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Quantum dynamics in double-well system with polychromatic perturbation using entangled trajectory molecular dynamics method

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Abstract. We investigate quantum dynamics in a double-well system subject to time-dependent perturbation with some incommensurate frequencies using entangled trajectory molecular dynamics method. We first compare time evolution of energy of the driven system with two mode perturbation under classical dynamics with one under quantum dynamics, it is found that quantum dynamics obeys Ehrenfest's theorem and it shows our results obtain from entangled trajectory molecular dynamics method are correct and accurate. Quantum coherence suppresses trajectory which initial energy is higher than barrier to get over it, this process is named coherent destruction of tunneling (CDT). We show this interesting phenomenon through showing entangled trajectory and corresponding classical trajectory in phase space and discuss their energy fluctuation with time. Furthermore, we discuss quantum dynamics with different frequency mode perturbation from the perspective of autocorrelation evolution, classical chaos brings remarkable influence on quantum-mechanical phenomena.

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Key words: Entangled trajectory molecular dynamics method, Coherent destruction of tunneling, Classical chaos.

1 Introduction

Quantum dynamics process in a double-well system with a periodically oscillating external field has been one of the subjects of long-lasting interest in diverse branches of physics and chemistry [1]. In particular, the coherent destruction of tunneling (CDT) which is an interesting phenomenon originally discovered by Grossmann has attracted

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much attention [2–4]. CDT phenomenon displays coherent control of quantum dynamics via a periodically oscillating external field, it has potential application in quantum motor and quantum-information processing and is very useful to understand much fundamental time-dependent processes [5, 6]. Even weak chaos can bring huge effect on quantum dynamics, Igarashi investigated classical chaos effect on dynamical behavior in driven quantum double-well systems numerically and showed chaos-assisted tunneling [7–9]. Recently, we study chaos-assisted tunneling of driven double-well system uses a new method named entangled trajectory molecular dynamics method (ETMD) and show this quantum phenomenon vividly with the help of entangled trajectories in phase space [10].

Entangled trajectory molecular dynamics method is an effective improvement to classical dynamics molecular method [10,13–18], it includes quantum effect during dynamics process at the cost of statistical independence of trajectories. ETMD not only can be used as a numerical method to solve the quantum Liouville equation, but also can show quantum underline dynamics process through showing entangled trajectories in phase space. A lot of works show ETMD method is an effective tool to deal with quantum dynamics of small molecular system, such as: capturing quantum tunneling effect [13–15, 19, 20], calculating photodissociation crosssection of water in its first absorption band [16], and autocorrelation function [21]. Most of these works used ETMD method used Wigner function which is well-known quantum phase space distribution function can be negative even the initial is positive anywhere to describe the evolution of the system. ETMD needs a positive definited ansatz approximation to fit the relationship between trajectories and distribution function, this approximation makes ETMD method based on Wigner presentation miss some important information about quantum dynamics, such as can not describe quantum interference phenomenon accurately [22]. For avoiding it, we use Husimi function which is positive anywhere to study quantum dynamics process in double-well system with polychromatic perturbation.

In the present paper, we study quantum dynamical behaviors in double well system with polychromatic perturbation using ETMD method based on Husimi presentation. Different from other similar works, we not only give some numerical calculation results, but also show quantum underline dynamics process through showing entangled trajectories in phase space. Firstly, we indirectly demonstrate ETMD method is a correct and accurate tool to deal with quantum driven system through checking the evolution of energy, the accord with Ehrenfest's theorem shows our numerical calculation is reasonable and precisely. The core of this paper is show coherent destruction of tunneling phenomenon with entangled trajectory and compare to classical one, we show the evolution of quantum coherence in this interesting case. At the end, we discuss quantum dynamics with different frequency mode perturbation from the perspective of autocorrelation evolution, the classical dynamics of some frequency mode perturbation shows chaotic behavior, chaos brings remarkable influence on quantum-mechanical phenomena.

