Dynamical correlation between quantum entanglement and intramolecular energy in molecular vibrations: An algebraic approach*

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The dynamical correlation between quantum entanglement and intramolecular energy in realistic molecular vibrations is explored using the Lie algebraic approach. The explicit expression of entanglement measurement can be achieved using algebraic operations. The common and different characteristics of dynamical entanglement in different molecular vibrations are also provided. The dynamical study of quantum entanglement and intramolecular energy in small molecular vibrations can be helpful for controlling the entanglement and further understanding the intramolecular dynamics.

Keywords: quantum entanglement, molecular vibration, intramolecular energy, Lie algebra

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1. Introduction

Since the definition of quantum computers was put forward by Feynman in 1982, various physical systems have been proposed to realize quantum computing algorithms. These physical systems have been proven suitable for representing a limited number of qubits in experiments, such as cavity quantum electrodynamics,[1,2] trapped ions,[3,4] and nuclear magnetic resonance.[5–7] Quantum computing based on the vibrational states of molecules has also been proposed by Vivie–Riedle and her co-workers in 2001.[8–10] In this approach, the qubit is performed by different eigenstates of vibrational modes or rotational modes of molecules, and a high-fidelity quantum logic gate is achieved by applying the optimal control theory. In 2002, the experimental implementation of the Deutsch–Jozsa algorithm for three-qubit functions using the pure coherent Li_2 superpositions of rovibrational eigenstates has also been proposed.[11] Since then, this approach has attracted increasing attention and achieved tremendous progress in recent years.[12–20]

The molecular vibrational or rotational state approach has three advantages compared with the previously referred physical models. First, the operations of multiple qubits can be realized conveniently by several vibrational modes or rotational modes in the molecules. Second, the nonlinear coupling in the vibrational modes or rotational modes of the molecules is the most important resource for quantum parallel computing. Third, various research has shown that quantum calculation based on molecules may obtain a high fidelity of quantum computing.[11,12,21,22] The following is the five basic requirements for performing a high-fidelity quantum computing:[23,24] (i) the energy level structure of the system can represent the quantum state of a qubit; (ii) the system can be initialized to produce one specific quantum state (the desired input); (iii) the coupling of the system and the external environment is weak, which can maintain a long decoherence time; (iv) a certain interaction between different qubits should exist so that the qubits can be manipulated; and (v) the output results can map the measurements. The molecular system has rich and clear vibrational energy level structure that can be used to present qubits. Extensive theoretical and experimental research has fully proven the controllability of the laser field to the vibration and rotation of a molecule. Thus, the desired quantum state of the system can be prepared and the interaction between the vibrational modes can be adjusted. Based on this analysis, the vibrational or rotational modes of polyatomic molecules are good candidates for presenting the qubits.

Several molecular systems have been proposed to perform the qubits;[8,9,11,15,25–30] the quantum coherent control theory (QCCT) and the optimal control theory (OCT) have been applied to implement the quantum logic gate operation.[15,25–27] Given the progress of the research on quantum computing based on molecular vibrational or rovibrational eigenstates, the importance of studying the intramolecular properties is gradually highlighted. For example, the entanglement between different rovibrational modes is a key resource for quantum computing; the intramolecular energy transfer can be used to control the transition between different eigenstates of vibrational or rotational modes; and the control of the intramolecular decoherence can be helpful for quantum computing. Thus, the dynamical study of quantum entangle-

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ment and intramolecular energy in different systems is becoming an important issue, and several research groups have focused on the dynamical behavior of entanglement in molecules in recent years.[31–39] Although the definition of entanglement is irrelevant to the dynamics, both the preparation and transmission of entanglement are the dynamical processes. Quantum entanglement is important in quantum computation and quantum information, as well as a key element in quantum teleportation, fast quantum algorithms, and quantum error-correction.[23] The research on dynamical entanglement can help further understand the behavior of entanglement in the quantum information process and control the entanglement in quantum computing.

