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Dynamical entanglement of vibrations in an algebraic model

Xi-Wen Hou,[1] Jing-Hua Chen,[2] and Zhong-Qi Ma[3]
[1]*Department of Physics, Huazhong Normal University, Wuhan 430079, China*
[2]*Department of Information Management, Huazhong Normal University, Wuhan 430079, China*
[3]*Institute of High Energy Physics, P.O. Box 918(4), Beijing 100049, China*

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The dynamical entanglement of two stretching vibrations of triatomic molecules H_2O and SO_2 in an algebraic model is studied in terms of the reduced-density linear entropy with initial states that are, respectively, taken to be the product of Fock states and coherent states on each bond. It is shown that the entanglement of initial states with local-mode character is regular while that of states with normal-mode character is irregular. For initial coherent states with a small amplitude, the regularity of the entanglement with a long period appears in SO_2 and "classical" beat phenomena of the entanglement happens in H_2O, where the period of the beat is longer. Those long periods of the entanglement indicate that the entanglement sustains long enough so that quantum information process and quantum computation could be accomplished.

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I. INTRODUCTION

Entanglement has become a potential physical resource in quantum information processing that has undergone a rapid development in recent years [1]. Since quantum entangled states play an important role in deposit and transport of quantum information, considerable effort has been devoted to generating entanglement in quantum-optic and atomic systems [2–6] and characterizing entanglement in various systems [7–10]. The possibility for the generation of the maximally entangled states with a fixed photon number from squeezed vacuum states is theoretically studied [3]. Such a kind of entangled source can be applied in quantum teleportation [4]. Atom-photon entangled coherent states in atomic Bose-Einstein condensate can be prepared via electromagnetically induced transparency [5]. It is shown that the nonlinear interaction between excitons in two coupled semiconductor microcrystallites increases the maximum values of entanglement more than that of the linear coupling model [6]. Besides generation of entangled states, the study of entanglement characteristics of various systems helps one to relate those characteristics to the understood properties of the system. It is demonstrated that the entanglement in several models [8] is largest near quantum critical points. It is suggested that the small-amplitude oscillations of entanglement might be regarded as fingerprints of the underlying classical chaos in smooth Hamiltonian systems [9]. A signature of quantum chaos based on dynamics of entanglement is given for the kicked top [10]. One of the important properties in those studies [2–10] is that the model parameter can be varied for various behaviors of entanglement. Thus those works are, to some extent, limited to ideal theoretical models. For molecular systems, a scheme is proposed to generate entanglement between two coupled identical polar molecules separated in a distance of tens of nanometers [11]. The possibility that a suitably excited molecule can mimic the logical action of a Turing machine is analyzed in [12]. Recent theoretical studies have highlighted the feasibility of using molecular vibrational states for quantum computing [13,14]. It is shown that extremely high quantum gate fidelity can be achieved for a qubit system based on vibrational eigenstates of diatomic molecules and the increase of fidelity can be realized with an increase of anharmonicity parameter in the molecules [14]. In present work, we investigate the entanglement dynamics of realistic molecular vibrations in an algebraic model [15], where model parameters have been optimized from experimental spectra of triatomic molecules H_2O and SO_2. Thereby the quite different behaviors of entanglement in those two molecules can be characterized.

