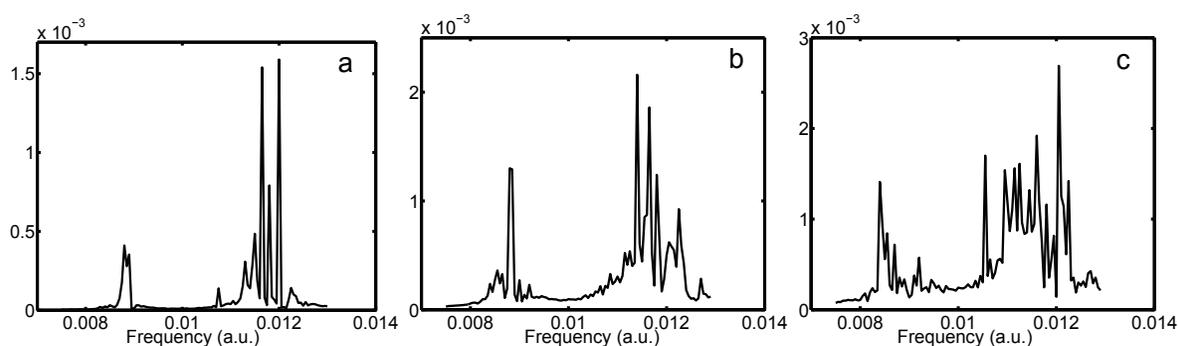


Fig. 4. Long-time-averaged energy absorption spectra of DCN at different laser intensities. (a) $E_0=0.003$ a.u. ($I\approx 0.32$ TW/cm²), (b) $E_0=0.009$ a.u. ($I\approx 2.84$ TW/cm²), (c) $E_0=0.015$ a.u. ($I\approx 7.89$ TW/cm²).

The Hamiltonian (28) is used to calculate the stretch vibrational levels of HCN and DCN molecules successfully, and it can reproduce the real situations of the molecules (Feng & Ding, 2007). The molecular parameters of HCN and DCN are given in Table 1 of Ref. (Feng & Ding, 2007).

We investigate the effects of linearly chirped pulses with different shapes on the molecules. The linear chirped pulses is successfully used to study vibrational excitation and dissociation of the molecules (Liu et al., 1995; 1999; Lin et al., 1998). Furthermore, a linearly chirped pulse can be readily be prepared in the laboratory (Witte et al., 2003; Balling et al., 1994). The laser

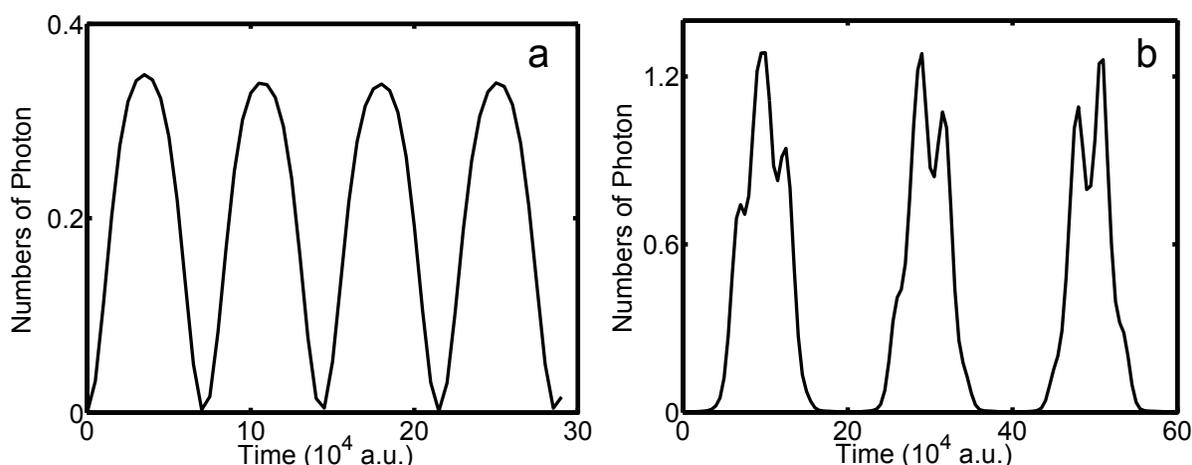


Fig. 5. Time-dependent average number of photons absorbed by DCN with $E_0=0.003\text{a.u.}$ ($I\approx 0.32\text{TW/cm}^2$). (a) $\omega_L=0.012\text{a.u.}(2630\text{cm}^{-1})$, the single-photon resonance, (b) $\omega_L=0.01075\text{a.u.}(2360\text{cm}^{-1})$, the seven-photon resonance.

