Chapter 4
Does The Classically Chaotic Henon-Heiles Oscillator Exhibit Quantum Chaos Under Intense Laser Fields?

4.1 Introduction

During the last three decades the quantum domain behaviour of the fascinating and classically chaotic Henon-Heiles Oscillator (HHO) [1] has been studied in considerable detail [2-13]. The quantum HHO has dissociation energy of 13.3333 a.u. and 99 bounds states [14]; some of the bound state energy levels are doubly degenerate [5]. The regime of stochastic motion sets in at around $E = 9.0$, which is about two-third of the well-depth of dissociation limit, approximately half of the bound states are in this regime \textit{i.e.} between $E = 9.0$ and the dissociation limit [13,14]. In studying the possibility of quantum chaos for a classically chaotic system, earlier efforts were essentially directed towards evolving an external Gaussian wave packet under the HH potential by numerically solving the time-dependent Schrödinger equation (TDSE) wherein the \textit{Hamiltonian was time-independent} [7-12].

In contrast to a classically chaotic system, where the exponential divergence of trajectories in phase space is an unambiguous and confirmatory signature of chaos [15-17], the decision about whether a quantum system is chaotic or not is frequently not unambiguous and one cannot generally depend on only one "signature" of quantum chaos. This is mainly because the deterministic concept of an orbit or trajectory in classical mechanics vanishes in quantum mechanics. The difficulty is further compounded by the fact that in quantum domain, classical chaos might vanish due to quantum fluctuations although this need not be universally true (see the argument in [18]).
In spite of previous extensive works, the question whether the classically chaotic HHO can exhibit quantum chaos under a time-dependent Hamiltonian does not seem to have been addressed so far. In this chapter, we pursue this question by taking the TD potential as arising from an intense laser field acting on an electron moving as an HHO, without employing an external wave packet. The reasons for this are the following:

i. Atoms, molecules, clusters and solids reveal highly interesting nonlinear phenomena under intense laser fields, e.g. high-order harmonic generation (HHG), above-threshold ionization (ATI), stabilization under superintense laser fields, etc. During the last two decades, these aspects have been extensively studied both theoretically and experimentally [19-24].

ii. It has been speculated that helium atom might exhibit quantum chaos under intense laser fields [25].

iii. Where response to intense laser fields is concerned, e.g. HHG, ATI and stabilization, a striking parallelism has been established between atoms and molecules on one hand and one-dimensional nonlinear oscillators (with or without infinite barriers) on the other [26,27]. Note that HHO is a coupled two-dimensional system and the same parallelism should remain valid here.

iv. An intense laser field excites an electron in an atom to the continuum. Since the HHO is classically chaotic, it might manifest quantum chaos on excitation to the continuum.

v) Instead of evolving an external wave packet under the TDSE, it might be preferable to let the system itself (HHO in an intense laser field) generate its own wavepacket which would continue to evolve under the strong TD perturbation.

In this chapter, we would consider quantum chaos to be characterized by sensitive dependence on initial conditions, like classical chaos. Therefore our approach to studying the motion of an electron under the HH potential in intense laser field is as follows:
a) We first calculate the ground-state energy of the unperturbed HHO by numerically solving the TDSE in imaginary time. The first and second excited states are also calculated. Apart from settling the question of accuracy (see later) of our numerical algorithm for TDSE, it may be noted that the ground-state is the solution at zero time (initial input) for the real time Schrödinger equation involving the laser field.

b) Two slightly different initial inputs are generated in two different ways:

1) Keeping the Hamiltonian the same, the ground state is obtained under two different prescribed tolerances (convergence conditions). This leads to two slightly different ground-state energies and wavefunctions.

2) Keeping the tolerance the same, two slightly different laser fields, i.e. Hamiltonians, are taken: The initial input at t = 0 (unperturbed ground state), is the same for both the Hamiltonians. But after the first time step, the same input state is now slightly different for the two Hamiltonians involving slightly different laser intensities.

c) With respect to such different initial inputs, the TDSE was numerically solved in real time for lasers of varying intensities. Thus, the system generates its own wave packet. Finally, we examine a combination of several different signatures of quantum chaos which have emerged over the years.

The method of calculation is described in section 4.2. Section 4.3 gives laser and grid specifications and section 4.4 gives slightly different initial inputs. Section 4.5 discusses the results while section 4.6 summarizes the conclusions.

4.2 Methodology

The two-dimensional TDSE is (atomic units employed unless mentioned otherwise)

\[ H \Psi(x, y, t) = i \frac{\partial \Psi(x, y, t)}{\partial t} \]  

(4.1)
Where the Hamiltonian for an electron moving under the HH potential in the presence of laser field in x-direction is given by

\[ H = -\frac{1}{2} \frac{\partial^2}{\partial x^2} - \frac{1}{2} \frac{\partial^2}{\partial y^2} + V(x, y) - x \varepsilon_0 f(t) \sin(\omega_L t) \] (4.2)