Several measurement methods have been applied to qualify quantum entanglement, each representing different characteristics of entanglement. The Von Neumann entropy has been used as the entanglement measure in our previous paper.[38,40] However, this method is not appropriate for measuring entanglement when the state becomes a mixed one, which is consistent with Refs. [41] and [42]. Most research on entanglement has applied concurrence and negativity as entanglement measurement tools. The concept of negativity is important because it leads to the upper bounds for the entanglement of distillation.[41,43] The concurrence is monotonically related to the entanglement of formation and does not change under local operations and classical communications. The range of the value for the concurrence is from zero to one: zero denotes that the system is in disentanglement, whereas one indicates that the system is in entanglement.[44,45] A precise quantitative picture of entanglement is also important to obtain a better insight into the corresponding physical systems. In addition, the definition of the tripartite quantum system based on the concurrence measure has also been proposed.[46,47] Given that the algebraic model can be expanded to discuss the influence of the molecular bending vibration, the tripartite entanglement can be also discussed using the concurrence measure later. Furthermore, studies of the dynamical correlation between quantum entanglement and intramolecular vibrational energy are worthwhile because the occurrence of intramolecular vibrational energy redistribution is inevitable in molecular vibrations.

In this study, a dynamical algebraic approach has been proposed to discuss the dynamical correlation between entanglement and intramolecular energy in realistic molecular vibrations. The Lie algebraic model of the molecule has been applied successfully to study vibrations in polyatomic molecules,[48–52] and the highly excited vibrational states can be well described by the algebraic method. In recent years, the dynamical algebraic method has been extended to study an interacting quantum system, the dynamical symmetries in nanophysics, the decoherence in a general three-level system,[53–56] and so on. In the present work, the Lie algebra quadratic anharmonic model is used to represent an isolated linear triatomic molecular system; the HCN and DCN molecules are chosen as examples for comparison. The stretching vibrational energies have also been calculated, and are consistent with the experimental results, indicating that the Hamiltonian can reproduce the real situations of stretch vibrations in the molecules.[57]

This paper proceeds as follows. In Section 2, the Lie algebra quadratic anharmonic model is first reviewed briefly, and the relevant theoretical formulas are given using the algebraic model. In Section 3, the concurrence and the negativity are calculated and discussed; the mean values of the stretching vibrational energy and the Lyapunov function are studied; and the dynamical correlation between entanglement and intramolecular energy is discussed. Finally, some concluding remarks are given in Section 4.

2. Theory

2.1. The algebraic model of molecules

The Lie algebra quadratic anharmonic model provides the algebraic Hamiltonian of a nonrotating Morse oscillator. The algebraic Hamiltonian can be expanded as a power series in the linear Casimir invariant of the subalgebra O(2) of the algebra U(2), leading to accurate fits to the vibrational energy levels of the diatomic molecules up to the dissociation limit. In addition, the coordinate space representation of the algebraic Hamiltonian can become a power series expansion in half integral powers of the Morse Hamiltonian with a corresponding classical potential expanded in half integral powers of the Morse potential function. This algebraic model has been proven to accurately fit the vibrational energy levels of diatomic molecules, as well as potential function, including the zero-point energy, and achieve the known value of dissociation energy,[58,59] which can be represented as

$$\mathcal{H} = \hbar\omega_0\left(\hat{C}^\dagger\hat{C} + \frac{\hat{C}_0}{2}\right), \tag{1}$$

where the three operators $\hat{C}^\dagger$, $\hat{C}$, and $\hat{C}_0$ are the quadratic operators constructed by the generators of $U(2)$ algebra. They obey the close commutation relations

$$\begin{aligned}
&[\hat{C}, \hat{C}^\dagger] = \hat{C}_0,\\
&[\hat{C}_0, \hat{C}] = -2\alpha_0\hat{C},\\
&[\hat{C}_0, \hat{C}^\dagger] = 2\alpha_0\hat{C}^\dagger,
\end{aligned} \tag{2}$$

where α_0 is the anharmonicity parameter. Then, operating on the eigenstate $|N, v\rangle$, we can obtain

$$\begin{aligned}
&\hat{C}^\dagger|N, v\rangle = \sqrt{(1-\alpha_0 v)(v+1)}|N, v+1\rangle,\\
&\hat{C}|N, v\rangle = \sqrt{[1-\alpha_0(v-1)]v}|N, v-1\rangle,\\
&\hat{C}_0|N, v\rangle = (1-2\alpha_0 v)|N, v\rangle,
\end{aligned} \tag{3}$$

where N denotes the total vibrational quantum number.[58]