Algebraic methods [16] are developed to be an effective theory for descriptions of vibrations, rotations, and rotation-vibration interactions in molecules [17]. For highly excited vibrations of polyatomic molecules, various algebraic models [15,18–22] and some important physical quantities such as band intensities [19] and potential-energy surfaces [20] are satisfactorily analyzed and interpreted. The simplest Lie algebraic Hamiltonian is within the u(2) algebraic framework. It is shown that a u(2) force-field expansion has a much faster convergence than the conventional force-field expansion [21]. Furthermore, a simple scheme is proposed to extract the corresponding classical dynamics from the u(2) model parameters [15] and to incorporate bend modes with possible interactions between vibrational modes [22]. However, the quantum dynamics of highly excited vibrations within algebraic frameworks [15,18–22], especially on the dynamics of quantum entanglement, has not yet been considered. Here we study the dynamical entanglement of stretching vibrations of H_2O and SO_2 in a u(2) algebraic model [15] that conserves a multiplet quantum number (MQN). The reduced-density linear entropy is employed to measure the entanglement with various MQN and initial states. The initial states are taken to be the product of Fock states and coherent states on each bond, respectively. It is demonstrated that the entanglement of initial states with local-mode character is regular while that of states with normal-mode character is irregular. Using a simplified model, we explain the period of regular entanglement of a pure state in H_2O and coherent states with a small amplitude in H_2O and SO_2. The entanglement of coherent states in H_2O can, under a condition, show a beat phenomenon with a time evolution very similar to that in classical physics. The entanglement of coherent states with small amplitude for SO_2 exhibits regular with a longer period, indicating that the entanglement lasts long enough for the quantum information process and quantum computation to be accomplished.

The paper is organized as follows. Section II presents u(2) algebraic model for H_2O and SO_2. In Sec. III we study the dynamics of entanglement with two kinds of initial states. We conclude the paper with discussions in the last section.

II. u(2) ALGEBRAIC MODEL

For stretching vibrations in a symmetrical A-B-A triatomic molecules, the algebraic Hamiltonian [15] reads,

$$\hat{H} = \hat{H}_0(v_a,v_c) + \hat{H}_i, \tag{1}$$

where $\hat{H}_0(v_a,v_c)$ and $\hat{H}_i$ is, respectively, diagonal and nondiagonal Hamiltonian with $v_{a(c)}$ representing the vibrational quantum number on stretch bond $a(c)$, which is given by,

$$\begin{aligned}\hat{H}_0(v_a,v_c) &= a^{(1,0)}_{(a,c)}(\hat{C}_a + \hat{C}_c) + a^{(2,0)}_{(a,c)}(\hat{C}_a^2 + \hat{C}_c^2) + 2a^{(1,1)}_{(a,c)}\hat{C}_a\hat{C}_c \\ &\quad + a^{(3,0)}_{(a,c)}(\hat{C}_a^3 + \hat{C}_c^3) + a^{(2,1)}_{(a,c)}\hat{C}_a\hat{C}_c(\hat{C}_a + \hat{C}_c),\end{aligned} \tag{2}$$

$$\hat{H}_i = m^{(1)}_{(a,c)}(\hat{f}_a^+\hat{f}_c^- + \hat{f}_a^-\hat{f}_c^+) + m^{(2)}_{(a,c)}(\hat{f}_a^+\hat{f}_a^+\hat{f}_c^-\hat{f}_c^- + \hat{f}_a^-\hat{f}_a^-\hat{f}_c^+\hat{f}_c^+), \tag{3}$$

where $a^{(i,j)}_{(a,c)}$ and $m^{(i)}_{(a,c)}$ $(i=1,2,3$, and $j=0,1)$ are parameters, the subscript a and c denote the two equivalent local-stretch vibrations, and $\hat{C}_i$ and $\hat{f}_i^{\pm}$ $(i=a,c)$ are u(2) operators with the matrix elements being,

$$\langle N_i,v_i'|\hat{C}_i|N_i,v_i\rangle = \left[\left(1+\frac{1}{N_i}\right)\left(v_i+\frac{1}{2}\right) - \frac{1}{N_i}\left(v_i+\frac{1}{2}\right)^2\right]\delta_{v_i',v_i}, \tag{4}$$

$$\langle N_i,v_i'|\hat{f}_i^+|N_i,v_i\rangle = \sqrt{(v_i+1)\left(1-\frac{v_i}{N_i}\right)}\,\delta_{v_i',v_i+1}, \tag{5}$$

$$\langle N_i,v_i'|\hat{f}_i^-|N_i,v_i\rangle = \sqrt{v_i\left(1-\frac{v_i-1}{N_i}\right)}\,\delta_{v_i',v_i-1}, \tag{6}$$