The structure of this paper is as follows. In Section 2, we describe the basic theoretical formalism of the entangled trajectory molecular dynamics method based on Husimi representation and the model of double-well system with polychromatic perturbation. In

Section 3, we present numerical results of the quantum dynamics to show the coherent destruction of tunneling phenomenon, a visual physical picture of entangled trajectories and corresponding classical ones is showed together with their energy evolution to present the underline dynamics process. Furthermore quantum dynamics under different mode perturbation is presented in the view of autocorrelation. Some conclusions are drawn in Section 4. Atomic units are used throughout the paper unless noted and $\hbar=1$ is used in our calculations.

2 Theory

Quantum probability distribution function can be used to describe the state of quantum system instead of wavefunction, this description is correct and complete. We use Husimi function ρ^H to describe the evolution of system. Husimi function is positive definited and can be got by smoothing Wigner function ρ^w (a well-known quantum probability distribution function with negative values) with diffusion operator [11,12]:

$$\rho^H(q,p;t)=S_qS_p\rho^w(q,p;t), \tag{1}$$

where

$$S_q=\exp\left(\frac{1}{2}\sigma_q^2\frac{\partial^2}{\partial^2 q}\right),\ S_p=\exp\left(\frac{1}{2}\sigma_p^2\frac{\partial^2}{\partial^2 p}\right). \tag{2}$$

The parameters σ_q and σ_p is chosen to satisfy the minimum uncertainty condition $\sigma_q\sigma_p=\frac{\hbar}{2}$. The inverse operations for this sharpening are similarly defined:

$$S_q^{-1}=\exp\left(-\frac{1}{2}\sigma_q^2\frac{\partial^2}{\partial^2 q}\right),\ S_p^{-1}=\exp\left(-\frac{1}{2}\sigma_p^2\frac{\partial^2}{\partial^2 p}\right). \tag{3}$$

Then quantum Liouville equation based on Husimi representation is given:

$$\frac{\partial\rho^H}{\partial t}=-\frac{1}{m}(S_ppS_p^{-1})\frac{\partial\rho^H}{\partial q}+(S_qV'(q)S_q^{-1})\frac{\partial\rho^H}{\partial p}-\frac{\hbar^2}{24}(S_qV'''(q)S_q^{-1})\frac{\partial^3\rho^H}{\partial p^3}+\ldots, \tag{4}$$

where the operator relations are:

$$S_qf(q)S_q^{-1}=f\left(q+\sigma_q^2\frac{\partial}{\partial q}\right),\ S_pf(p)S_p^{-1}=f\left(p+\sigma_p^2\frac{\partial}{\partial p}\right), \tag{5}$$

which can be verified by Taylor expanding of operator. For instance,

$$\begin{aligned}
&S_qqS_q^{-1}=q+\sigma_q^2\frac{\partial}{\partial q},\\
&S_qq^2S_q^{-1}=q^2+\sigma_q^2+2\sigma_q^2q\frac{\partial}{\partial q}+\sigma^4\frac{\partial^2}{\partial q^2},\\
&S_qq^3S_q^{-1}=q^3+3\sigma_q^2+3\sigma_q^2(q^2+\sigma_q^2)\frac{\partial}{\partial q}+3\sigma_q^4q\frac{\partial^2}{\partial q^2}+\sigma^6\frac{\partial^3}{\partial q^3},
\end{aligned} \tag{6}$$

it is the same case for p.

In this paper, we consider double-well system subject to time-dependent perturbation with some incommensurate frequencies with the following Hamiltonian:

$$\mathcal{H}(t)=\frac{p^2}{2}+\frac{q^4}{4}-A(t)\frac{q^2}{2}, \tag{7}$$

where q and p represent the coordinate and the momentum, respectively. The time-dependent perturbation takes form