According to the above model, the Hamiltonian of a linear triatomic molecule can be represented as two-coupled quadratic anharmonic oscillators.[57,60] In other words, the two molecules can be taken as the coupling of the two Morse oscillators

$$\begin{aligned}\mathcal{H} &= \hbar\omega_{01}\left(\hat{C}_1^{\dagger}\hat{C}_1+\frac{\hat{C}_{01}}{2}\right)+\hbar\omega_{02}\left(\hat{C}_2^{\dagger}\hat{C}_2+\frac{\hat{C}_{02}}{2}\right)\\ &\quad-\kappa(\hat{C}_1^{\dagger}\hat{C}_2+\hat{C}_2^{\dagger}\hat{C}_1),\end{aligned} \tag{4}$$

where ω_{01} and ω_{02} are the angular frequencies of the two bonds of the triatomic molecule, and κ is the coupling coefficient that can be determined by the experimental values.[57]

2.2. Entanglement measurement

A generalization of the concurrence of a bipartite quantum state in an arbitrary dimension is defined by[44]

$$\mathcal{C}(|\Psi\rangle)=\sqrt{2(\langle\Psi|\Psi\rangle^2-\mathrm{Tr}\rho_{\mathrm{r}}^2)}, \tag{5}$$

where the reduced density matrix $\rho_{\mathrm{r}}=\mathrm{Tr}_{\mathrm{p}}|\Psi\rangle\langle\Psi|$ is obtained as a partial trace of the bipartite state. The "ρ_{r}" denotes the reduced density matrix and "Tr_{p}" denotes a partial trace of the bipartite state. The negativity is defined as twice the absolute value of the sum of the negative eigenvalues[41]

$$\mathcal{N}(\rho)=2\max\{0,-\lambda_{\mathrm{neg}}\}, \tag{6}$$

where λ_{neg} is the sum of the negative eigenvalues of the partial transpose of the state ρ, and the partial transpose is given by

$$\rho_{i\alpha,j\beta}^{\mathrm{T}_2}=\rho_{i\beta,j\alpha}, \tag{7}$$

where ρ^{T_2} denotes the partial transpose for the second subsystem.

For the time-independent Hamiltonian $\mathcal{H}$, the time-dependent wave function is given by

$$|\Psi(t)\rangle=\hat{U}(t)|\Psi(0)\rangle=\mathrm{e}^{-\mathrm{i}t\mathcal{H}}|\Psi(0)\rangle, \tag{8}$$

where $|\Psi(0)\rangle$ is the initial state of the quantum system. In this function, $\hbar=1$ as we choose the atomic units. The whole dynamical information of a quantum system can be achieved from the time-dependent wave function. The reduced density matrix can be represented as

$$\rho_1(t)=\mathrm{Tr}_2\,\rho(t)=\mathrm{Tr}_2|\Psi(t)\rangle\langle\Psi(t)|, \tag{9}$$

where Tr_2 denotes the partial trace over the second subsystem.

The mean values of the stretching vibrational energy of bond i can be given by the full density matrix

$$\langle\mathcal{E}_i\rangle(t)=\mathrm{Tr}\{\rho(t)\mathcal{H}_i\}, \tag{10}$$

where H_i is the stretching Hamiltonian of bond i.