where $|N_i,v_i\rangle$ $(i=a,c)$ is the local basis on the stretch bond i with N_i being related to the number of bound states for the corresponding anharmonic oscillator [17]. The algebraic Hamiltonian Eq. (1) conserves MQN, which is defined by

$$v_m = v_a + v_c, \tag{7}$$

so that in addition to the energy there are two integrals of the motion, and the Hamiltonian matrix is block diagonal with respect to MQN. Fitting the experimental spectra of H_2O and SO_2, Iachello and co-workers have listed the optimized parameters in the model in Table I of Ref. [15], where the classical dynamics of the classical counterpart of Eq. (1) has been analyzed.

III. DYNAMICS OF ENTANGLEMENT

We study the dynamics of entanglement in the algebraic model Eq. (1). There have been various measures of entanglement in the literature, and we choose the linear entropy for its simplicity as well as its wide applicability. The linear entropy of entanglement, $s(t)$, is defined by [23]

$$s(t) = 1 - \mathrm{Tr}_a[\rho_a(t)]^2, \tag{8}$$

where Tr_a denotes the trace over the first subsystem a, and $\rho_a(t)$ is the reduced-density matrices, $\rho_a(t)=\mathrm{Tr}_c|\psi(t)\rangle\langle\psi(t)|$, where index a and c stand for the degree of freedom on stretch mode a and c, respectively, and $|\psi(t)\rangle$ is the quantum state of the full system, which evolves in time under the action of algebraic Hamiltonian Eq. (1).

We employ two kinds of initial states, Fock states and coherent states, to inspect the time evolution of the entanglement. The importance of studying in detail the entanglement in different initial states is threefold. It may be viewed as a key to the understanding of some of the striking differences between the quantum and classical description of the world. Since Fock states for H_2O are of local-mode or normal-mode character, we show that they display quite different behaviors of the entanglement. Continuous-variable-type entangled states including squeezed states and coherent states have been widely applied to quantum teleportation [4], quantum cryptography [24], and quantum computation [25]. Therefore it is an interesting topic to investigate the entanglement of coherent states.

A. Entanglement of Fock states

In order to explore the dynamical properties of quantum entanglement of highly excited vibrations in H_2O and SO_2, we take MQN v_m to be 15, 20, and 25, as an example. Within every v_m two initial states are taken to be

$$|\psi(0)\rangle_A = |N_a,0\rangle \otimes |N_c,v_m\rangle \equiv |0,v_m), \tag{9}$$

$$|\psi(0)\rangle_B = \begin{cases} \left|N_a,\dfrac{v_m}{2}\right\rangle \otimes \left|N_c,\dfrac{v_m}{2}\right\rangle \equiv \left|\dfrac{v_m}{2},\dfrac{v_m}{2}\right), & v_m \text{ even}, \\ \left|N_a,\dfrac{v_m-1}{2}\right\rangle \otimes \left|N_c,\dfrac{v_m+1}{2}\right\rangle \equiv \left|\dfrac{v_m-1}{2},\dfrac{v_m+1}{2}\right), & v_m \text{ odd}. \end{cases} \tag{10}$$

Because the two vibrational modes are identical for H_2O and SO_2, so $N_a=N_c\equiv N$ and the exchange of the vibrational quantum number on each bond a and c gives the same result. The states $|\psi(0)\rangle_A$ and $|\psi(0)\rangle_B$ correspond, respectively, to a typical local mode and a normal mode for H_2O while both states are of typical normal mode for SO_2 [26]. It should be