Here \( \omega_L \) is the laser frequency, \( f(t) \) is the ramp function, \( \varepsilon_0 \) is to be obtained from the laser intensity \( \varepsilon_0 = (8\pi I/c)^{1/2} \), \( c \) is the velocity of light and \( V(x, y) \) is the HH potential

\[ V(x, y) = \frac{1}{2}(x^2 + y^2) + \lambda x(y^2 - \frac{x^2}{3}) \] (4.3)

where, the coupling constant \( \lambda \) is taken as 0.11180340. The ground state wave function of unperturbed HH potential is obtained by numerically solving the TDSE in imaginary time. The method is based on transforming the TDSE into an equation that resembles a diffusion quantum Monte Carlo (DQMC) equation [28]. Successive higher energies are calculated by employing the same imaginary time evolution but additionally requiring that an excited state is orthogonal to all the lower states. The numerical method reported earlier for one-dimensional oscillators [29,30] (see chapter 2) has been adopted for two-dimensional oscillators. Our calculated ground- and first two-excited states energies are in excellent agreement with the literature values (given in parentheses)[14]: \( E_0 = 0.998595 \) (0.9986), \( E_1 = 1.990768 \) (1.9901), \( E_2 = 2.956244 \) (2.9563). In order to evolve \( \psi(x, y, t=0) \) in real time under the HH potential in intense laser fields; TDSE is solved in real time as follows: Eq. (4.1) can be rewritten as

\[ -iH\Psi(x, y, t) = \frac{\partial \Psi(x, y, t)}{\partial t} \] (4.4)

i.e. \( \frac{\partial}{\partial t} = -iH \)

The Taylor expansion of \( \psi(x, y, t+\Delta t) \) around \( \psi(x, y, t) \) is given by
\[ \Psi(x, y, t + \Delta t) = \left[ 1 + \frac{dt}{\partial t} \frac{\partial^2}{\partial t^2} + \cdots \right] \Psi(x, y, t) \] (4.5)

So,

\[ \Psi(x, y, t + \Delta t) = e^{\frac{dt}{\partial t}} \Psi(x, y, t) \] (4.6)

\[ e^{-iH\Delta t} \] is the time-propagator which is a nonlinear evolution operator propagating the wavefunction \( \psi(x, y, t) \) from \( t \) to an advanced time \( t + \Delta t \).

Eq. (6) can be written in more symmetrical form as

\[ e^{iH\Delta t/2} \Psi(x, y, t + \Delta t) = e^{-iH\Delta t/2} \Psi(x, y, t) \] (4.7)

Putting the Hamiltonian given by Eq. (4.2) in Eq. (4.7)

\[ \exp\left( i \left( -\frac{1}{2} \frac{\partial^2}{\partial x^2} + \frac{V'(x, y)}{2} \right) \Delta t \right) \Psi(x, y, t + \Delta t) = \]

\[ \exp\left( -i \left( -\frac{1}{2} \frac{\partial^2}{\partial x^2} + \frac{V'(x, y)}{2} \right) \Delta t \right) \Psi(x, y, t) \] (4.8)

Here, \( V'(x, y) \) includes the oscillator potential, \( V(x, y) \) and the electric field term in the \( x \)-direction, i.e.

\[ V'(x, y) = V(x, y) - x \varepsilon_0 f(t) \sin(\omega_L t) \] (4.9)

Separating \( x \) and \( y \) terms on both sides, Eq. (4.8) can be rewritten as

\[ \exp\left( i \left( -\frac{1}{2} \frac{\partial^2}{\partial x^2} + \frac{V'(x, y)}{2} \right) \Delta t \right) \exp\left( -i \left( -\frac{1}{2} \frac{\partial^2}{\partial y^2} + \frac{V'(x, y)}{2} \right) \Delta t \right) \Psi(x, y, t + \Delta t) = \]

\[ \exp\left( -i \left( -\frac{1}{2} \frac{\partial^2}{\partial x^2} + \frac{V'(x, y)}{2} \right) \Delta t \right) \exp\left( -i \left( -\frac{1}{2} \frac{\partial^2}{\partial y^2} + \frac{V'(x, y)}{2} \right) \Delta t \right) \Psi(x, y, t) \] (4.10)

Expansion of the exponential operator on both sides of the equation followed by truncation after the second term leads to
\[
\left(1 + i \left( \frac{-1}{2} \frac{\partial^2}{\partial x^2} + \frac{V'(x,y)}{2} \right) \frac{\Delta t}{2} \right) \left(1 + i \left( \frac{-1}{2} \frac{\partial^2}{\partial y^2} + \frac{V'(x,y)}{2} \right) \frac{\Delta t}{2} \right) \Psi(x,t,t + \Delta t) = \nabla L + V'(x,y) \Delta t
\]

\[
\left(1 - i \left( \frac{-1}{2} \frac{\partial^2}{\partial x^2} + \frac{V'(x,y)}{2} \right) \frac{\Delta t}{2} \right) \left(1 - i \left( \frac{-1}{2} \frac{\partial^2}{\partial y^2} + \frac{V'(x,y)}{2} \right) \frac{\Delta t}{2} \right) \Psi(x,y,t) \]

(4.11)

Using Peaceman-Rachford Splitting, the above equation leads to following two equations

\[
\left(1 + i \left( \frac{-1}{2} \frac{\partial^2}{\partial x^2} + \frac{V'(x,y)}{2} \right) \frac{\Delta t}{2} \right) \Psi_{l,m}^* = \left(1 - i \left( \frac{-1}{2} \frac{\partial^2}{\partial y^2} + \frac{V'(x,y)}{2} \right) \frac{\Delta t}{2} \right) \Psi_{l,m}^n
\]

