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Effects of the laser field on dynamical entanglement in triatomic molecules

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Abstract. Algebraic approach to dynamical entanglement of triatomic molecules in laser fields are proposed. The effects of laser fields on dynamical entanglement are discussed. The researches show that the changes of two stretch vibrational entanglement along with the laser frequency are corresponding to vibrational transitions from different initial states to final states. The sustained entanglement will be implemented by using a two-step laser excitation.

1. Introduction

It have been suggested that molecular vibrational states can be applied to quantum computing for more than ten years, theoretical and experimental researches have attracted much attention [1–11], along with advancement in laser technology. The quantum logic gate operation and arbitrary superposition states in molecular system have been achieved using the quantum coherent control theory (QCCT) and the optimal control theory (OCT) [6, 7]. These progresses promote studying entanglement dynamics in molecular system [12–17]. The entanglement of molecular vibrations emerge initial-state dependence and dynamical connection characteristic relating to intramolecular energy. It is well known that entanglement is the key element in quantum computing, the crux of using molecular vibrational modes for quantum computing is the generation and control of entanglement between the vibrational states. Since the quantum logic gate operation based on molecular vibrational or rotational states can be implemented using the suited laser pulse scheme, it is necessary to explore the influence of laser field on the entanglement dynamics in molecular system. The dynamical Lie algebraic approach has been successfully used to study time-dependent problems [18], the advantage lies in avoiding the complex process of solving the time-dependent Schrödinger equation. The time-evolution operator can be expressed a product of a finite number of exponential operators if the operators in Hamiltonian close under commutation. In this paper, the dynamical Lie algebraic approaches are expanded to study effects of laser field on entanglement in linear triatomic molecules. The long-time-averaged probability and concurrence for different initial states are calculated and compared.

This paper is structured as follow. In Sec.2, the algebraic molecular model in laser fields is reviewed briefly, and the concurrence of the two vibrational states in a linear triatomic molecule is given using the algebraic method. In Sec.3, the influence of the laser field on the dynamical entanglement is discussed. The correlations between vibrational excitation and vibrational entanglement in the laser field are proposed. Finally, conclusions and some remarks are given in Sec.4.

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2. Theoretical Methods

2.1 Reviews to the algebraic molecular model in the laser field

The algebraic Hamiltonian for a linear triatomic molecule in a laser field can be represented as follows [18, 19]

$$
\begin{aligned}
\hat{H} &= \hat{H}_{mol} + \hat{H}_{int} \\
&= \hbar\omega_{01}(\hat{A}_1^\dagger \hat{A}_1 + \frac{\hat{I}_{01}}{2}) + \hbar\omega_{02}(\hat{A}_2^\dagger \hat{A}_2 + \frac{\hat{I}_{02}}{2}) - \lambda(\hat{A}_1^\dagger \hat{A}_2 + \hat{A}_2^\dagger \hat{A}_1), \\
&\quad + d_1(\hat{A}_1^\dagger + \hat{A}_1) + d_2(\hat{A}_2^\dagger + \hat{A}_2)
\end{aligned} \quad (1)
$$

where ω_{01} and ω_{02} are the angular frequencies of the triatomic molecule corresponding to the bond 1 and bond 2. λ is the coupling coefficient which depend on the experimental values of realistic molecular spectra. d_1 and d_2 are the linear dipole moment forms [20] of the molecules

$$
d_i = -\frac{1}{2\alpha_i}\sqrt{\frac{\hbar\omega_{0i}}{D_i}}\mu_i \cdot \varepsilon(t), \quad (2)
$$

in which μ_i (i= 1,2) is the first derivative of the dipole moment, $\varepsilon(t)$ presents the laser field.

The quadratic operators $\hat{A}_i^\dagger, \hat{A}_i, \hat{I}_{0i}$ act on the state $|N_i, V_i\rangle$ [20],

$$
\begin{aligned}
\hat{A}_i^\dagger |N_i, v_i\rangle &= \sqrt{(1 - x_{0i} v_i)(v_i + 1)}\,|N_i, v_i + 1\rangle \\
\hat{A}_i |N_i, v_i\rangle &= \sqrt{[1 - x_{0i}(v_i - 1)]v_i}\,|N_i, v_i - 1\rangle, \\
\hat{I}_{0i} |N_i, v_i\rangle &= 1 - 2x_{0i} v_i |N_i, v_i\rangle,
\end{aligned} \quad (3)
$$

where $x_{0i} = 1/N_i$ is the anharmonic correction [21, 22].

