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Entropy correlation and entanglement for mixed states in an algebraic model

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Abstract

As an alternative with potential connections to actual experiments, other than the systems more usually used in the field of entanglement, the dynamics of entropy correlation and entanglement between two anharmonic vibrations in a well-established algebraic model, with parameters extracted from fitting to highly excited spectral experimental results for molecules H₂O and SO₂, is studied in terms of the linear entropy and two negativities for various initial states that are respectively taken to be the mixed density matrices of thermal states and squeezed states on each mode. For a suitable parameter in initial states the entropies in two stretches can show positive correlation or anti-correlation. And the linear entropy of each mode is positively correlated with the negativities just for the mixed-squeezed states with small parameters in H₂O while they do not display any correlation in other cases. For the mixed-squeezed states the negativities exhibit dominantly positive correlations with an effective mutual entropy. The differences in the linear entropy and the negativities between H₂O and SO₂ are discussed as well. Those are useful for molecular quantum computing and quantum information processing.

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1. Introduction

Entanglement is a quantum-mechanical feature that can be used for many fascinating applications including teleportation [1], quantum cryptography [2] and quantum computation [3]. To quantify the degree to which a state is entangled is a central task for quantum information processing and, correspondingly, several measures of it have been introduced.

Those are entanglement of formation [4], entanglement of distillation [5], relative entropy of entanglement [6], linear entropy of entanglement [7], concurrence [8], negativity [9, 10] and so forth. Those measures of entanglement have motivated considerable research. One of such researches is to characterize the properties of entangled states [11–14] and, consequently, some important ideas such as collapse and revival of entanglement in two interacting qubits [12], entanglement reciprocation between qubits and continuous variables [13], and entanglement transfer from continuous variables to multiple qubits [14] have been proposed, where continuous-variable-type entangled states are considered. Since all bipartite entangled states are useful for information processing [15], it is still interesting to investigate entanglement in a bipartite system.

Another research is to explore the correlation between one measure of entanglement and other quantities that are easier to compute [16–18]. A relationship between negativity and uncertainty products has been established for asymmetric two-mode Gaussian states [17]. For a mixed state anti-correlation is observed between the entanglement of formation and participation ratio, whereas a strong correlation is observed between the entanglement of formation and negativity [18]. In addition, calculation of a more tractable measure can allow one to at least work out the correct ordering for other measures [19–21]. However, those investigations [16–21] concentrate on static properties of entanglement. We are here interested in the negativity as a measure of entanglement for its simplicity as well as its wide applicability in order to discuss its dynamical correlation with quantum mutual entropy [26].

The purpose of this paper is twofold. On the one hand, we shall study the dynamics of entropy correlation and entanglement between vibrations in molecules H_2O and SO_2 , where molecular parameters have been optimized from experimental spectra by Iachello and coworkers [22]. The entropy in each vibration is described by the linear entropy [7] as it is related to purity. Two negativities are employed to measure the entanglement with two kinds of initial states, which are taken to be the mixed density matrices of thermal states and squeezed states on each vibrational mode, respectively. Two cases of the entropy correlation, in which partial entropies fluctuate together in time and they are anti-correlated, happen for a suitably chosen state. An effective mutual entropy that is defined as the sum of linear entropies in two modes shows strong correlation with the negativities for the mixed-squeezed states. The differences between H_2O and SO_2 in the entropy correlation and the negativities are still noted. On the other hand, recent theoretical studies have highlighted the feasibility of using molecular vibrational states for quantum computing [23–25]. It is shown that extremely high quantum gate fidelity can be achieved for a qubit system based on vibrational eigenstates of diatomic molecules [24], and the interplay of the anharmonicity and the coupling is of prime importance in quantum computing based on vibrational qubits [25]. Therefore, the dynamical properties of both entropy correlation and entanglement in realistic molecules H_2O and SO_2 may be useful for molecular quantum computing.

The paper is organized as follows. Section 2 presents an algebraic Hamiltonian for highly excited vibrational spectra in molecules H_2O and SO_2 . Section 3 studies the dynamics of entropy correlation and entanglement for initial mixed thermal states and squeezed states, where the entropy of each mode and the entanglement are described in terms of the linear entropy and two negativities, respectively. The mutual entropy of those states is discussed as well. Last section concludes the paper with discussions.