(4.12)

\[
\left(1 + i \left( \frac{-1}{2} \frac{\partial^2}{\partial x^2} + \frac{V'(x,y)}{2} \right) \frac{\Delta t}{2} \right) \Psi_{l,m}^{n+1} = \left(1 - i \left( \frac{-1}{2} \frac{\partial^2}{\partial y^2} + \frac{V'(x,y)}{2} \right) \frac{\Delta t}{2} \right) \Psi_{l,m}^* \]

(4.13)

Here $\Psi_{l,m}^*$ is an intermediate function that acts as a bridge between $\Psi_{l,m}^n$ and $\Psi_{l,m}^{n+1}$. We now approximate $\partial^2 / \partial x^2$ and $\partial^2 / \partial y^2$ by three-point difference formula as follows

\[
\frac{\partial^2}{\partial x^2} \Psi_{l,m}^{n+1} = \frac{1}{h^2} \delta x^2 \Psi_{l,m}^{n+1} = \frac{1}{h^2} \left( \Psi_{l+1,m}^{n+1} - 2 \Psi_{l,m}^{n+1} + \Psi_{l-1,m}^{n+1} \right)
\]

(4.14)

and

\[
\frac{\partial^2}{\partial x^2} \Psi_{l,m}^n = \frac{1}{h^2} \delta x^2 \Psi_{l,m}^n = \frac{1}{h^2} \left( \Psi_{l+1,m}^n - 2 \Psi_{l,m}^n + \Psi_{l-1,m}^n \right)
\]

(4.15)

\[
\frac{\partial^2}{\partial y^2} \Psi_{l,m}^{n+1} = \frac{1}{h^2} \delta y^2 \Psi_{l,m}^{n+1} = \frac{1}{h^2} \left( \Psi_{l,m+1}^{n+1} - 2 \Psi_{l,m}^{n+1} + \Psi_{l,m-1}^{n+1} \right)
\]

(4.16)

and

\[
\frac{\partial^2}{\partial y^2} \Psi_{l,m}^n = \frac{1}{h^2} \delta y^2 \Psi_{l,m}^n = \frac{1}{h^2} \left( \Psi_{l,m+1}^n - 2 \Psi_{l,m}^n + \Psi_{l,m-1}^n \right)
\]

(4.17)
Using Eq. (4.14) to (4.17) in Eqs. (4.12) we obtain

\[
\left( \Psi_{l,m}^{n} - i\left( -\frac{1}{2h^2} \left( \Psi_{l+1,m}^{n} - 2\Psi_{l,m}^{n} + \Psi_{l-1,m}^{n} \right) + \frac{V'(x,y)}{2} \Psi_{l,m}^{*} \right) \Delta t \right) = \\
\left( \Psi_{l,m}^{n} + i\left( -\frac{1}{2h^2} \left( \Psi_{l+1,m+1}^{n} - 2\Psi_{l,m+1}^{n} + \Psi_{l-1,m+1}^{n} \right) + \frac{V'(x,y)}{2} \Psi_{l,m}^{*} \right) \Delta t \right)
\]

(4.18)

On rearranging various terms, we obtain

\[
\begin{align*}
-\frac{i\Delta t}{4h^2} \Psi_{l+1,m}^{n} + \left( 1 + \frac{i\Delta t}{2h^2} + \frac{iV'(x,y)\Delta t}{4} \right) \Psi_{l,m}^{n} - \frac{i\Delta t}{4h^2} \Psi_{l-1,m}^{n} = \\
\frac{i\Delta t}{4h^2} \Psi_{l,m+1}^{n} + \left( 1 - \frac{i\Delta t}{2h^2} - \frac{iV'(x,y)\Delta t}{4} \right) \Psi_{l,m}^{n} + \frac{i\Delta t}{4h^2} \Psi_{l,m-1}^{n}
\end{align*}
\]

(4.19)

Eq. (4.19) can be written as

\[
\alpha_{l,m}^{*} \Psi_{l-1,m}^{n} + \beta_{l,m}^{*} \Psi_{l,m}^{n} + \gamma_{l,m}^{*} \Psi_{l+1,m}^{n} = \xi_{l,m}^{n}
\]

(4.20)

where,

\[
\begin{align*}
\alpha_{l,m}^{*} & = -\frac{i\Delta t}{4h^2} \\
\beta_{l,m}^{*} & = 1 + \frac{i\Delta t}{2h^2} + \frac{iV'(x,y)\Delta t}{4} \\
\gamma_{l,m}^{*} & = -\frac{i\Delta t}{4h^2}
\end{align*}
\]

\[
\xi_{l,m}^{n} = \frac{i\Delta t}{4h^2} \Psi_{l,m+1}^{n} + \left( 1 - \frac{i\Delta t}{2h^2} - \frac{iV'(x,y)\Delta t}{4} \right) \Psi_{l,m}^{n} + \frac{i\Delta t}{4h^2} \Psi_{l,m-1}^{n}
\]

(4.21)

Eq. (4.20) can be recast into a tridiagonal matrix equation

\[
\begin{bmatrix}
\beta_{1}^{*} & \gamma_{1}^{*} & 0 \\
\alpha_{2}^{*} & \beta_{2}^{*} & \gamma_{2}^{*} \\
0 & \gamma_{N_{1}-1}^{*} & \alpha_{N_{1}}^{*}
\end{bmatrix}
\begin{bmatrix}
\Psi_{1}^{*} \\
\Psi_{2}^{*} \\
\Psi_{N_{1}}^{*}
\end{bmatrix}
= \\
\begin{bmatrix}
\xi_{1}^{n} \\
\xi_{2}^{n} \\
\xi_{N_{1}}^{n}
\end{bmatrix}
\]