The Lyapunov method is applied to describe the stability and other properties of the dynamical system. The Lyapunov function has been used to study the stability of the quantum control system.[61–65] Pykh[66] has also proven that the Lyapunov function (fitness-like and entropy-like energy Lyapunov functions) can be used to study the diversity of the system. Several definitions of the Lyapunov function have been proposed, such as the definition of the Lyapunov function given by Mirrahimi[61,62] as $\mathcal{L}(\Psi)=||\Psi-\Phi||^2/2$; the other definitions have been given by Beauchard[65] as $\mathcal{L}(\Psi)=1-|(\Psi|\Phi)|^2$. Both definitions are employed to investigate the control of the target states in the linear systems. According to both definitions, the Lyapunov function is applied in this study to describe the distance between the final state $|\Psi(t)\rangle$ and the initial state $|\Psi(0)\rangle$ at time t

$$\mathcal{L}(t)=1-|\langle\Psi(0)|\Psi(t)\rangle|^2, \tag{11}$$

where its zero values describe the recurrence times from the definition.

The initial states can be written as[33]

$$|\Psi(0)\rangle=|N_1,n_1\rangle\otimes|N_2,n_2\rangle\equiv|n_1,n-n_1\rangle, \tag{12}$$

where $n=n_1+n_2$ is the total quantum number of two stretching bonds. Given that the product Fock state can hold clues to quantum memory components, and has an important role in quantum computing,[67] discussion on the dynamical behaviors of concurrence is useful for quantum computation.

Putting Eq. (12) into Eq. (8) yields

$$\begin{aligned}|\Psi(t)\rangle &= \mathrm{e}^{-\mathrm{i}t\mathcal{H}}|\Psi(0)\rangle\\ &= \mathrm{e}^{-\mathrm{i}t\mathcal{H}}|n_1,n-n_1\rangle\\ &= \sum_{k=0}^{\infty}\frac{(-\mathrm{i}t)^k}{k!}\mathcal{H}^k|n_1,n-n_1\rangle\\ &= \sum_{k=0}^{\infty}\sum_{\max\{-k,-n_1\}}^{\min\{k,n-n_1\}}\mathcal{Y}_k^j(t)|n_1+j,n-n_1-j\rangle\\ &= \sum_{j=-n_1}^{n-n_1}\mathcal{Y}^j(t)|n_1+j,n-n_1-j\rangle,\end{aligned} \tag{13}$$

where $\mathcal{Y}^j(t)=\sum_{k=|j|}^{\infty}\mathcal{Y}_k^j(t)$. When $k=0$, $\mathcal{Y}_0^j(t)=\delta_{0,j}$, and when $k=1,2,\ldots,\infty$, the recursive expression $\mathcal{Y}_k^j(t)$ is

$$\begin{aligned}\mathcal{Y}_k^j(t) &= -\frac{\mathrm{i}t}{k}\cdot\Big\{\eta_0(n_1+j,n-n_1-j)\mathcal{Y}_{k-1}^j(t)\\ &\quad+\eta_+(n_1+j,n-n_1-j)\mathcal{Y}_{k-1}^{j+1}(t)\\ &\quad+\eta_-(n_1+j,n-n_1-j)\mathcal{Y}_{k-1}^{j-1}(t)\Big\},\end{aligned} \tag{14}$$

where the function η_0, η_+, η_- can be obtained using algebraic operations (see Eq. (3))

$$\begin{aligned}\eta_0(v_1,v_2) &= \hbar\omega_{01}(v_1^2-\alpha_{01}v_1^2)+\hbar\omega_{02}(v_2^2-\alpha_{02}v_2^2),\\ \eta_+(v_1,v_2) &= -\kappa\sqrt{(1-\alpha_{01}v_1)(v_1+1)}\end{aligned}$$

$$\times\sqrt{[1-\alpha_{02}(v_2-1)]v_2},$$
$$\eta_-(v_1,v_2) = -\kappa\sqrt{[1-\alpha_{01}(v_1-1)]v_1}$$
$$\times\sqrt{(1-\alpha_{02}v_2)(v_2+1)}, \quad (15)$$

where $v_1 = n_1 + j$, and $v_2 = n - n_1 - j$.