field is as follows

$$\begin{aligned} E(t) &= E_0 f(t) \cos \Phi(t), \\ \Phi(t) &= \Omega_0 t \left(1 - \frac{\alpha_c t}{2\tau}\right). \end{aligned} \quad (53)$$

In our numerical calculation, Ω_0 is taken as the anharmonic frequency of the selected bond, namely, we let $\Omega_0 = \omega_{01}$ or ω_{02} . The instantaneous frequency is taken as $\Omega_{ins}(t) = \Omega_0 [1 - \alpha_c (\frac{t}{\tau})]$, the amount of chirping is then determined by the parameters α_c . The target excitation state is chosen $v = 10$ which determines the value of the parameters α_c . Researches as in Ref. (Gong et al., 2005; Dion et al., 1999) show the ionization threshold is estimated to be at 10^{14}W/cm^2 , the laser intensities chosen in this investigation are all far below this value and other parameters are similar to Ref. (Brezina & Liu, 2004) and given in figure captions. We here consider three kinds of laser shapes: *rectangular*, *Gaussian* and *Sech-shaped*.

3.2.1 Resonant excitation probability

The dipole moment of DCN molecule is given by using Eq. (17) with experimental values (Hyde & Hornig, 1952). We assume the molecule to be in the ground state at $t = 0$. Fig. 4 gives the long-time-averaged absorption energy spectra of DCN molecule at different laser intensities. Fig. 5 gives the averaged number of photons absorbed by the molecule.

Based upon Eq. (49), 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 $\omega_L = 0.007\text{a.u.}$ to $\omega_L = 0.013\text{a.u.}$

Fig. 4 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 (Borsella et al., 1983).

The average number of photons absorbed by the molecule have been studied at $E_0=0.003\text{a.u.}$. At $\omega_L=0.012\text{a.u.}$ and $\omega_L=0.01075\text{a.u.}$, the single-photon and the seven-photon resonant

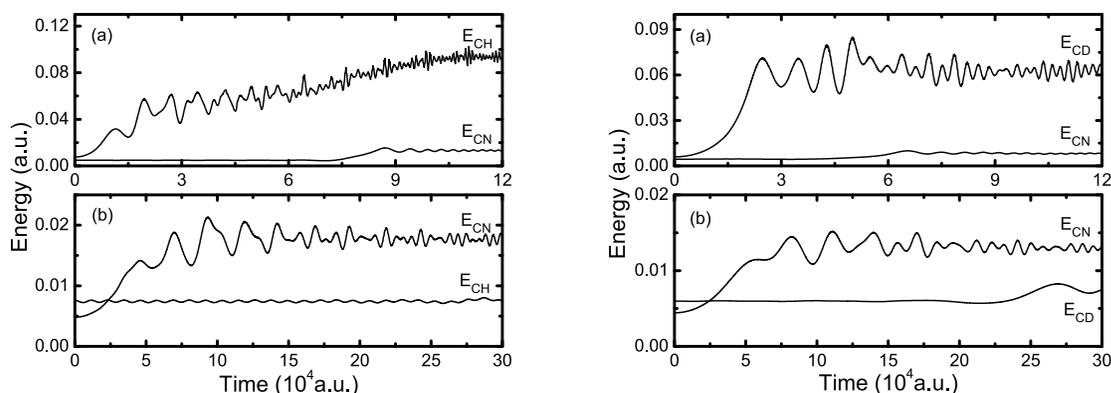


Fig. 6. The selective excitations of C-H bond and C-N bond for HCN (left column) and C-D bond and C-N bond for DCN (right column) to the $v = 10$ level by a resonance linearly chirped pulse with a rectangular shape. (a) Time-dependence of energy in C-H bond (E_{CH}) (left) or C-D bond (E_{CD}) (right) and in C-N bond (E_{CN}). The pulse parameters are $I = 0.9 \times 10^{12} \text{ W/cm}^2$; $\tau \approx 2.8 \text{ ps}$. $\alpha_c = 0.38$ for HCN and $\alpha_c = 0.26$ for DCN. (b) Time-dependence of energy in C-H bond (E_{CH}) (left) or C-D bond (E_{CD}) (right) and in C-N bond (E_{CN}). The pulse parameters are $I = 0.5 \times 10^{13} \text{ W/cm}^2$; $\tau \approx 7.2 \text{ ps}$. $\alpha_c = 0.13$ for HCN and $\alpha_c = 0.11$ for DCN.

excitations are found; the corresponding curves of the time-dependent average number of photons absorbed by the molecule are given in Fig. 5. 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 (Walker & Preston, 1977).