2. Algebraic Hamiltonian

There are various algebraic schemes for molecular systems in the literature [22, 27–31], in which $U(4)$ and $U(2)$ algebras are extensively used [22, 29–31]. The $U(4)$ algebraic model

becomes quite complicated when the number of atoms in a molecule is larger than four, while the $U(2)$ algebraic model is particularly well suited for dealing with highly excited vibrations of polyatomic molecules. Since anharmonic vibrations are usually described by Morse oscillators that can be realized in terms of the $U(2)$ algebra, the $U(2)$ force-field expansion has a much faster convergence than the conventional force-field expansion [29]. Furthermore, a simple scheme is proposed to extract the corresponding classical dynamics from the $U(2)$ model parameters [22]. We are interested in the $U(2)$ algebraic model [22] for XY-stretching vibrations in a symmetrical triatomic molecule XY_2 , where the YY-bending mode is not taken into account since the bending fundamental is about half the wave number of the stretching fundamental. More importantly, such model with a few parameters has accurately described the highly excited stretching vibrations in the interested molecules. The algebraic Hamiltonian [22] reads

$$\hat{H} = \hat{H}_0(v_b, v_c) + \hat{H}_i, \quad (1)$$

where $\hat{H}_0(v_b, v_c)$ and \hat{H}_i are respectively diagonal and non-diagonal Hamiltonians with $v_{b(c)}$ representing the vibrational quantum number on the XY-stretch bond $b(c)$, which are given by

$$\begin{aligned} \hat{H}_0(v_b, v_c) = & a_{(b,c)}^{(1,0)}(\hat{C}_b + \hat{C}_c) + a_{(b,c)}^{(2,0)}(\hat{C}_b^2 + \hat{C}_c^2) + 2a_{(b,c)}^{(1,1)}\hat{C}_b\hat{C}_c \\ & + a_{(b,c)}^{(3,0)}(\hat{C}_b^3 + \hat{C}_c^3) + a_{(b,c)}^{(2,1)}\hat{C}_b\hat{C}_c(\hat{C}_b + \hat{C}_c), \end{aligned} \quad (2)$$

$$\hat{H}_i = m_{(b,c)}^{(1)}(\hat{f}_b^+\hat{f}_c^- + \hat{f}_b^-\hat{f}_c^+) + m_{(b,c)}^{(2)}(\hat{f}_b^+\hat{f}_b^+\hat{f}_c^-\hat{f}_c^- + \hat{f}_b^-\hat{f}_b^-\hat{f}_c^+\hat{f}_c^+), \quad (3)$$

where $a_{(b,c)}^{(j,k)}$ and $m_{(b,c)}^{(j)}$ ($j = 1, 2, 3$ and $k = 0, 1$) are parameters, the subscripts b and c denote the two equivalent XY-stretch vibrations, and \hat{C}_i and \hat{f}_i^\pm ($i = b, c$) are $U(2)$ operators with the matrix elements being

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

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

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

where $|N_i, v_i\rangle$ 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 [31], and v_i varies from 0 to N_i without dissociating the bond. If N_i is taken to be infinite, equation (4) becomes the matrix elements of the usual harmonic oscillator with equations (5) and (6) being the corresponding ones of its creation and annihilation operators, respectively. Fitting highly excited spectra of H_2O and SO_2 in experiments, Iachello and coworkers have presented the optimized parameters in the model in table I of [22] with $N_b = N_c = 50$ for H_2O and 170 for SO_2 , where the classical dynamics of the classical counterpart of equation (1) has been analyzed. The extension of the $U(2)$ algebraic model to incorporate bending modes with possible interactions between vibrational modes [30] becomes a useful model for the description of the highly excited vibrational spectra of polyatomic molecules. Details for algebraic methods are referred to [31].