The analytical expression of the reduced-density matrix can be given based on Eqs. (9) and (13) by

$$\rho_1(t) = \mathrm{Tr}_2\{|\Psi(t)\rangle\langle\Psi(t)|\}$$
$$= \sum_{j=-n_1}^{n-n_1}\sum_{j'=-n_1}^{n-n_1}\mathcal{Y}^j(t)\mathcal{Y}^{j'}(t)^*|n_1+j\rangle\langle n_1+j'|\delta_{j,j'}$$
$$= \sum_{j=-n_1}^{n-n_1}|\mathcal{Y}^j(t)|^2|n_1+j\rangle\langle n_1+j|. \quad (16)$$

The concurrence and the Lyapunov function for a general pure bipartite state can be given, respectively, by putting Eq. (16) into Eqs. (5), and (11)

$$\mathcal{C}(t) = \sqrt{2(1-\mathrm{Tr}_1\rho_1(t)^2)} = \sqrt{2\left(1-\sum_{j=-n_1}^{n-n_1}|\mathcal{Y}^j(t)|^4\right)}, \quad (17)$$

$$\mathcal{L}(t) = 1-|\langle\Psi(0)|\Psi(t)\rangle|^2 = 1-|\mathcal{Y}^0(t)|^2. \quad (18)$$

The negativity can be calculated by using Eqs. (6) and (13). The mean values of the stretching vibrational energy for each bond can also be obtained from Eqs. (10) and (13). The dynamical study of entanglement and intramolecular energy in small triatomic molecules can be discussed.

The mean values of the Lyapunov function and the concurrence over a long time T are defined by

$$\overline{\mathcal{C}} = \frac{1}{T}\int_0^T\mathcal{C}(t)\,\mathrm{d}t,$$
$$\overline{\mathcal{L}} = \frac{1}{T}\int_0^T\mathcal{L}(t)\,\mathrm{d}t. \quad (19)$$

3. Dynamical study of quantum entanglement and intramolecular stretching vibrational energies

As a concrete example, the triatomic molecules HCN and DCN are used to study the dynamical correlation between entanglement and stretching vibrational energies. The corresponding molecular parameters have been given in Table 1 published in our previous work.[57] The parameters m, ω_0, α_0, a, and D denote the reduced mass, angular frequency, anharmonicity parameter, Morse parameters, and the dissociation energy of the corresponding chemical bond, respectively. The coefficient κ is the coupling coefficient of HCN or DCN determined by the experimental energy values. The stretch vibrational levels of HCN and DCN molecules have been calculated using the algebraic Hamiltonian. The relative errors are all smaller than 0.9% (see Table 2 in Ref. [57]), which indicates that the Hamiltonian can reproduce the real situations of the stretch vibrations in molecules. All calculations have been carried out in atomic units except that the unit of energy is cm^{-1}, so the stretching vibrational energies are provided in units of cm^{-1}.

Here is the organization of this section. In subsection 3.1, quantum entanglement is studied in order to measure the concurrence and the negativity. In subsection 3.2, we provide the mean values of the stretching vibrational energy, the mean values of the concurrence, and the mean Lyapunov function. Then, the dynamical correlation between entanglement and intramolecular energy is discussed. The same and different characteristics of the two molecules are also summarized.

3.1. Quantum entanglement measures

First, the negativity and concurrence for various initial states $n_1 = 0, 1, \ldots, 20$ $(n = 20)$ are calculated to obtain a better exhibition of the difference of dynamical entanglement with various initial states. Only the results $n_1 = 0, 5, 10, 15, 20$ are shown. The concurrences for various initial states of the HCN and DCN molecules are given in Figs. 1 and 2. The negativities for various initial states of the HCN and DCN molecules are shown in Figs. 3 and 4. Although the pendulous curves have some differences, the two measures provide similar periodic behaviors. Thus, we only discuss the concurrence. Figures 1 and 2 show that the concurrences have better quasi-periodic behaviors for the local-mode initial states ($|0,20\rangle$ and $|20,0\rangle$) than those for the quasi-normal-mode states ($|5,15\rangle$ and $|15,5\rangle$) and normal-mode states ($|10,10\rangle$). The entanglement can achieve and preserve the maximum value during evolution for the quasi-normal-mode initial states, especially for the state $|10,10\rangle$. That is, a quantum normal-mode initial state can be prepared to obtain larger entanglement in quantum computing. Compared with the dynamical entanglement of DCN, the oscillation periods of HCN are shorter and the maximum of entanglement occurs earlier. The angular frequency of the CH bond is higher than the CD bond, as shown in Table 1 of Ref. [57]. In addition, the CH bond is easier to excite based on our pervious study of multiphoton excitation of the two molecules, which may be the reason why the maximum entanglement occurs earlier for the HCN molecule.