$$A(t)=a-\frac{1}{\sqrt{M}}\sum_{i=1}^{M}\epsilon\sin(\omega_i t), \tag{8}$$

the external filed $A(t)$ consists of M sinusoidal modes which are coupled to the system with strength $\epsilon=0.4$ and $a=5$ is constant. We set the frequencies of i_{th} sinusoidal mode in $A(t)$ is represented by ω_i as mutually incommensurate. The double well system driven by a periodically force characterizes the physics of a wide class of systems, such as the transfer of hydrogen in atoms [23] and molecules along chemical bonds and macroscopic quantum coherence phenomena in SQUIDs [24]. In the classical dynamics of this model with some frequency mode perturbation, it shows chaotic behavior caused by perturbation. The quantum Liouville equation for this system can been written as :

$$\begin{aligned}\frac{\partial\rho^H}{\partial t}=&-\frac{p}{m}\frac{\partial\rho^H}{\partial q}+[q^3+3\sigma_q^2-A(t)q]\frac{\partial\rho^H}{\partial p}+\left[3\sigma_q^2(q^2+\sigma_q^2)-A(t)\sigma_q^2-\frac{\sigma_p^2}{m}\right]\frac{\partial^2\rho^H}{\partial q\partial p}\\&+3\sigma_q^4 q\frac{\partial^3\rho^H}{\partial q^2\partial p}-\frac{\hbar^2}{4}q\frac{\partial^3\rho^H}{\partial p^3}+\sigma_q^6\frac{\partial^4\rho^H}{\partial q^3\partial p}-\frac{\hbar^2}{4}\sigma_q^2\frac{\partial^4\rho^H}{\partial q\partial p^3}.\end{aligned} \tag{9}$$

In ETMD method, it needs Husimi function used as distribution function and satisfy the continuity equation,

$$\frac{\partial\rho^H}{\partial t}+\frac{\partial}{\partial q}(\rho^H\dot{q})+\frac{\partial}{\partial p}(\rho^H\dot{p})=0. \tag{10}$$

Finally, the evolution equations of entangled trajectories in this model can be written as:

$$\begin{aligned}\dot{q}=&\frac{p}{m}-[3\sigma_q^2(q^2+\sigma_q^2)-A(t)\sigma_q^2-\frac{\sigma_p^2}{m}]\frac{1}{\rho^H}\frac{\partial\rho^H}{\partial p}+\frac{\hbar^2}{4}\sigma_q^2\frac{1}{\rho^H}\frac{\partial^3\rho^H}{\partial p^3},\\\dot{p}=&-[q^3+3\sigma_q^2-A(t)q]-3\sigma_q^4 q\frac{1}{\rho^H}\frac{\partial^2\rho^H}{\partial q^2}+\frac{\hbar^2}{4}q\frac{1}{\rho^H}\frac{\partial^2\rho^H}{\partial p^2}-\sigma_q^6\frac{1}{\rho^H}\frac{\partial^3\rho^H}{\partial q^3}.\end{aligned} \tag{11}$$

It is obvious that quantum effect has been considered in the evolution of trajectory because the state of system has been involved in the evolution equation, the evolution of trajectories are no longer statistically independent. To numerically solve Eq. 11, we use an

ansatz approximation of the Husimi function, namely, the continuous Husimi function ρ^H is represented by a finite ensemble of N trajectories [25,26],

$$\rho^H(\mathbf{q},\mathbf{p};t)=\frac{1}{N}\sum_{j=1}^{N}\delta(\mathbf{q}-\mathbf{q}_j(t))\delta(\mathbf{p}-\mathbf{p}_j(t)). \tag{12}$$

Because Husimi function is positive anywhere, this assumption is accurate.

3 Results and discussion

The entangled trajectory approach to quantum dynamics was suggested as a method for numerically evolving the Husimi function fully quantum mechanically using an ensemble of interacting trajectories, and this theoretical formalism was extended to the case of general potential and multi-dimensional case. The ETMD method's time consumption linearly increases with the increase of dimension (not exponential form as traditional quantum numerical calculation), it is a great advantage compared to traditional quantum numerical calculation. We use Gaussian wavepacket with zero momentum localized in the right well of the potential as an initial state in this paper:

$$\rho^H(q,p)=\frac{1}{2\pi\hbar}e^{-\frac{(q-q_0)^2}{4\sigma_q^2}-\frac{p^2}{4\sigma_p^2}}, \tag{13}$$

where we choose $q_0=\sqrt{5}$ is the right bottom of the unperturbed potential well and $\sigma_q=\sqrt{\frac{1}{6.8}}$,$\sigma_p=\sqrt{1.7}$ is the same as previous works [7–9]. The value of expectation of system's energy in phase space is defined as:

$$E(t)=\int H(\mathbf{q},\mathbf{p};t)\rho^H(\mathbf{q},\mathbf{p};t). \tag{14}$$