2.2 The derivation of the reduced density

Since the Hamiltonian of the system (eq.(1)) is time-dependent, it is suitable to consider this problem in the intercation representation, the system Hamiltonian can be written as

$$
\begin{aligned}
\hat{H}_I(t) &= e^{i\hat{H}_0 t/\hbar}\hat{V}e^{-i\hat{H}_0 t/\hbar} \\
&= \sum_{i=1,2} d_i(\gamma_{i+}\hat{A}_i^\dagger + \gamma_{i-}\hat{A}_i) - \lambda(\gamma_{1+}\gamma_{2-}\hat{A}_1^\dagger\hat{A}_2 + \gamma_{1-}\gamma_{2+}\hat{A}_2^\dagger\hat{A}_1),
\end{aligned} \quad (4)
$$

in which

$$
\begin{aligned}
\hat{H}_0 &= \hbar\omega_{01}(\hat{A}_1^\dagger\hat{A}_1 + \frac{\hat{I}_{01}}{2}) + \hbar\omega_{02}(\hat{A}_2^\dagger\hat{A}_2 + \frac{\hat{I}_{02}}{2}), \\
v &= -\lambda(\hat{A}_1^\dagger\hat{A}_2 + \hat{A}_2^\dagger\hat{A}_1) + d_1(\hat{A}_1^\dagger + \hat{A}_1) + d_2(\hat{A}_2^\dagger + \hat{A}_2), \\
\gamma_{i+} &= e^{i\omega_{0i}(x_{0i}+I_{0i})t}, \gamma_{i-} = e^{-i\omega_{0i}(x_{0i}-I_{0i})t}, (i = 1, 2).
\end{aligned} \quad (5)
$$

The whole Hamiltonian are partitioned into two parts for the simplicity of algebraic structure,

$$\hat{H}_I = \hat{H}_I^{(0)} + \hat{H}_I^{(1)},$$
$$\hat{H}_I^{(0)}(t) = d_1(\gamma_1 + \hat{A}_1^\dagger + \gamma_1 - \hat{A}_1) + d_2(\gamma_2 + \hat{A}_2^\dagger + \gamma_2 - \hat{A}_2), \quad (6)$$
$$\hat{H}_I^{(1)} = -\lambda(\gamma_1 + \gamma_2 - A_1^\dagger A_2 + \gamma_1 - \gamma_2 - A_2^\dagger A_1),$$

so the the time-evolution operator can also be split into two parts

$$\hat{U}_I(t) = \hat{U}_I^{(0)}(t) \cdot \hat{U}_I^{(1)}(t) \quad (7)$$

According to the Lie algebra Wei-Norman approach [23, 24], the first term evolution operator can be written as

$$\hat{U}_I^{(0)}(t) = \prod_{rj} \hat{U}_{rj}(t), \text{ and } \hat{U}_{rj}(t) = e^{-\frac{i}{\hbar}\mu_{rj}(t)X_{rj}}, \quad (8)$$

where the coefficients $\mu_{rj}(t)$ ($r = 0,+,-$; $j = 1,2$) are known as the Lagrange parameters which can be solved by

$$i\hbar \frac{\partial}{\partial t}\hat{U}_I(t) = \hat{H}_I(t)\hat{U}_I(t), \quad (9)$$

with the initial condition $\hat{U}_I = (t = 0) = 1$. The operators $X_{0j} \equiv I_{0j}, X_{+j} \equiv A_j^\dagger$ and $X_{-j} \equiv A_j$. The second term evolution operator can be obtained by the Magnus approximation

$$\hat{U}_I^{(1)}(t) = \exp[\sum_{m=1}^{\infty}\Gamma_m(t)], \quad (10)$$

where Γ_m denotes the integrals of m-fold multiple commutators [18, 25].

Once the time evolution operator is known, the time-dependent wave function can be obtained

$$|\psi(t)\rangle = \hat{U}_I(t)|\psi(0)\rangle, \quad (11)$$

The probability of vibrational excitation from state $|V_{01}, V_{02}\rangle$ to state $|V_{f1}, V_{f2}\rangle$ is

$$P_{0f}(t) = |\langle v_{f1}, v_{f2}|\hat{U}_I(t)|v_{01}, v_{02}\rangle|^2, \quad (12)$$

and the long-time-averaged transition probability is defined by

$$\bar{P}_{0f} = \lim_{T\to\infty}\frac{1}{T}\int_0^T P_{0f}(t)dt. \quad (13)$$

The reduced density matrices are well-known as

$$\rho_1(t) = Tr_2\rho(t) = Tr_2\{|\psi(t)\rangle\langle\psi(t)|\}, \quad (14)$$

where Tr_2 denotes the partial trace over the second subsystem. Putting eq.(11) into eq.(14), the reduced density matrices of the system can be obtained.