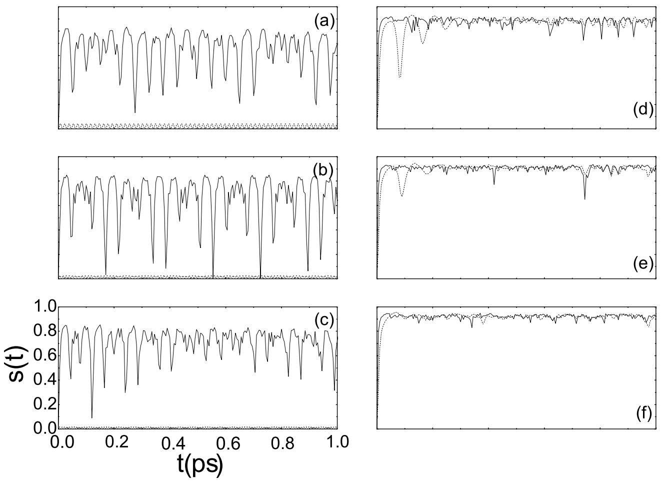

FIG. 1. The linear entropy $s(t)$ for H_2O (a), (b), (c), and SO_2 (d), (e), (f) with $v_m=15$, 20, and 25, respectively. Here two initial states are used: $|\psi(0)\rangle_A$ (dotted line) and $|\psi(0)\rangle_B$ (solid line).

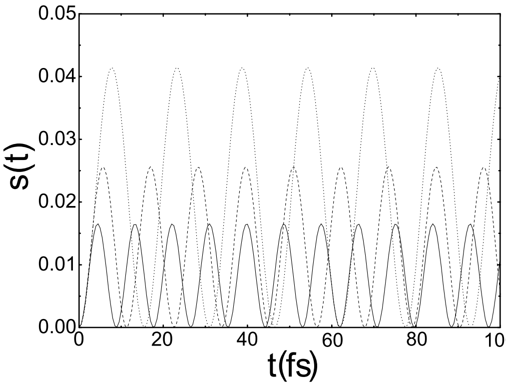

FIG. 2. The linear entropy $s(t)$ of initial state $|\psi(0)\rangle_A$ for H_2O with $v_m=15$ (dotted line), 20 (dashed line), and 25 (solid line).

mentioned that some important ideas were introduced for understanding the properties of local and normal modes, including dynamical tunnelling [27], locality parameter [28], polyad phase sphere [29], and susceptibility of eigenstates [30]. Here we explore the differences between those two typical states in dynamics of quantum entanglement.

With those initial states, Figs. 1(a)–1(c) for H_2O, 1(d)–1(f) for SO_2 show the entanglement dynamics described by the linear entropy, which is for $v_m=15$, 20, and 25, respectively. Some observations in Fig. 1 are as follows. For those two molecules, the increasing rate of the linear entropy in the early-time evolution of state $|\psi(0)\rangle_B$ is more rapid than that of state $|\psi(0)\rangle_A$, and the amplitude and the frequency of oscillations of the entropy of state $|\psi(0)\rangle_B$ are larger that those of state $|\psi(0)\rangle_A$. The larger the MQN v_m, the larger the difference of the entropy between those two states for H_2O, but the smaller for SO_2. The first maximal entropy of state $|\psi(0)\rangle_B$ for H_2O and both $|\psi(0)\rangle_A$ and $|\psi(0)\rangle_B$ for SO_2 increases as v_m increases, while that of state $|\psi(0)\rangle_A$ for H_2O decreases. Considering the local-mode or the normal-mode characters of those two initial states, we notice the main difference between the entropy of the typical local-mode and the typical normal-mode state, that is, the entropy of the typical local-mode state $|\psi(0)\rangle_A$ is better quasiperiodic than that of the typical normal-mode states $|\psi(0)\rangle_B$ for H_2O with the maximal entropy of local-mode states being smaller than that of normal-mode states. For those two states with normal-mode character that difference is small for SO_2.