Similarly, using Eqs. (4.14) to (4.17) in Eq. (4.13) leads to
\[ - \frac{i \Delta t}{4h^2} \Psi_{l,m+1}^{n+1} + \left( 1 + \frac{i V(x,y) \Delta t}{2h^2} + \frac{i V'(x,y) \Delta t}{4} \right) \Psi_{l,m}^{n+1} - \frac{i \Delta t}{4h^2} \Psi_{l,m-1}^{n+1} = \]  
\[ \frac{i \Delta t}{4h^2} \Psi_{l+1,m}^* + \left( 1 + \frac{i V(x,y) \Delta t}{2h^2} - \frac{i V'(x,y) \Delta t}{4} \right) \Psi_{l,m}^* + \frac{i \Delta t}{4h^2} \Psi_{l-1,m}^* \]  
\text{(4.22)}

Eq. (4.22) can be written as
\[ \alpha_{l,m} \Psi_{l,m}^{n+1} + \beta_{l,m} \Psi_{l,m-1}^{n+1} + \gamma_{l,m} \Psi_{l,m+1}^{n+1} = \xi_{l,m} \]  
\text{(4.23)}

where,
\[ \alpha_{l,m} = -\frac{i \Delta t}{4h^2} \]
\[ \beta_{l,m} = 1 + \frac{i \Delta t}{2h^2} + \frac{i V'(x,y) \Delta t}{4} \]
\[ \gamma_{l,m} = -\frac{i \Delta t}{4h^2} \]
\[ \xi_{l,m}^* = \frac{i \Delta t}{4h^2} \Psi_{l+1,m}^* + \left( 1 + \frac{i V(x,y) \Delta t}{2h^2} - \frac{i V'(x,y) \Delta t}{4} \right) \Psi_{l,m}^* + \frac{i \Delta t}{4h^2} \Psi_{l-1,m}^* \]

Eq. (4.23) can also be recast into a tridiagonal matrix equation
\[ \begin{bmatrix} \beta_1 & \gamma_1 & 0 & \ldots & 0 \\ \alpha_2 & \beta_2 & \gamma_2 & \ldots & 0 \\ 0 & \ldots & \gamma_{N_1-1} & \alpha_{N_1} & \beta_{N_1} \\ 0 & \ldots & \alpha_{N_1} & \beta_{N_1} & \gamma_{N_1-1} \end{bmatrix} \begin{bmatrix} \Psi_{1}^{n+1} \\ \Psi_{2}^{n+1} \\ \ldots \\ \Psi_{N_1}^{n+1} \end{bmatrix} = \begin{bmatrix} \xi_1^* \\ \xi_2^* \\ \ldots \\ \xi_{N_1}^* \end{bmatrix} \]  
\text{(4.24)}

Eqs. (4.21) and (4.24) are solved by using a modified Thomas algorithm [31].

4.3 Laser and Grid Specifications

i) Laser wavelength $\lambda_L = 1064$ nm

ii) Laser frequency $\omega_L = 0.0428228$ a.u.

iii) Laser intensity $I = 5 \times 10^{13} \cdot 2.001 \times 10^{17}$ Wcm$^{-2}$

iv) Time increment $\Delta t = 0.05$.

v) Total time of laser $0 \leq t \leq 2252.75$ (55 fs), 15 optical cycles.
vi) For $I = 5 \times 10^{13} - 1 \times 10^{17}$ Wcm$^{-2}$ the computations were carried on till 29 optical cycles. Since no noticeable difference was observed compared to 15 optical cycles, all the results are reported up to 15 optical cycles.

vii) The linear ramp is $f(t) = \frac{t}{t_0}$ up to 5 optical cycles and unity thereafter.

viii) In the present work, the spatial grid is taken as $-150 \leq x \leq 150$, $-15 \leq y \leq 15$ for $I = 2 \times 10^{17}$, 2.001$\times 10^{17}$ Wcm$^{-2}$ and $-15 \leq x \leq 15$, $-5 \leq y \leq 5$ for $I = 5 \times 10^{13} - 1 \times 10^{17}$ Wcm$^{-2}$ and $\Delta x = \Delta y = 0.1$ for $I = 2 \times 10^{17}$, 2.001$\times 10^{17}$ Wcm$^{-2}$ while $\Delta x = \Delta y = 0.02$ for $I = 5 \times 10^{13} - 1 \times 10^{17}$ Wcm$^{-2}$.

4.4 Slightly Different Initial Inputs

The two different initial inputs (Section 4.1) generated in two different ways are:

i. The ground-state eigenfunctions $\psi_1(x,y,t=0)$ and $\psi_2(x,y,t=0)$ obtained with tolerance limits $10^{-12}$ and $10^{-8}$ respectively.

ii. The wavefunction $\psi_1(x,y,t=0)$ evolved under HH potential in laser fields of slightly different intensities, i.e. $2 \times 10^{17}$ Wcm$^{-2}$ and 2.001$\times 10^{17}$ Wcm$^{-2}$. When $\psi_1(x,y,t=0)$ is evolved in time under the laser field of $I = 2.001 \times 10^{17}$ Wcm$^{-2}$, it is designated as $\psi_1/(x,y,t)$.