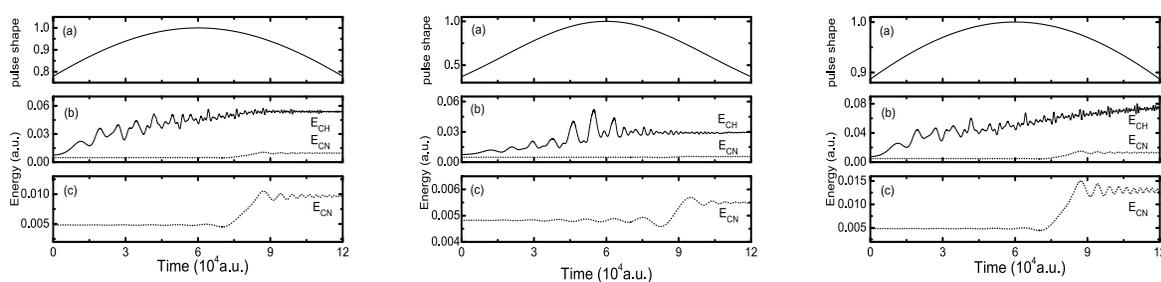


Fig. 7. The selective excitation of C-H bond of HCN to the $v = 10$ level by a resonance linearly chirped pulse with a Gaussian shape. (a) Time dependence of Gaussian pulse shape $f(t)$. (b) Time-dependence of energy in C-H bond (E_{CH}) and in C-N bond (E_{CN}). (c) The enlargement of C-N bond (E_{CN}) energy in panel (b). The pulse parameters are $I = 0.9 \times 10^{12} \text{ W/cm}^2$; $\alpha_c = 0.38$; $\tau \approx 2.8 \text{ ps}$. The left column is for $f(t) = \exp[-(t - \tau/2)^2 / \tau^2]$, the central column is for $f(t) = \exp[-(t - \tau/2)^2 / (\tau/2)^2]$, and the right column is for $f(t) = \text{sech}[(t - \tau/2) / \tau]$.

3.2.2 The selective bond excitation via the rectangular laser shape

The selective vibrational excitation of C-H (C-D) and C-N bonds in HCN and DCN molecules can be achieved by using the rectangular laser shape as shown in Fig. 6. The selective target bond picks up a higher energy but the energy of the other bond has a relatively smaller values at the end of the pulse. The selective bond excitation in molecule HCN is plotted in the left column of Fig. 6. In the top-left panel of Fig. 6, we can see the energy in C-N bond does not oscillate near the initial energy but it has a pronounced increase after the energy in C-H bond grows. There are two possibilities about this phenomenon. One is due to the intramolecular vibrational redistribution (IVR) as energy leaks from the highly excited C-H bond; the other is due to an accidental resonance with the pulse frequency which is near the C-N frequency (ω_{02}) at the end of the laser pulse. From the bottom-left panel of Fig. 6, we see the selective vibrational excitation of the C-N bond needs longer pulse duration and more higher laser intensity. The C-N bond is much heavier and stronger than the C-H bond, so the C-H bond may break firstly before the C-N bond is excited to the higher excitation level or dissociation and the selectivity of the C-N bond decreases. The results are consonant with the other study (Brezina & Liu, 2004).

The selective vibrational excitation of DCN is given in the right column of Fig. 6 and the similar case is observed. However, from top-right panel of Fig. 6, we can see that the energy in C-D bond does not increase always but begin to oscillate at some value, and the energy in C-N bond begin to increase earlier than that of HCN. In bottom-right panel of Fig. 6 when the C-N bond is excited, the C-D bond picks up energy at the end of the pulse. Compared to the left column of Fig. 6, energies in the selective C-D bond or the selective C-N bond are smaller than those of HCN. These results represent IVR (intramolecular vibrational redistribution) in DCN molecule is faster and the selectivity of DCN is much less than that of HCN which consist with the reference (Chelkowski & Bandrauk, 1991).

3.2.3 C-H bond excitation in HCN molecule by a Gaussian and Sech-shaped laser pulses

In this subsection we show the influences of laser pulse on the energy oscillations in a bond of HCN molecule. Fig. 7 (the left and center columns) shows that the oscillations of energy in C-H bond decreased at the end of the Gaussian shaped laser pulse and the pulsewidth of the Gaussian shape have a deep influence on oscillations of energy. But the energy in C-H bond also decreases in the Gaussian shaped laser pulse and the value is smaller than in the rectangular laser shape. In contrast, using the Sech-shaped laser pulses, the value of energy becomes large but still smaller than in the rectangular laser shape as shown in the right column of Fig. 7.