Additionally, we still note that there is a significant property in the period and the shape of the entropy for state $|\psi(0)\rangle_A$ for H_2O, as shown in Fig. 2, that is, the evolution of the entropy is periodic and regular. Such a period is explained with the analytic form of the entropy for state $|\psi(0)\rangle_A$. In diagonalization of the algebraic Hamiltonian we note that for a given v_m, the contributions of $|0,v_m)$ and $|1,v_m-1)$ to the eigenenergy of $|0,v_m)$ are one order of magnitude larger than those of other states for H_2O. Thus we neglect the 2:2 resonance term with $m^{(2)}_{(a,c)}$ in Eq. (2) and consider the interaction between just two states $|0,v_m)$ and $|1,v_m-1)$. In this case we obtain the linear entropy of the initial state $|0,v_m)$, given by,

$$s(t)=\frac{2\gamma^2}{\Omega^2}\left[1-\frac{\gamma^2}{\Omega^2}\sin^2(\Omega t)\right]\sin^2(\Omega t), \tag{11}$$

where $\gamma=m^{(1)}_{(a,c)}\sqrt{v_m[1-(v_m-1)/N]}$ and $\Omega=\frac{1}{2}\sqrt{[H_0(0,v_m)-H_0(1,v_m-1)]^2+4\gamma^2}$, where $H_0(v_a,v_c)$ is the eigenvalue of $\hat{H}_0(v_a,v_c)$ on local base $|v_a,v_c)$. It is easy to get the period τ of the entropy from Eq. (11),

$$\tau=\frac{\pi}{\Omega}. \tag{12}$$

In this way the period given by Eq. (12) is 0.0163, 0.0119, and 0.0093 ps for H_2O with $v_m=15$, 20, and 25, respectively, which quite approximates to the corresponding period 0.0164, 0.0122, and 0.0096 ps obtained in Fig. 2. The amplitude of Eq. (11) indeed decreases as v_m increases for H_2O. It would be usable for a quantum information process since the state $|\psi(0)\rangle_A$ for H_2O is in entanglement but goes with disentanglement, because entanglement and disentanglement are both necessary for quantum computation.

It should be pointed out that Eq. (11) with $v_m=1$ is the exactly analytic form for those two molecules, which was employed to check the numerical method we used here. Those yield the same results in the case of $v_m=1$. In simulation of the entropy we found that the difference between Eq. (11) and the numerical results increase as v_m increases. That is because Eq. (11) is based on a simple model. For other initial states with larger v_m, it is probably impossible to relate the period to the parameters in Eq. (1) for each molecule since there are anharmonic interactions between stretches and many frequencies take part in the evolution.

B. Entanglement of coherent states

We now study the entanglement of continuous-variable-type states of H_2O and SO_2. Here we are interested in coherent states, and the discussion for other kinds of continuous-variable-type states is straightforward. The initial states are given by,

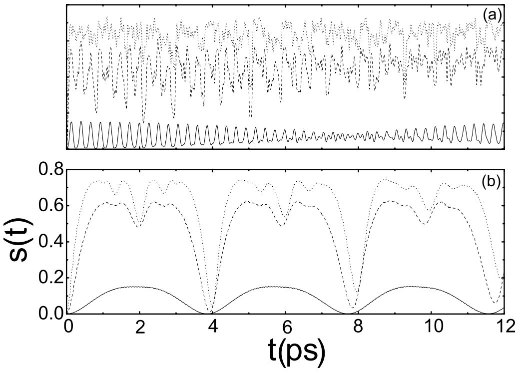

FIG. 3. The linear entropy $s(t)$ for H_2O (a) and SO_2 (b) with initial coherent states. Here the amplitude of coherent states is taken to be $\alpha=0.2$ (solid line), 1.0 (dashed line), and 1.8 (dotted line).

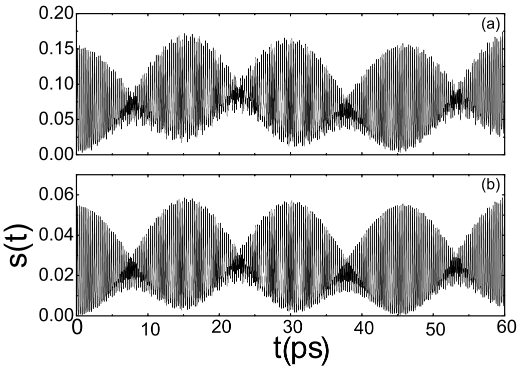

FIG. 4. The linear entropy $s(t)$ for H_2O in longer time evolution with initial coherent states. Here the amplitude of coherent states is 0.2 (a) and 0.1 (b).