The concurrence for a general pure bipartite state is defined by [26]

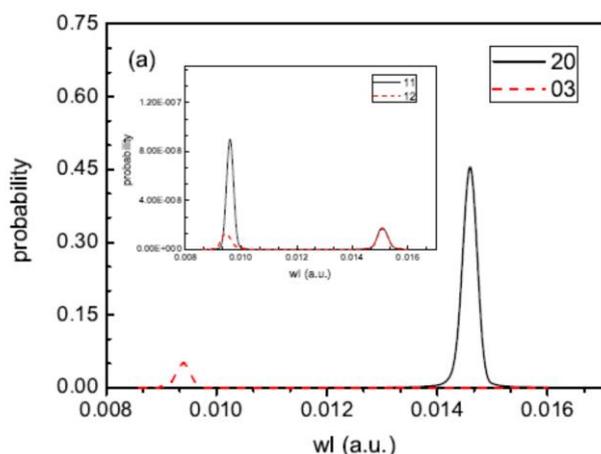$$C(t) = \sqrt{2(1 - Tr_1\rho_1(t)^2)}, \quad (15)$$

Figure 1: The long-time-averaged probability as a function of laser frequency

and the mean values of the concurrence over long time T can be given as follows

$$\bar{C} = \frac{1}{T}\int_0^T C(t)dt, \quad (16)$$

then the dynamical entanglement for the two stretch vibrational states can be solved.

3. Results and discussion

The triatomic molecule HCN as a concrete example is studied. The stretch vibrational energies have been successfully calculated which can reflect the real vibrational situation, and the parameters of HCN molecule are given in our previous work [19]. The laser field is chosen to be $E_0\sin(\omega_L t)$. The long-time-averaged probability and concurrence for different initial states are calculated, here we only show the situation of the initial state $|\psi(0)\rangle = |v_{01}, v_{02}\rangle = 0.2\rangle|$, see in **Figures 1** and **2**, since the essential features of the calculations are similar for those states.

The **Figure 1** give the long-time-averaged probabilities from initial state $|0,2\rangle$ to final states $|1,1\rangle$, $|2,0\rangle$, $|0,3\rangle$ and $|1,2\rangle$, respectively. The strength of the laser field is set as E_0=0.009au. The higher vibrational excitation probabilities are not given because the values are extremely small. The highest peak of the transitional probabilities appears when the laser frequency is at ω_L = 0.0146au, which represents the transition from $|0,2\rangle$ to $|2,0\rangle$. **Figure 2** give the mean values of the concurrence over long time T for initial state $|0,2\rangle$. The value of the laser frequency is also 0.0146au when the concurrence arrives at the highest peak, which indicates the entanglement between states $|0,2\rangle$ and $|2,0\rangle$ is strongest at the present laser frequency. From **Figures 1** and **2**, we can also see the second peak value denotes the transition from $|0,2\rangle$ to $|0,3\rangle$, the value of the corresponding laser frequency is also same both in vibrational excitation and entanglement. When the initial state is changed, the value of the laser frequency is different; however, the corresponding relation between the vibration excitation and entanglement is still invariant. According to the property, the long-time preservation of the entanglement can be achieved by means of generating the superposition states between the two stretch vibration modes. We can obtain the needed initial state $|0,2\rangle$ using the first chirped laser pulse, then adding the second appropriate chirped laser pulse which frequency is need to set as 0.0146au, the
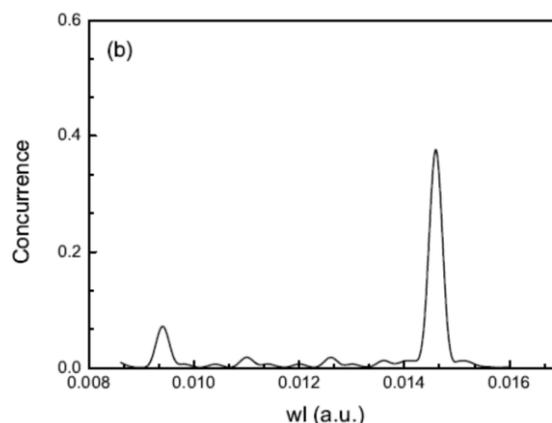

Figure 2: The mean values of the concurrence over long time T for initial state $|0,2\rangle$ as a function of laser frequency