4.5 Results and Discussion

For the two highest intensities, a larger grid (specified above) was employed in order to take care of reflection/transmission of the wavefunction at/through the boundaries. Nevertheless, the norm $N(t)$ changed by $\pm 3\%$ as shown in Fig. 4.1 and these fluctuations were taken care of by renormalizing the wavefunction to the last stable value (1.000267). For comparison, we also report a similar study on an electron moving in two dimensional harmonic oscillator (HO) potential under a laser field of intensity $2 \times 10^{17}$ Wcm$^{-2}$.
Fig. 4.1 Plots of normalization constant $N(t)$ against time $t$ in a.u. for Henon-Heiles oscillator (a) at $I = 5 \times 10^{16}$, (b) at $I = 2 \times 10^{17}$ W cm$^{-2}$ respectively.
The quantum dynamical motion of the electron moving in the HH potential under intense laser fields is studied through various time-dependent quantities which also help in identifying the possible signatures of quantum chaos.

### 4.5.1 Autocorrelation function

The correlation of the system with its initial state is measured in terms of the autocorrelation function [8]

\[ C(t) = \left| \langle \Psi_1(x, y, t = 0) | \Psi_1(x, y, t) \rangle \right|^2 \]  

(4.25)

Fig. 4.2 shows the variation of \( C(t) \) with time, the laser electric field \( \varepsilon(t) \) is given in Fig. 4.2(a). For the HH potential under low intensity (5x10\(^{13}\)-5x10\(^{16}\) Wcm\(^{-2}\), Figs. 4.2(c) and (d)) and HO potential under intensity (2x10\(^{17}\) Wcm\(^{-2}\), Fig. 4.2(a)), the periodicity in \( C(t) \) is twice that of \( \varepsilon(t) \). Although the value of \( C(t) \) falls below unity after 5 optical cycles, it periodically returns to its initial value. Thus, in all the three cases the system is well correlated to its initial state. However, note that the fall in the value of \( C(t) \) increases as the laser intensity is increased. But, when the HH potential is under the intensity 2x10\(^{17}\) Wcm\(^{-2}\), \( C(t) \) decays rapidly and does not return to its starting value of unity. This behaviour is an indication of the departure of the system from its initial quantum state which may be regarded as a signature of quantum chaos.

### 4.5.2 Power spectrum

Similar behaviour is reflected in the power spectra \( A(\omega) \) obtained through the fast fourier transform (FFT) of \( C(t) \) for the last 6 optical cycles with integration limits \( t_1 = 819.15 \text{ a.u.} \) to \( t_2 = 1638.35 \text{ a.u.} \). \( A(\omega) \) is given by
A relatively simple spectrum with a few lines is obtained for the HH potential at $I = 5 \times 10^{16}$ Wcm$^{-2}$ (Fig. 4.3(b)) and for HO at $I = 2 \times 10^{17}$ Wcm$^{-2}$ (Fig. 4.3(a)). The HH potential at $I = 2 \times 10^{17}$ Wcm$^{-2}$ shows a rich spectrum (Fig. 4.3(c)) that has some characteristics of atomic ATI spectrum as the higher energy peaks are less intense than the lower ones. The complete spectrum of energy levels of the HH potential below the dissociation energy (13.3333 a.u.) is given in the literature [14]. The comparative analysis of power spectrum obtained for HH at $I = 2 \times 10^{17}$ Wcm$^{-2}$ with the available energy spectrum reveals that about 25 states appear in the power spectrum with significant intensity. Higher excited states and the peak corresponding to the last bound state also appears, although with much less intensity. This implies that the continuum states are contributing to the power spectrum.

### 4.5.3 Nearest neighbour spacing distribution

The statistical analysis of the power spectrum is done by nearest-neighbour spacing distribution. This shows Poisson's distribution

$$P_p(s) = \exp(-s)$$

(4.27)

for integrable systems and Wigner distribution

$$P_w(s) = \frac{\pi S}{2} \exp(-\pi S^2/4)$$

(4.28)

for classically chaotic systems [1], where $S$ is the gap between two nearest energy levels. For HH at $I = 5 \times 10^{16}$ Wcm$^{-2}$, the statistical behaviour is neither clear-cut Wigner nor Poisson (Fig. 4.3(e)). The HO (Fig. 4.3(d)) and HH (Fig. 4.3(f)) at $I = 2 \times 10^{17}$ Wcm$^{-2}$ seem to follow Poisson and Wigner-like statistics respectively. It may be noted that while a small number of peaks obtained in the spectrum in Fig. 4.3(a) and 4.3(b) is an impediment in doing a statistical
analysis, it also does not work when there is a rich spectrum like Fig. 4.3(c). In general, it can be said that statistical analysis is not helpful when the quantum dynamics of a system is studied in presence of an external periodic field. This is because the peaks in the spectrum are equally spaced (equal to photon energy) and if the peaks are shifted they are shifted equally and the separation between the peaks is given by a multiple of photon energy. Hence, the statistical divergence is not seen in the spectrum. It appears likely that statistical analysis may also not be successful in the study of atoms and molecules in intense laser fields.