$$|\psi(0)\rangle_{\alpha,\beta} = e^{\frac{-|\alpha|^2-|\beta|^2}{2}} \sum_m \frac{\alpha^m}{\sqrt{m!}}|N_a,m\rangle \otimes \sum_n \frac{\beta^n}{\sqrt{n!}}|N_c,n\rangle, \tag{13}$$

where α and β are the amplitude of the coherent states on bond a and c, respectively, and m (n) is the vibrational quantum number on bond a (c). The values of α and β can be taken to be any complex number. Here we let $\alpha=\beta$ be a real number. In simulation of the entropy we truncated the set of Fock states that compose the Poissonian distribution at some n_t with $\sum_{n=0}^{n_t} e^{-\alpha^2}\alpha^{2n}/n! \simeq 1$. The accuracy of the results obtained was tested by adding more Fock states to that distribution to see whether the calculated values changed.

Figure 3 shows the linear entropy of initial coherent states, where the amplitude α of coherent states is taken to be 0.2, 1.0, and 1.8, as an example. The maximal entropy increases as the amplitude α increases for those two molecules. The frequency of oscillation of the entropy for H_2O is larger than that for SO_2. The oscillation of the entropy in the cases of $\alpha=1.0$ and 1.8 for H_2O is irregular while that for SO_2 is quasiperiodic. For smaller amplitude $\alpha=0.2$, the oscillation of the entropy is periodic for SO_2 while that exhibits quasiperiodic in the early-time evolution for H_2O. To see that more clearly, Fig. 4 shows the entropy in longer time evolution for $\alpha=0.2$, where the similar thing for $\alpha=0.1$ is plotted for comparison. It is nicely observed that "classical" beat phenomena of the entropy of the initial coherent states happens for H_2O with the similar shape for $\alpha=0.2$ and 0.1, as in classical physics and in the well-known Jaynes-Cummings model [31,32]. In Fig. 4 the quasiperiod of beat is far longer than that of early-time evolution of the entanglement. The later period will be discussed below. The beat phenomenon of entanglement has been very recently observed for two distinguishable qubits in a one-dimensional harmonic trap [33]. The beat can be used as the carrier of information. Hence the long period of beat is necessary for quantum information processing. The period for SO_2 and quasiperiod for H_2O are already explained in terms of the analytic form of the entropy for the coherent state with a small amplitude α.

In the case of a small amplitude α, the initial coherent state can be approximately taken to be,

$$|\psi(0)\rangle_{\alpha,\alpha} \cong P(0,0)|0,0) + P(1,0)|1,0) + P(0,1)|0,1) + P(2,0) \times|2,0) + P(0,2)|0,2) + P(1,1)|1,1), \tag{14}$$

where $P(m,n)=e^{-\alpha^2}\alpha^{(m+n)}/\sqrt{m!\,n!}$, $m(n)=0$, 1, and 2 is the vibrational quantum number on bond a (c). We again neglect the 2:2 resonance term with $m_{(a,c)}^{(2)}$ in Eq. (2) and consider the 1:1 resonance interaction. Thus, due to $P(0,2)=P(2,0)$, that state evolves as follows,

$$\begin{aligned}|\psi(t)\rangle_{\alpha,\alpha} = {} & P(0,0)|0,0) + e^{-iH_0(1,0)t}\{[P(1,0)\cos(m_{(a,c)}^{(1)}t) - iP(0,1)\sin(m_{(a,c)}^{(1)}t)]|1,0) \\ & + [P(0,1)\cos(m_{(a,c)}^{(1)}t) - iP(1,0)\sin(m_{(a,c)}^{(1)}t)]|0,1)\} + e^{-i\sigma_1 t}\left\{\left[P(2,0)\cos(\omega t) - i\frac{P(2,0)\sigma_2 + P(1,1)\lambda}{\omega}\sin(\omega t)\right][|2,0) \right. \\ & \left. + |0,2)] + \left[P(1,1)\cos(\omega t) - i\frac{2P(2,0)\lambda - P(1,1)\sigma_2}{\omega}\sin(\omega t)\right]|1,1)\right\},\end{aligned} \tag{15}$$