3. Dynamics of entropy correlation and entanglement

We investigate the dynamics of both entropy correlation and entanglement between vibrational modes of H₂O and SO₂ in the model equation (1). Two kinds of initial mixed states, thermal states and squeezed states, are used to inspect the time evolution of entropy correlation and entanglement. The importance of studying them in different initial states is twofold. 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. Continuous-variable-type entangled states including squeezed states and thermal states have been widely applied to quantum teleportation [32], quantum cryptography [33] and quantum computation [34]. The mixed density matrices of initial states of the full system is taken to be

$$\rho_{bc}(0) = \rho_b(0) \otimes \rho_c(0), \quad (7)$$

where $\rho_i(0) = \sum_{v_i=0}^{N_i} P_A(v_i, \bar{v}_i) |N_i, v_i\rangle \langle N_i, v_i|$ is the initial density matrix of mode i . The subscripts $A = T$ and S respectively represent the thermal states and the squeezed states on mode i with the coefficients being [35]

$$P_T(v_i, \bar{v}_i) = \frac{1}{1 + \bar{v}_i} \left[\frac{\bar{v}_i}{1 + \bar{v}_i} \right]^{v_i} \quad (8)$$

$$P_S(v_i, \bar{v}_i) = \begin{cases} \frac{v_i!}{[(v_i/2)!]^2 \cosh r} \left[\frac{\tanh r}{2} \right]^{v_i}, & v_i \text{ even,} \\ 0, & v_i \text{ odd,} \end{cases} \quad (9)$$

where $r = \text{arcsinh} \sqrt{\bar{v}_i}$, and \bar{v}_i is the average quantum number on mode i , which will be taken as a parameter in what follows. In simulation of entropy correlation and entanglement we truncated the set of Fock states that compose the corresponding distribution at some v_i with $\sum_{v_i=0}^{v_i} P_A(v_i, \bar{v}_i) \simeq 1$. The accuracy of the results obtained is further tested by adding more Fock states to that distribution to see whether the calculated values are changed. Now that the initial density matrices and the quantum Hamiltonian equation (1) are at hand, we are able to investigate dynamics of both entropy correlation and entanglement in H₂O and SO₂.

3.1. Entropy correlation

We now study the entropy of each stretching mode in order to explore the behaviors of entropy correlation between two stretches in H₂O and SO₂. The entropy of mode $b(c)$ is given by the linear entropy [7]:

$$S_{b(c)}(t) = 1 - \text{Tr}[\rho_{b(c)}(t)]^2, \quad (10)$$

where $\rho_{b(c)}(t) = \text{Tr}_{c(b)} \rho_{bc}(t)$ is the reduced-density matrix with $\rho_{bc}(t)$ being the full density one, which evolves in time with the Liouville equation,

$$\dot{\rho}_{bc}(t) = -i[\hat{H}, \rho_{bc}(t)]. \quad (11)$$

The purity of mode $b(c)$ is given by $\text{Tr}[\rho_{b(c)}(t)]^2$. Thus, an increase in the linear entropy is parallel to a decrease in purity. The entropy of mode $b(c)$ can be described in terms of the von Neumann entropy, $-k_B \text{Tr}[\rho_{b(c)}(t) \ln \rho_{b(c)}(t)]$. Both entropies have the same trend for a bipartite system [36] so that the results of the linear entropy are presented below. In the calculation of the entropy, we have used many sets of parameters (\bar{v}_b, \bar{v}_c). We would like to discuss in detail the results for two sets of parameters as examples.

Figures 1 and 2 respectively show the partial entropy changes (PEC) ($\Delta S(t) = S(t) - S(0)$) of each stretching mode in H₂O and SO₂ with initial mixed thermal states (MTS) and