4.5.4 Distance function

Fig. 4.3 depicts the variation of distance function [32,33] with time. Since the present work employs initial inputs generated in two different ways, D(t) is defined separately for each initial input:

(i) Distance function obtained from slightly different initial wavefunctions \( \psi_1(x,y,t=0) \) and \( \psi_2(x,y,t=0) \) that are evolved under the same Hamiltonian is given by

\[
D(t) = \left\{ \left( \langle x_1(t) \rangle - \langle x_2(t) \rangle \right)^2 + \left( \langle p_{x1}(t) \rangle - \langle p_{x2}(t) \rangle \right)^2 \right\}^{1/2}
\]  

(4.29)

where \( \langle x_1(t) \rangle, \langle p_{x1}(t) \rangle \) and \( \langle x_2(t) \rangle \) and \( \langle p_{x2}(t) \rangle \) are expectation values at time \( t \) of position and momentum operators in x-direction with respect to \( \psi_1(x,y,t) \) and \( \psi_2(x,y,t) \) respectively. Here expectation values in y-direction, i.e. \( \langle y \rangle \) and \( \langle p_y \rangle \) are not considered as the potential is symmetric in y-direction and therefore these values vanish.

(ii) Distance function obtained from same initial wavefunction that is evolved under slightly different Hamiltonians is given by

\[
D_H(t) = \left\{ \left( \langle x_1(t) \rangle - \langle x'_1(t) \rangle \right)^2 + \left( \langle p_{x1}(t) \rangle - \langle p'_{x1}(t) \rangle \right)^2 \right\}^{1/2}
\]  

(4.30)
Here $<x_1'(t)>$ and $<p_{x_1}'(t)>$ are expectation values calculated with $\psi_1'(x,y,t)$. Note that $\psi_1(x,y,t=0) = \psi_1'(x,y,t=0)$. Fig. 4.4(a) shows minor fluctuations in $D(t)$ that fall on a straight line for HH at $I = 5 \times 10^{13}$- $1 \times 10^{17}$ Wcm$^{-2}$ and HO for the laser intensity $2 \times 10^{17}$ Wcm$^{-2}$. When the HH potential is under this intensity, there are prominent peaks after 5 optical cycles (Fig. 4.4(b)). But the change in $D(t)$ is negligible as compared to the increase in $D_H(t)$ (Fig. 4.4(c)). Also, the peak height decreases in $D(t)$ whereas it increases for $D_H(t)$. Thus, the distance corresponding to two identical wavefunctions that are evolving in “phase space” under slightly different Hamiltonians increases. This is analogous to the exponential divergence of classical phase space trajectories characteristic of chaotic systems. This behaviour also exhibits greater sensitivity of quantum motion to small changes in the Hamiltonian rather than in the initial quantum state when the underlying classical motion is chaotic.

In the spirit of classical dynamics, a quantum Lyapunov exponent has been defined as [32,34]

$$\Lambda_+ = \lim_{D(0) \to 0} \frac{1}{t \to \infty} \ln \frac{D(t)}{D(0)}$$

(4.31)

The corresponding Kolmogorov-Sinai (KS) entropy has been defined as

$$E_{KS} = \sum_{\Lambda_+ > 0} \Lambda_+$$

(4.32)

It was noticed here that the KS entropy is positive at all laser intensities. At the final time, $t = 2252.75$, it has the highest value for HO, $E_{KS} = 3774.628 (D_0 = 5.444 \times 10^{-18})$ whereas $E_{KS} = 449.31 (D_0 = 3.862 \times 10^{-6})$ for HH at $I = 2 \times 10^{17}$ Wcm$^{-2}$. The criterion of increased KS entropy associated with exponential sensitivity to initial conditions is not found here to be unambiguously valid.
Fig. 4.2 Plots of correlation function $C(t)$ against time $t$ in a.u. for (a) two-dimensional harmonic oscillator at $I = 2 \times 10^{17}$ W cm$^{-2}$ and the Henon-Heiles oscillator at (b) $I = 2 \times 10^{17}$, (c) $I = 5 \times 10^{16}$, (d) $I = 5 \times 10^{13}$ W cm$^{-2}$ respectively. In (a) the laser electric field $\varepsilon(t)$ is shown in dotted line for $I = 2 \times 10^{17}$ W cm$^{-2}$. For other lower intensities the field amplitude decreases but the periodicity remains the same.
Fig. 4.3 Power spectra plotted against the photon energy ($\omega$), in a.u. for (a) the two-dimensional harmonic oscillator at $I = 2 \times 10^{17}$ W cm$^{-2}$ and the Henon-Heiles oscillator at (b) $I = 5 \times 10^{16}$, (c) $I = 2 \times 10^{17}$ W cm$^{-2}$ respectively. The corresponding nearest-neighbour spacing distribution $P(s)$ is plotted against the energy level spacing $S$ in (d), (e) and (f) respectively.
Fig. 4.4 Plot of distance function $D(t)$, in a.u. of Henon-Heiles oscillator against time. Both (a), (b) refer to $D(t)$ calculated with two slightly different initial states and (c) refers to $D(t)$ calculated with two slightly different Hamiltonians. (a) $I = 5 \times 10^{13} - 1 \times 10^{17}$, (b) $I = 2 \times 10^{17}$ and (c) $I = 2.001 \times 10^{17}$ Wcm$^{-2}$ respectively.
4.5.5 Overlap integral