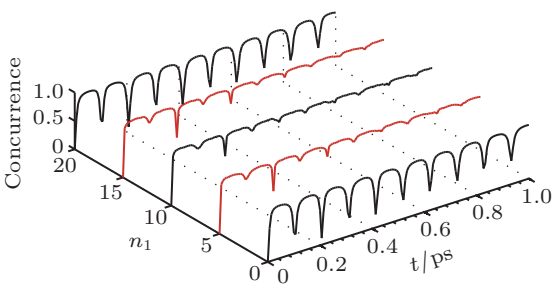


Fig. 1. (color online) The concurrence for the initial states $|n_1, n-n_1\rangle$ in the HCN with $n_1 = 0$, 5, 10, 15, 20.

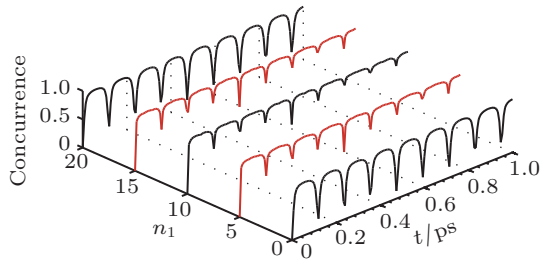


Fig. 2. (color online) The concurrence for the initial states $|n_1, n-n_1\rangle$ in the DCN with $n_1 = 0$, 5, 10, 15, 20.

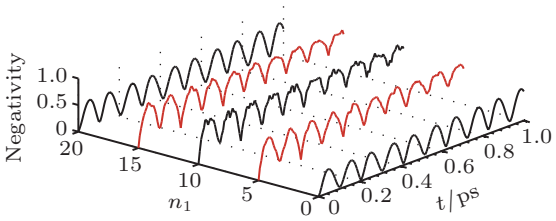


Fig. 3. (color online) The negativity for the initial states $|n_1, n-n_1\rangle$ in HCN with $n_1 = 0$, 5, 10, 15, 20.

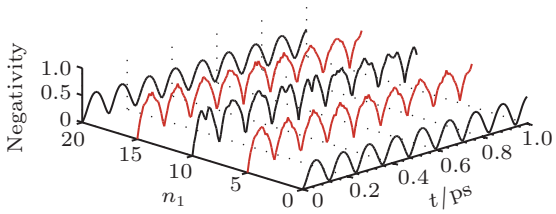


Fig. 4. (color online) The negativity for the initial states $|n_1, n-n_1\rangle$ in the DCN with $n_1 = 0$, 5, 10, 15, 20.

3.2. Dynamical correlation between entanglement and intramolecular energy

This subsection provides the dynamical behavior of the stretching vibrational energies for the initial states ($n_1 = 0$, 5, 10, 15, 20) in the HCN and DCN molecules. The corresponding eigenvalues of stretching vibrational energies for different initial states of the two molecules are given in Table 1. The dynamical behavior of the stretching vibrational energies of each bond in the HCN and DCN are given in Figs. 5 and 6. These figures show that the stretching vibrational energies for the local-mode initial states $|0, 20\rangle$ and $|20, 0\rangle$ present a typical normal mode vibrational behavior, whereas energies are the typical local mode vibrations for the normal-mode initial state $|10, 10\rangle$. Both figures show that vibrations of the C–H (C–D) and C–N bonds represent normal-local-normal transitions with the increase in the initial state n_1. Furthermore, the periods of stretching vibrational energies and dynamical entanglement have the same characteristics, and the oscillation periods of HCN are shorter than that of DCN. The mean values of the concurrence and the Lyapunov function for the initial states $|n_1, n-n_1\rangle$ in the HCN and DCN with $n_1 = 0$, 1, …, 20 are given in Fig. 7, which shows that the concurrence has larger values near the normal-local-normal transitions. At the same time, the maximum values of entanglement also arise near the normal-mode initial states, as shown in Figs. 1 and 2. Therefore, the behavior of vibrational entanglement can be taken as the dynamical sign of normal-local-normal vibrational mode transitions, which coincides with other literatures.[32]

Table 1. Vibrational energies (in cm^{-1}) of different initial states for the HCN and DCN molecules.