From the figures, we find the values of energy in the bond excitation is decided by the pulse area, and the oscillations of energy is related to the pulse shape. In order to get small oscillation and high energy, we take a Super-Gaussian shape laser pulse and give the C-H and C-N bond selective excitation in Fig. 8. As we expected, the energy in the C-H bond has a larger values and smaller oscillations compared to the other shaped cases. The C-N bond excitation has also a better selectivity than the rectangular case at the same laser intensity.

4. Conclusion

We have studied selective vibrational excitation for small molecules in an algebraic model. It is achieved state-selective excitation of diatomic molecules and bond selective excitation of triatomic molecules successfully. The results are in good agreement with the other researches. In this investigation, we find the optimal laser frequency must decrease as a function of time

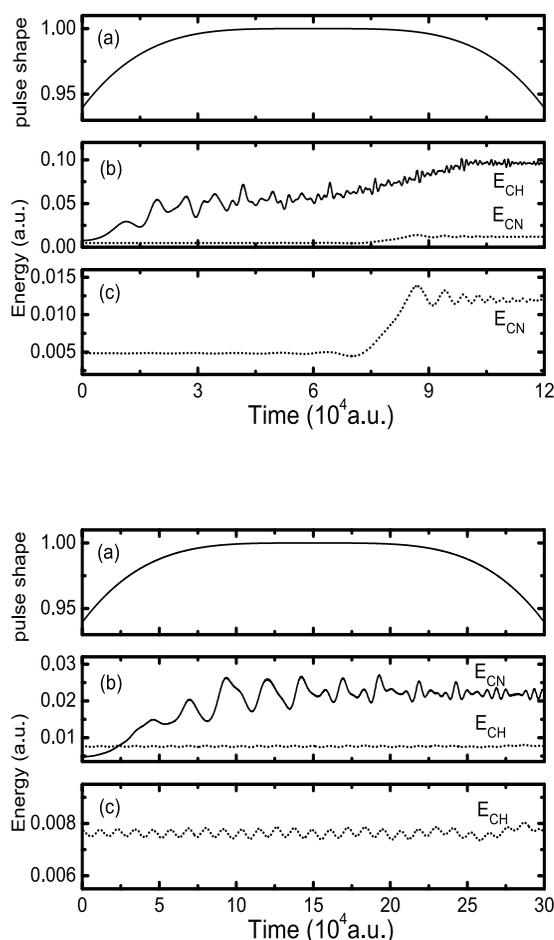


Fig. 8. The selective excitation of C-H bond of HCN to the $v = 10$ level by a resonance linearly chirped pulse with a Super-Gaussian shape. (a) Time dependence of Super-Gaussian pulse shape: $f(t) = \exp[-(t - \tau/2)^4/\tau^4]$. (b) Time-dependence of energy in C-H bond ($-E_{CH}$) and in C-N bond ($\cdots E_{CN}$). (c) The enlargement of C-N bond ($\cdots E_{CN}$) energy in panel (b). The pulse parameters for left column are $I = 0.9 \times 10^{12} \text{ W/cm}^2$; $\alpha_c = 0.38$; $\tau \approx 2.8 \text{ ps}$. The pulse parameters for right column are $I = 0.5 \times 10^{13} \text{ W/cm}^2$; $\alpha_c = 0.13$; $\tau \approx 7.2 \text{ ps}$.

and the transition probability is very sensitive to the laser frequency to the control state excitation of a diatomic molecule. For the triatomic molecule, although the intramolecular vibrational redistribution makes selective excitation difficult, selective excitation to a specific vibrational bond can be achieved by using the appropriate chirped and shaped pulse. To get a good selectivity of bond excitation, the pulse area and the pulse shape are of the same importance. The pulse area decides the value of energy in bonds and the proper laser shape decreases the oscillations of energy. In our cases, a Super-Gaussian shape laser can be used to obtain a better selectivity of bond excitation. Furthermore, the bending motions and rotations can be taken into account and this method can also be extended to study the tetra-atomic molecular case.

More fundamental extensions are possible. The dynamical entanglement of realistic small molecules in the external laser fields can be investigated based on the algebraic models. The quantum control of the von Neumann entropy, and geometric phase of realistic molecules could be a natural investigation in the future.

5. Acknowledgments

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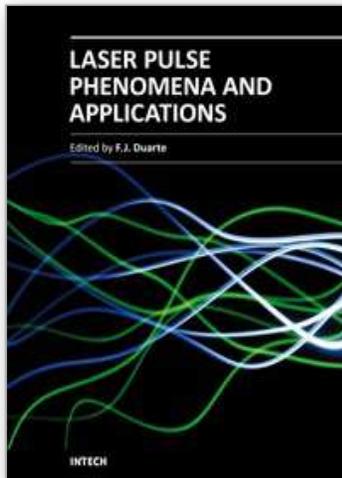
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