Another criterion to measure the sensitivity of quantum dynamical motion to the initial quantum state or the Hamiltonian is provided by the overlap integral, \( I(t) \) [18]. For the slightly different initial wavefunctions, \( \psi_1(x,y,t=0) \) and \( \psi_2(x,y,t=0) \) that are evolved under the same Hamiltonian, \( I(t) \) is given by

\[
I_1(t) = \left| \langle \Psi_1(x,y,t) \mid \Psi_2(x,y,t) \rangle \right|^2
\]

(4.33)

and when \( \psi_1(x,y,t=0) \) and \( \psi'_1(x,y,t=0) \) \((\psi_1 = \psi'_1\) only at \( t=0 \)) are evolved under slightly different Hamiltonians, \( I(t) \) is defined as

\[
I_2(t) = \left| \langle \Psi_1(x,y,t) \mid \Psi'_1(x,y,t) \rangle \right|
\]

(4.34)

Fig. 4.5(b) shows that \( I_2(t) \) decays rapidly after 5 optical cycles and falls to 0.72 at the end of the 15th optical cycle. Due to the unitarity of the time-evolution operator, \( I_1(t) \) maintains a steady initial value, Fig. 4.5(a), \( I_2(t) \) changes as the Hamiltonian is slightly changed. The decay of the overlap between the time-evolved quantum states under slightly different Hamiltonians again exhibits greater sensitivity of quantum motion to slight changes in the Hamiltonian.

4.5.6 Quantum “phase space” volume

The variation of quantum “phase space” volume [8] \( V(t) \), or the uncertainty product, with time and the laser electric field \( \varepsilon(t) \) is given in Fig. 4.6. \( V(t) \) is defined as

\[
V(t) = \left( \left( \langle x^2 \rangle - \langle x \rangle^2 \right) \left( \langle p_x^2 \rangle - \langle p_x \rangle^2 \right) \left( \langle y^2 \rangle - \langle y \rangle^2 \right) \left( \langle p_y^2 \rangle - \langle p_y \rangle^2 \right) \right)^{1/2}
\]

(4.35)

where all the expectation values are with respect to \( \psi_1(x,y,t) \). \( V(t) \) remains nearly constant for the HH potential under lower field intensity \( (5 \times 10^{13} - 1 \times 10^{17} \text{ Wcm}^{-2}) \), Fig. 4.6(b)) and for HO potential under intensity \( 2 \times 10^{17} \text{ Wcm}^{-2} \).
(Fig. 4.6(b)). It increases after 5 optical cycles and is around 1800 for the HH potential at $2 \times 10^{17}$ W cm$^{-2}$ intensity (Fig. 4.6(a)). Interestingly, the maxima in $V(t)$ correspond to the minima in the laser electric field $\varepsilon(t)$. A large increase in $V(t)$ can be interpreted as the fingerprint of quantum chaos as it implies a loss of information about the electron being represented by the wavepacket. There is a likelihood of the system characterized by large $V(t)$ to follow a chaotic trajectory.

### 4.5.7 Quantum “phase space” trajectory

Fig. 4.7 shows the quantum “phase space” trajectories where $\langle x(t) \rangle$ is plotted against $\langle p_x(t) \rangle$ in the spirit of Ehrenfest theorem [35].

$$\frac{m}{i} \frac{d}{dt} \langle \hat{r} \rangle = \int \Psi^* \frac{\hbar}{i} \nabla \Psi \, d^3r = \langle p \rangle$$  \hspace{1cm} (4.36)

$$\frac{d}{dt} \langle p \rangle = - \int \Psi^* (\nabla \Psi) \Psi \, d^3r = -\langle \nabla^2 \rangle = \langle F \rangle$$  \hspace{1cm} (4.37)

Fig. 4.7(a),(c), and (d) show the spiral trajectory obtained for HH potential in laser fields of lower intensities and for HO potential at $2 \times 10^{17}$ W cm$^{-2}$ intensity. When the HH potential is under the laser field of this intensity, the trajectory starts as a spiral but rapidly diverges into a complicated pattern (Fig. 4.7(b)).

It may be mentioned here that the trajectories obtained by evolving slightly different wavefunctions under the same Hamiltonian superimpose on each other. This fact, in addition to earlier observations (KS entropy values, distance function and overlap integral) indicate that the time evolution of quantum systems is much more sensitive to small changes in the Hamiltonian. This is in accord with earlier studies [36,37] which demonstrated that the evolution of a quantum state was altered under a slightly perturbed Hamiltonian, the overlap between the perturbed and unperturbed states tended to a comparatively small value if the analogous classical motion is chaotic than if it is regular. This may be explained by
saying that in quantum dynamical motion all the states are mixed (linear combination of other states), whereas the classical exponential sensitivity to the initial state applies only to an individual state [18]. However, it has also been argued that quantum systems exhibit a state sensitivity very similar to classical state sensitivity, by using computational motion reversal [18].

Fig. 4.8 shows \( <p_x(t)> \) for HH at \( I = 2 \times 10^{17} \) Wcm\(^{-2} \) against \( <x(t)> \) at the end of each optical cycle. The values lie close together for the first few optical cycles but diverge by the end of the 15\(^{th} \) optical cycle, thus displaying greater sensitivity to the initial Hamiltonian.