$(n_1, n-n_1)$	(0,20)	(5,15)	(10,10)	(15,5)	(20,0)
HCN	29207.92	30267.95	31208.43	33092.33	34196.36
DCN	26878.52	27462.93	28759.17	30879.27	31084.93

Furthermore, entanglement and chaos may have certain relationships based on the three aspect analysis of the results. The first reason is the mean value of the calculated concurrence. Figure 7 shows that the maximum entanglement occurs in the intermediate energy range when the initial local-mode state transforms into the normal-mode state. The second reason is the calculated Lyapunov function, which can describe the "distance" between the final state and the initial state at time t. The final state in the evolution may still stay at the initial state when the value of the Lyapunov function is zero at time t, and the state in the evolution will transit to other states completely when the value of the Lyapunov function is equal to 1. Thus, the values of the Lyapunov function are close to one in the intermediate energy range, as shown in Fig. 7. The third reason is the area of the chaotic motion. Higher excitation energy corresponds to higher degree of chaos in general; however, the energy of excitation is not a unique factor for the occurrence of chaos. Researchers have found that the chaotic motion occurs near the separatrix of the local and normal vibrational modes in the intermediate energy range.[68,69] The range of producing the maximum entanglement is consistent with the area of the chaotic motion, which inspires us to study the relationship or the influence between them. Although we have found some chaotic trajectories in the intermediate energy range of the two molecules, we have not found the direct correspondence between the initial states and the chaotic trajectories. Therefore, this problem will be discussed elsewhere. Compared with the DCN molecule, the mean values of the Lyapunov function and the concurrence in HCN decrease rapidly when the vibrational quantum number increases at

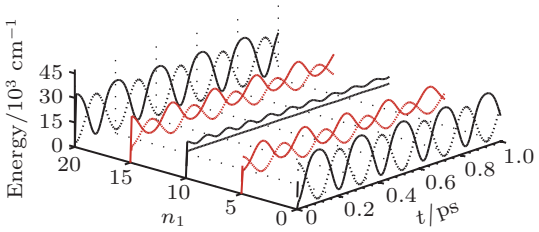


Fig. 5. (color online) Dynamical behavior of the stretching vibrational energies for the initial states $|n_1, n-n_1\rangle$ in HCN with $n_1 = 0$, 5, 10, 15, 20. The solid line represents the stretching vibrational energy of the C–H bond and the dashed line represents the stretching vibrational energy of the C–N bond.

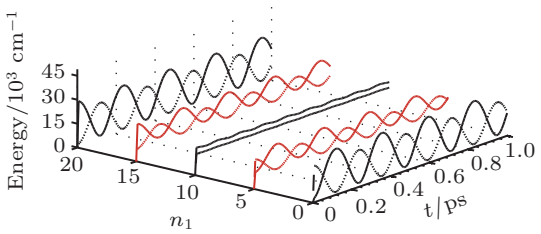


Fig. 6. (color online) Dynamical behavior of the stretching vibrational energies for the initial states $|n_1, n-n_1\rangle$ in the DCN with $n_1 = 0$, 5, 10, 15, 20. The solid line represents the stretching vibrational energy of the C–D bond and the dashed line represents the stretching vibrational energy of the C–N bond.