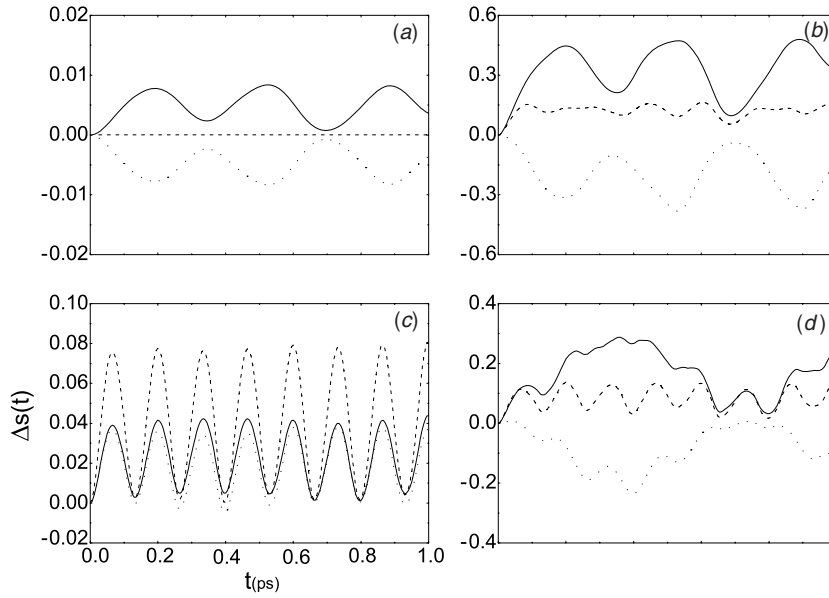


Figure 1. Linear entropy changes $\Delta S(t) = S(t) - S(0)$ of modes b (the solid line) and c (the dotted line) and the effective mutual entropy (the dashed line) in H_2O . Here two kinds of initial states are used: (a) and (b) for the mixed thermal states with parameters $(\bar{\nu}_b, \bar{\nu}_c) = (0.21, 0.20)$ and $(0.80, 0.01)$, respectively; (c) and (d) for the mixed-squeezed states with same parameters as the corresponding (a) and (b).

mixed-squeezed states (MSS) given by equations (7)–(9), where two sets of parameters $(\bar{\nu}_b, \bar{\nu}_c)$ are taken to be $(0.21, 0.20)$ and $(0.80, 0.01)$. For both kinds of states with small parameters $(\bar{\nu}_b, \bar{\nu}_c) = (0.21, 0.20)$, the PEC of modes b and c in both molecules are regular with the period in H_2O being larger than that in SO_2 . In that case, it is easy to note that the PEC of MTS in figures 1 and 2(a) are anti-correlated while those of MSS in figures 1 and 2(c) are positively correlated. As the difference between parameters $\bar{\nu}_b$ and $\bar{\nu}_c$ increases in the case of $(\bar{\nu}_b, \bar{\nu}_c) = (0.80, 0.01)$, the PEC of two modes become anti-correlated for both states with the magnitude of PEC increasing, where the PEC oscillations of MSS are irregular, as shown in figures 1 and 2(b), (d). Once $\bar{\nu}_b$ is set to be equal to $\bar{\nu}_c$, we find that the PEC of the two modes are indeed identical at all times. We performed the similar simulation with other parameters and found that the larger the difference between the parameters $\bar{\nu}_b$ and $\bar{\nu}_c$, the more distinct the anti-correlations of entropies. For anti-correlations MTS has an advantage over MSS since a smaller difference between $\bar{\nu}_b$ and $\bar{\nu}_c$ is needed, as shown in figures 1 and 2(a). The positively correlated or anti-correlated behaviors of entropies offer a possibility of complement quantum computing and quantum information processing.

The entropy correlations of two modes can be quantitatively measured by the mutual entropy that indicates the entropy shared between the two subsystems. It should be mentioned that in classical information theory the mutual entropy is a very important quantity to analyze the communication processes and physical transformations. Similarly, the quantum mutual entropy is of importance in quantum information processing [26, 36] as well as in the study of the physics in general many-body systems [37].

The mutual entropy is defined by [26]