The ETMD method obeys Ehrenfest theorem as well, the ensemble behaves classically on average. This property can be used as judgment standard to check the accurate of our numerical calculation are and show ETMD is a powerful tool to study quantum dynamics of driven system. We show the evolution of system's energy perturbed with two mode different frequencies in Fig. 1. It is obvious that our result obtains from ETMD has only a little differences with one under classical dynamics, this result obeys Ehrenfest's theorem indicates our numerical calculations is accurate. To get careful comparison, a short time case has been shown in subplot (b). It also demonstrates that ETMD method is appropriate to deal with quantum driven system.

In this paper, we focus on coherent destruction of tunneling, one of the interesting phenomena in a quantum driven system. We define transition probability $P_L(t)$ which gives the probability that the wave packet is found in the left well in the Husimi presentation as:

$$P_L(t)=\int_{-\infty}^{0}dq\int_{-\infty}^{\infty}dp\rho^H(q,p,t)=\frac{1}{2}\left[1-\frac{1}{N}\sum_{j=1}^{N}erf\left(\frac{q_j(t)}{2h_q}\right)\right], \tag{15}$$

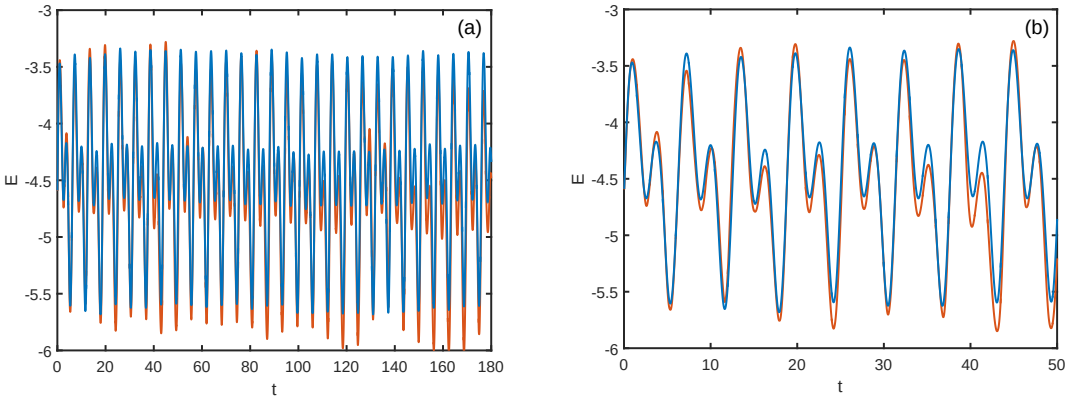

Figure 1: Time evolution of energy of the driven system. The blue line is the result of classical dynamics and tangerine line is the corresponding result of quantum dynamics with same initial state. Subplot (a) is the long time result, (b) is the short time result in order to provide better compare observation.

where $erf(x)=\frac{2}{\sqrt{\pi}}\int_0^x e^{-t^2}dt$ is error function and h_q is the width parameter determined by optimizing the fit of Eq. (12) to the initial Husimi distribution function. We show coherent destruction of tunneling phenomenon and its evolution of quantum coherence in Fig. 2 (a,b) and underline dynamical process with its energy evolution in Fig. 2 (c,d). In the subplot (a), all trajectories have been trapped in right well for all time under quantum dynamics at a glance, but some trajectories can conquer the barrier and go to the left well under classical dynamics. This phenomenon called coherent destruction of tunneling, entangled trajectories' barrier penetration have been suppressed by quantum coherence. Quantum coherence in phase space can be measure by purity defined as

$$Q=2\pi\hbar\int_{all}\rho^H(q,p;t)^2dqdp=\frac{2\pi\hbar}{2\pi h_q h_p N}\sum_{i,j=1}^{N}e^{-\frac{\left(q_{1j}(t)-q_{1i}(t)\right)^2}{2h_q^2}-\frac{\left(p_{1j}(t)-p_{1i}(t)\right)^2}{2h_p^2}}. \qquad (16)$$