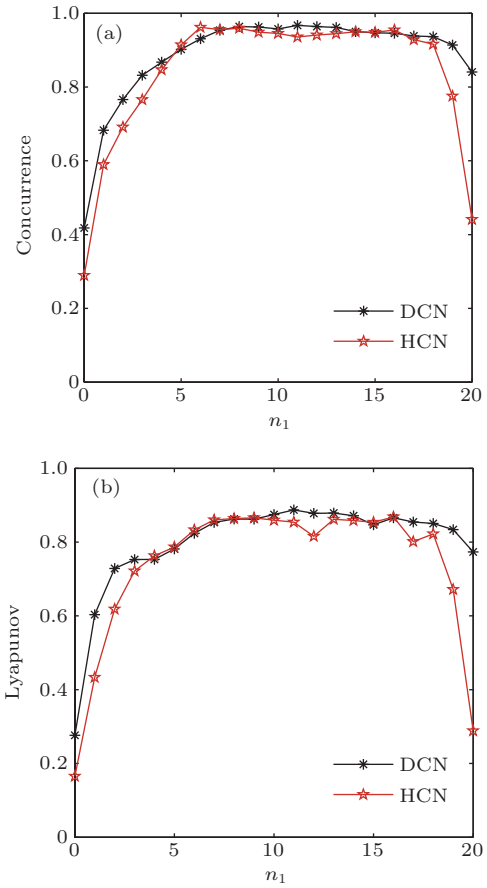


Fig. 7. (color online) Mean values of the concurrence (a) and the Lyapunov function (b) for the initial states $|n_1, n-n_1\rangle$ in HCN and DCN with $n_1 = 0, 1, \ldots, 20$.

$n = 20$. The vibrational anharmonicity of the C–H bond in the HCN is stronger than that of the C–D bond in DCN, as shown in Table 1. In addition, the decoherence caused by the IVR in HCN appear earlier than that in the DCN in our previous study of multiphoton vibrational excitation for the two molecules.[60] Thus, the phenomenon of earlier and faster decline of the HCN may indicate that decoherence will destroy the entanglement earlier in HCN during the process of the control on the entanglement.

4. Conclusions

We have studied the dynamical correlation between quantum entanglement and intramolecular energy using the Lie algebraic approach. We calculated the dynamical entanglement using the concurrence and negativity measures, and found similar periodic behaviors for the initial Fock states although the pendulous curves have some differences. We found the common and different characteristics of dynamical entanglement in the vibrations of the HCN and DCN molecules. The concurrence has better quasi-periodic behavior for the local-mode initial states than that for the normal-mode states. The maximum mean values of the concurrence and the Lyapunov function occur near the normal-mode initial states, indicating that the local-mode initial state may be favorable for preparing a qubit, and the normal-mode state can achieve larger entanglement in quantum computing. We also found that the normal-to-local transition of the stretching vibrational energies corresponds to the entanglement with the initial state from the local mode to the normal mode, indicating that the behavior of vibrational entanglement may become the dynamical sign of normal–local–normal vibrational transitions. In addition, the entanglement of DCN has longer period and larger mean values than that of HCN. The decoherence of entanglement appears later than HCN, indicating that DCN is a more reasonable candidate in quantum computation based on the vibrational states of realistic molecules.

Understanding of the dynamical properties of the entanglement extensively can help achieve the control of the entanglement, and better understand the intramolecular dynamics. Furthermore, the observed periodic pendulation phenomenon will also make the states usable, whether the maximum entanglement or the disentanglement are both necessary during the quantum transmission. However, for the operation of vibrational quantum computers, the main issues are the construction of the qubit and the implementation of a quantum logic gate, and the laser control is necessary for implementing the construction of the qubit and the quantum logic operation. We have begun to study the quantum control of the entanglement using the algebraic model, and strategies for the operation of the vibrational quantum computing will be introduced. An external bath is often needed in real situation, which will give rise to decoherence and energy transfer. For the molecular vibrational qubits, decoherence may come from the collision with other molecules, the intramolecular anharmonic resonances with the remaining vibrational modes, the rotational freedom, and the electronic freedom. Collisions may be ignored when the gas pressure is extremely low. Only the coupling of the two stretching vibrations are considered in the present work. However, when the algebraic model and the approach are proven to be applied to study the dynamical intermolecular entanglement of vibrations successfully, the approach can be extended to study the situation where an external bath (the bending vibration or the rotational effects) is considered. The relevant work is in progress.

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