$$S_m(t) = S_b(t) + S_c(t) - S_{bc}(t), \quad (12)$$

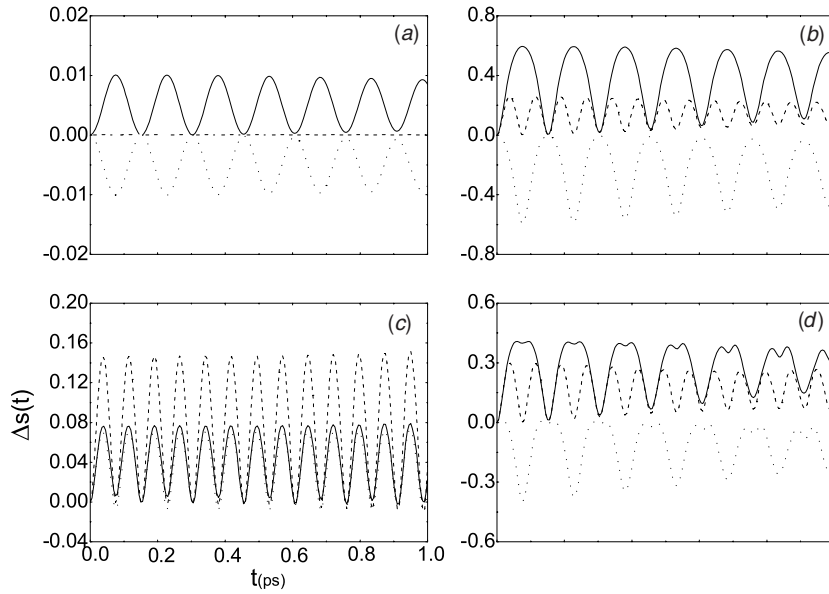


Figure 2. Same as in figure 1 but here is for SO₂.

where S_b and S_c are the linear entropy of modes b and c given by equation (10), respectively, and S_{bc} is the linear entropy of the full system. The latter is constant for the Hamiltonian evolution governed by equation (1). Thus one can take the sum of PEC of two modes as an effective mutual entropy (EME), which is plotted as the dashed curve in figures 1 and 2. EME is very small in the magnitude of 10^{-5} in figures 1 and 2(a) and it is irregular in figure 1(b) for MTS in H₂O. It is quasi-periodic as seen by its amplitude of fluctuations in figures 1(c), (d) and figure 2, which is significantly smaller than that of each PEC in anti-correlated cases. However, in positively correlated cases as shown in figures 1 and 2(c) its amplitude is larger than that of each PEC, where EME and PEC have the same period. In the following subsection, EME will be further discussed with the entanglement measured by the negative eigenvalues of the partially transposed density matrix.

3.2. Entanglement

We now investigate the entanglement of MTS and MSS in H₂O and SO₂. There are various measures of entanglement mentioned in the introduction. Although a particular measure of entanglement can be analytically determined in some cases, most entanglement measures are very difficult to calculate in general [19]. Here we are interested in the negativity as a measure of entanglement since it can be computed efficiently for any mixed states of an arbitrary bipartite system.

The negativity of a state $\rho_{bc}(t)$ is simply defined by [9]

$$N(t) = \max \{0, -\mu_{\min}\}, \tag{13}$$

where μ_{\min} is the minimum of the eigenvalues of the partial transpose of state $\rho_{bc}(t)$. Partial transposition is a blockwise transposition of a matrix, which is given by

$$\rho_{i\alpha, j\beta}^{T_2}(t) \equiv \rho_{i\beta, j\alpha}(t), \tag{14}$$

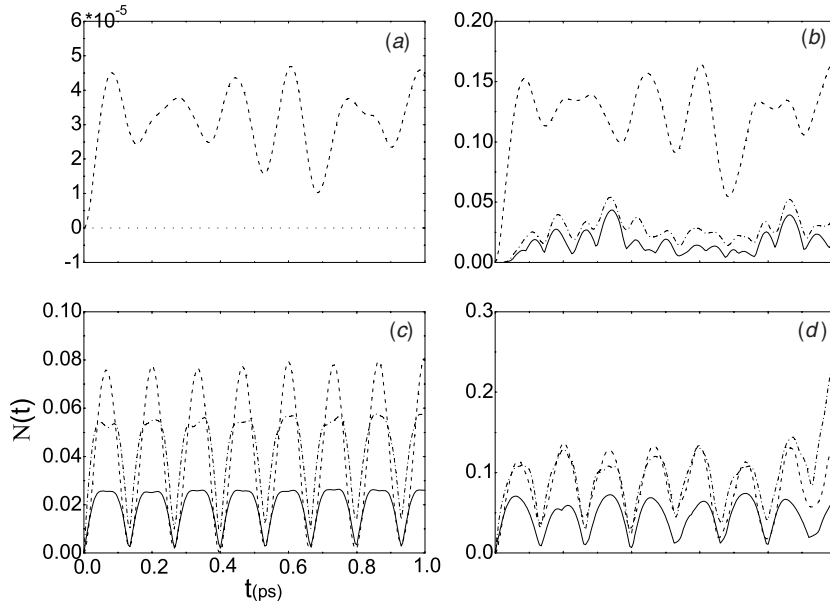


Figure 3. Two negativities $N(t)$ (the solid line) and $N_s(t)$ (the dot-dashed line) and the effective mutual entropy (the dashed line) in figure 1 for H_2O , where the same initial states as in figure 1 are used.