Quantum coherence is maximum for $Q=1$ in a pure state and the system is complete decoherence in the case of $Q=0$. We show the evolution of quantum coherence under classical dynamics and quantum dynamics in subplot (b), quantum coherence under quantum dynamics always higher than one under classical dynamics. Entangled trajectories have energy exchange during evolution, it is wondering that whether entangled trajectory cannot conquer the barrier because of losing energy. ETMD method can give underline dynamics process of quantum dynamics, we show entangled trajectory and corresponding classical trajectory with the same initial state in phase space in subplot (c). Entangled trajectory always was trapped in right well but classical one back and forth across between the double well, its energy is shown in subplot (d) compared to one under classical dynamics. The energy of classical trajectory quasi-periodical change because of driven force, and it is clear that entangled trajectory's energy higher than the barrier

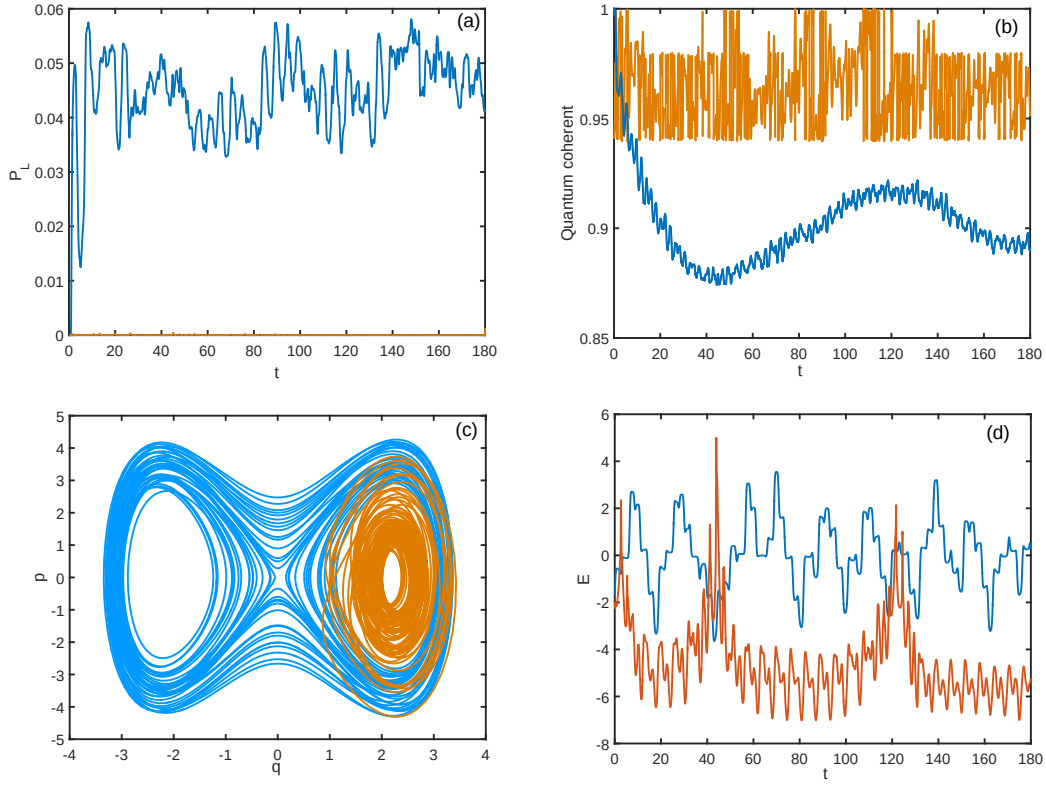

Figure 2: Quantum coherence suppress barrier penetration. Subplot (a) shows transition probability under quantum dynamics (blue line) and quantum one (tangerine line), Subplot (b) shows the evolution of quantum coherence under two different mechanics. The classical trajectory in subplot (c) conquer the barrier and back and forth between the two double well, but the entangled trajectory trapped in left well for all time. The energy fluctuation of classical trajectory and entangled trajectory is given in subplot (d). The detailed discussion is given in the text.