where $\sigma_1=[H_0(2,0)+H_0(1,1)]/2$, $\sigma_2=[H_0(2,0)-H_0(1,1)]/2$, $\lambda=m_{(a,c)}^{(1)}\sqrt{2(1-1/N)}$, and $\omega=\sqrt{2\lambda^2+\sigma_2^2}$. Therefore, the linear entropy $s(t)$ of that state can be expressed in analytic form, which is too lengthy to include here. For $\alpha=0.2$, the (quasi-)period of the entropy by Eq. (15) is 0.278 and 3.99 ps for H_2O and SO_2, respectively, which is quite close to the corresponding period 0.281 and 4.07 ps obtained in Fig. 3. For $\alpha=0.1$, the nearly same period as above is obtained analytically and numerically for H_2O. However, there is a larger difference in the amplitude and the beat frequency of the entropy between the analytical and numerical calculation although that amplitude in both cases decreases as α decreases. That is because the analytical calculation is performed with a simplified model.

It is worthwhile emphasizing that the long period of entanglement of coherent states with small amplitude for SO_2 and the longer period of beat in entanglement for H_2O mean that the entanglement lasts for a long time. The entanglement as a physical resource is available on the condition that the entanglement could sustain long enough so that some processing can be performed. For example, the entanglement between the atom and the cavity field must survive long enough so that it can be transferred to a next atom via coherent interaction [34]. The increasingly long period of entanglement in a generalized Jaynes-Cummings model can be realized by increasing the detuning parameter [35]. Therefore the long time sustained entanglement for H_2O and SO_2 would be applied to quantum information and quantum computation.

IV. CONCLUSION AND DISCUSSION

We have investigated dynamical entanglement in triatomic molecules H_2O and SO_2 in u(2) algebraic model [15] with various MQN and initial states, where the entanglement is measured by the linear entropy. It is shown that the maximal entropy of an initial state with the local-mode character is smaller than that of one with the normal-mode character. The entropy of typical local-mode state $|\psi(0)\rangle_A$ in H_2O exhibits a better period that has been explained with the simplest model based on two energy levels. That implies that two states $|0,v_m\rangle$ and $|1,v_m-1\rangle$ in H_2O could be reasonable candidates for a qubit in quantum communication. For initial state $|\psi(0)\rangle_B$ with normal-mode character, the linear entropy for those two molecules is irregular with the maximal entropy increasing as MQN increases. Those behaviors can be taken as dynamical signature to differ the local-mode from the normal-mode state. For initial coherent states with a small amplitude, it is shown that the periodicity of the entropy appears in SO_2 and classical beat phenomena of the entropy happens in H_2O, and that the maximal entropy of the coherent states increases as the amplitude of coherent states increases. The (quasi-)period of the entropy of $|\psi(0)\rangle_A$ in H_2O and coherent states with a small amplitude in H_2O and SO_2 has been explained with a simplified model based on a few energy levels. The long periods of entanglement of coherent states with small amplitude for H_2O and SO_2 show that the entanglement sustains long enough so that some task could be accomplished. For the stretching vibrations of H_2O and SO_2, the quantum state with large dynamical entanglement can be prepared by a typical normal-mode state or by coherent states with a large amplitude. We believe that those are useful in quantum information processing and quantum computing based on vibrational states in triatomic molecules H_2O and SO_2.

It is possible to study entanglement dynamics and entanglement transfer in other polyatomic molecules, especially to explore the influence of classical bifurcations or chaos, which occur frequently in highly excited molecules, on quantum entanglement, and part of the results will be discussed elsewhere [36]. It is desirable to investigate decoherence for molecules due to coupling between vibrational and other degrees of freedom (i.e., electronic or rational). It is worth to explore the relation between dynamics of quantum entanglement and quantum dynamics of localized excitations in molecules.

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