where T_2 is the partial transposition for the second subsystem. The negativity is also defined by [38]

$$N_s(t) = \max\{0, -\lambda_{\text{neg}}\}, \tag{15}$$

where λ_{neg} is the sum of the negative eigenvalues of $\rho_{i\alpha, j\beta}^{T_2}(t)$. Note that $N_s(t)$ corresponds to the negativity defined by $(\|\rho^{T_2}\| - 1)/2$, and its closely related cousin, the logarithmic negativity defined by $\log_2\|\rho^{T_2}\|$, where $\|\rho^{T_2}\|$ denotes the trace norm of ρ^{T_2} [10]. Negativity, $N_s(t)$, serves as an upper bound of quantum teleportation capacity, and the logarithmic negativity is an upper bound to distillable entanglement [10, 39]. How to use the properties of negativity for quantum teleportation and distillable entanglement will be left to discuss in the future.

Figures 3 and 4 respectively show both negativities for H_2O and SO_2 with the same states as figures 1 and 2, where EME is also plotted with a dashed curve for comparison. Some observations in figures 3 and 4 are in order.

First, comparing figures 3 and 4 with figures 1 and 2, we notice that both negativities differ from the linear entropies on their oscillatory behaviors, but they have the same period of oscillation for MSS with small parameters $(\bar{v}_b, \bar{v}_c) = (0.21, 0.20)$ in H_2O , as shown in figure 1(c) and figure 3(c).

Second, it is observed that the two negativities are almost zero in figures 3 and 4(a) for MTS with parameters $(\bar{v}_b, \bar{v}_c) = (0.21, 0.20)$ where the difference between \bar{v}_b and \bar{v}_c is small because the system is symmetric about modes b and c . The two negativities increase a little with the time evolution in figure 4(b) for SO_2 . From figures 3 and 4 it is easy to note that for the same parameters in initial states their magnitude of MTS is smaller than that of MSS. That implies that the larger negativities can be obtained with MSS. In addition, the two negativities are identical both in their minimal values and in oscillation, where the magnitude of $N(t)$ (the

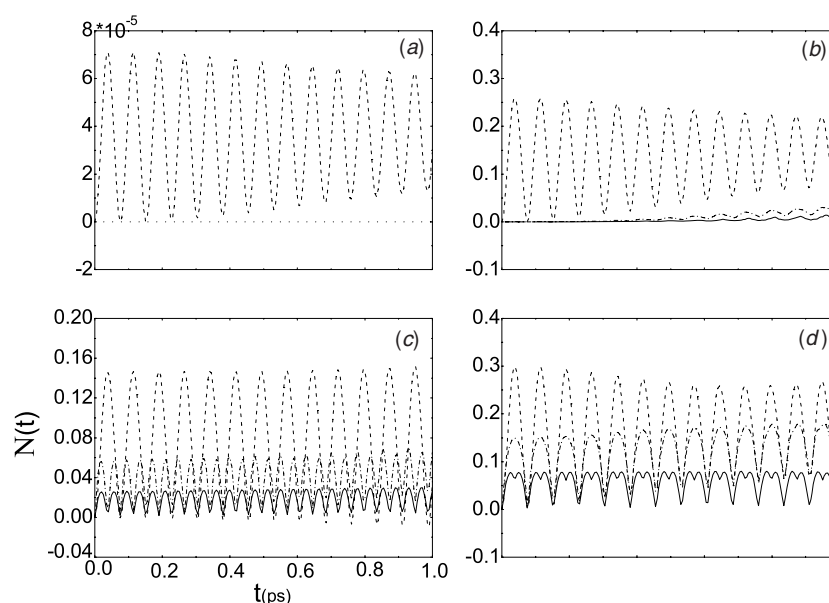


Figure 4. Same as in figure 3 but here is for SO_2 .

solid curve) is smaller than that of $N_s(t)$ (the dot-dashed curve). That indicates that the two negativities are indeed a good measure of entanglement.