at some time. It is proved that entangled trajectory has been suppressed in right well not because of losing energy but effect by quantum coherence.

Classical chaos brings huge changes in quantum dynamics process, our model shows chaotic behave under classical dynamics in the case of three and more mode perturbation. Autocorrelation function is commonly used to study quantum dynamics process, in the theory of quantum phase space, it is defined as follow:

$$C = 2\pi\hbar \int_{all} \rho^H(q,p;t)\rho^H(q,p;0)dqdp. \tag{17}$$

It characterizes the "distance" between the initial state and state after some evolution, we show autocorrelation function as a function of time t for various the number of frequency components in Fig. 3. It is shown that autocorrelation function under two mode perturbation random oscillation between with small numerical area, quantum state change

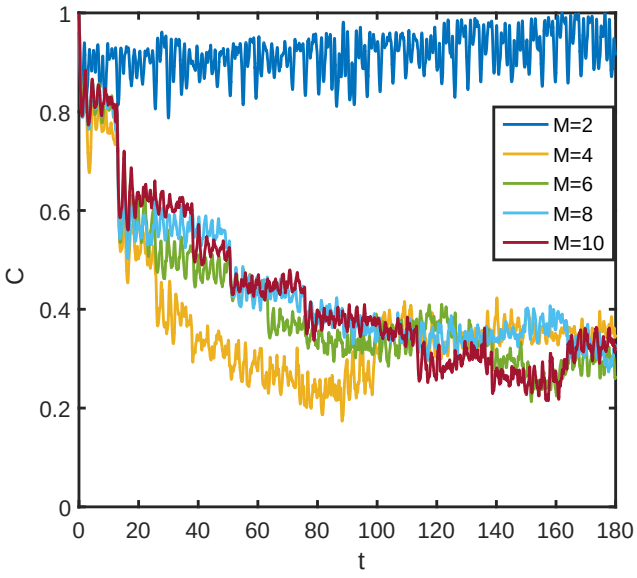

Figure 3: Autocorrelation function as a function of time t for various the number of frequency components. The values of M are $M=2$ (blue line), $M=4$ (yellowish line), $M=6$ (light green line), $M=8$ (baby blue line), $M=10$ (garnet line). The detailed discussion is given in the text.

around the initial state. For three and more mode perturbation, autocorrelation function evolution in all these case have same changes, there is only small difference between them. Because of classical chaos, quantum dynamics in all these cases deviate from the initial state rapidly and ruleless oscillation with time evolution.

4 Conclusion

We used entangled molecular dynamics method as a powerful tool to deal with quantum dynamics in double-well system with polychromatic perturbation. ETMD method obeys Ehrenfest's theorem, which can be used to check the correctness and accuracy of numerical calculation. It also shows that ETMD method is suitable to invest quantum dynamics in driven system. We gave the time evolution of energy in the case of system with two mode perturbation using ETMD method and compared to one under classical dynamics. It is clearly the two have delicate difference and our calculation is accurate. ETMD method gives underline process in quantum dynamics, a physical picture of coherent destruction of tunneling has been shown through showing entangled trajectory and its corresponding classical trajectory in phase space. Entangled trajectory's energy higher than barrier for some time, but it cannot come cross the barrier because of the coherence of quantum system, that is CDT phenomenon. Classical chaos has an enormous impact on quantum dynamics, our system show chaotic behavior for some mode per-

turbation under classical dynamics. In the view of autocorrelation evolution, quantum dynamics in this case deviate from the initial state rapidly and ruleless oscillation with time evolution.

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