Third, we compare both negativities with EME. For MTS the magnitude of EEM is much larger than that of the negativities, where the oscillation of the negativities is much irregular than that of EME, as seen in figures 3 and 4(b). For MSS in H_2O the negativity $N_s(t)$ is better identical with EME than the negativity $N(t)$ in both magnitude and period of oscillation while that keeps in part identical for SO_2 . That means that EME just for MSS can be regarded as a measure of entanglement. Such identity remains for MSS with other parameters. It is interesting to note that the shape of EME for two kinds of states with two sets of parameters is similar since the ground state and several lower excited states are always dominant in the initial mixed states.

Finally, two interesting differences in the negativities between H_2O and SO_2 are noted. As shown in figures 3 and 4(b), in the early-time evolution the negativities of MTS in H_2O are much larger than those in SO_2 . The quasi-periodicity of both negativities happens for MSS while that disappears for MTS, where the quasi-period in H_2O is larger than that in SO_2 . Those differences can be regarded as dynamical fingerprints of molecular intrinsic property, since H_2O and SO_2 are respectively the local- and normal-mode molecules in nature [22].

It should be pointed out that the entanglement properties of pure Fock and coherent states were discussed for two interacting modes in a pure theoretical model [11], where the entanglement was measured by the linear entropy and the von Neumann entropy. Since pure states are difficultly realized in experiments, we have considered two kinds of mixed states and studied the dynamics of the linear entropy correlation and two negativities for two stretching modes in a well-established algebraic model with parameters extracted from fitting to highly excited spectral experimental results for H_2O and SO_2 . That represents a nice alternative with potential connections to actual experiments, other than the systems usually used in the field of entanglement. The above-studied correlations among the linear entropy, the mutual entropy

and negativities in molecules H₂O and SO₂ are helpful in further understanding of molecular vibrations on the dynamical aspects of entanglement as well as to processing of molecular quantum information.

4. Conclusion and discussion

We have explored the dynamical properties of entropy correlation and entanglement of two anharmonic stretches in molecules H₂O and SO₂ [22] with various initial mixed states, where the entropy and the entanglement are respectively measured by the linear entropy and two negativities. The initial states are taken to be the mixed density matrices of thermal states and squeezed states on each vibrational mode. It is shown that positively correlated or anti-correlated behaviors of the linear entropies in each mode depend on initial states. For MSS with a small difference between parameters $\bar{\nu}_b$ and $\bar{\nu}_c$, the behaviors of linear entropies are positively correlated. For MTS and MSS with a large difference between the parameters $\bar{\nu}_b$ and $\bar{\nu}_c$, two linear entropies are anti-correlated.

It is still demonstrated that the negativities and the linear entropy of each mode are positively correlated only for MSS with small parameters ($\bar{\nu}_b, \bar{\nu}_c$) in H₂O while they do not exhibit any correlated behavior in other cases. In addition, for MSS in both molecules, H₂O and SO₂, it is found that the negativities are dominant in positive correlations with EME. Thus EME could be regarded as a measure of entanglement just for MSS. Any correlation between the linear entropy and the negativities was not found for MTS in both molecules. Both negativities of MTS with large difference between $\bar{\nu}_b$ and $\bar{\nu}_c$ in H₂O are much larger than those in SO₂ in the early-time evolution, and the quasi-period of both negativities for MSS in H₂O is larger than that in SO₂. That can be taken as dynamical signatures of their corresponding intrinsically local- and normal-mode property. We believe that those are useful in quantum computing based on vibrational states in molecules H₂O and SO₂ since using vibrational states of diatomic molecules for quantum computing [23, 24] is feasible.

It is desirable to employ those properties of entropy correlation and entanglement for molecular quantum computing. It is possible to investigate entanglement quantified by other measures in polyatomic molecules. It is worth studying other quantity correlations in molecular systems.

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