### 4.5.8 Potential energy surface and Probability density plots

The potential energy surfaces (PES) and the probability densities at \( t=0 \) as well as at the crest (\( \epsilon_0 = 2.3872021, \ t = 2090.85 \)) and trough (\( \epsilon_0 = -2.387202, \ t = 2164.20 \)) of the 15\(^{th} \) optical cycle for \( I = 2 \times 10^{17} \) Wcm\(^{-2} \) are shown in Fig. 4.9. The ground-state probability density is given by a single centrosymmetric peak (Fig. 4.9(b)). The shifting of the minima of the PES and likewise the spread of probability density is in the positive and negative x-direction respectively at the crest (Fig. 4.9(c), (d)) and at the trough (Fig. 4.9(e), (f)) of the laser electric field. It is clear that the initial quantum state is evolving into a mixture of numerous states. When the HH potential is under a low-intensity field, the norm was preserved at unity but it changed by ±3% when the laser intensity \( 2 \times 10^{17} \) Wcm\(^{-2} \) is applied, even though a larger grid was employed at this intensity. This indicates that the probability density is leaking into the continuum. Moreover, the spread of the probability density all over the grid and the existence of higher bound states in the power spectrum suggest that an electron moving under the HH potential in presence of a laser field of intensity \( 2 \times 10^{17} \) Wcm\(^{-2} \) has reached the continuum. This is further supported by the high harmonic generation spectrum.
Fig. 4.5 Overlap integral $I(t)$ plotted against optical cycle (a) refers to $I_1(t)$ calculated with two slightly different initial states while (b) refers to $I_2(t)$ obtained for slightly different Hamiltonian.
Fig. 4.6 Plot of "phase space" volume of Henon-Heiles oscillator against time. (a) refers to $I = 2 \times 10^{17}$ Wcm$^{-2}$ while (b) refers to $I = 1 \times 10^{17}, 5 \times 10^{16}, 5 \times 10^{15}, 5 \times 10^{13}$ Wcm$^{-2}$ as well as the two-dimensional harmonic oscillator at $I = 2 \times 10^{17}$ Wcm$^{-2}$. The laser electric field $\varepsilon(t)$ is also shown in dotted line. All quantities are in atomic units.
Fig. 4.7 Quantum "phase space" trajectories, in a.u., of (a) the two-dimensional harmonic oscillator at $I = 2 \times 10^{17}$ W cm$^{-2}$ and of the Henon-Heiles oscillator at (b) $I = 2 \times 10^{17}$ (c) $I = 5 \times 10^{16}$, and (d) $I = 5 \times 10^{13}$ W cm$^{-2}$ respectively.
Fig. 4.8 The expectation values (a.u) $\langle p_x(t) \rangle$ of the momentum operator in $x$-direction, plotted against the expectation value of position operator $\langle x(t) \rangle$ at the end of each optical cycle, for the Henon-Heiles oscillator. (a) $I = 2.001 \times 10^{17}$, (b) $I = 2 \times 10^{17}$ Wcm$^{-2}$. 
Fig. 4.9 The Henon-Heiles oscillator potential energy surface (a) and the corresponding probability density (b) in a.u at time $t = 0$. 
Fig. 4.9 continued. The Henon-Heiles oscillator potential energy surface (c) and the corresponding probability density (d) in a.u., for $I = 2 \times 10^{17}$ Wcm$^{-2}$ at the crest ($\varepsilon(t) = \varepsilon_0$, $t = 2090.5$ a.u.) of the 15th optical cycle.
Fig. 4.9 continued. The Henon-Heiles oscillator potential energy surface (e) and the corresponding probability density (f) in a.u., for $I = 2 \times 10^{17}$ Wcm$^{-2}$ at the trough ($\varepsilon(t) = -\varepsilon_0$, $t = 2164.20$ a.u.) of the 15$^{th}$ optical cycle.
4.5.9 High harmonic generation (HHG)

The source of high harmonic generation is the polarization of the medium caused by intense laser field. This involves induction of time-varying dipole moment, \( \mu(t) = \langle \psi_1(x,y,t) \mid x \mid \psi_1(x,y,t) \rangle \) in the species when exposed to the laser field. The HHG spectrum \( H(\omega) \) is obtained by taking the FFT of the time-varying dipole moment \( \mu(t) \) for the last 6 optical cycles with integration limits \( t_1 = 819.15 \) to \( t_2 = 1638.35 \), viz

\[
H(\omega) = \left| \int_{t_1}^{t_2} \mu(t) e^{-i\omega t} dt \right|^2 ; \quad -\infty \leq \omega \leq +\infty, \ \mu(t) = \mu(-t)
\]  

(4.38)

and plotted against the harmonic order. The HHG spectrum presents a series of peaks centered at the harmonics or the multiples of laser frequency due to the generation of frequency components of dipole moment at these multiples of the incident laser frequency. There is no significant harmonic spectrum below \( 2\times10^{17} \ Wcm^{-2} \) as the electron has not reached the highly excited states (Fig. 4.10(c), (d)). For HO (Fig. 4.10(a)) the spectrum shows a single peak as it is a bound potential. The plateau of harmonics obtained in the HHG spectrum at \( I = 2\times10^{17} \ Wcm^{-2} \) results from transitions to the continuum and subsequent emissions to lower levels, especially the ground-state. The spectrum has a rich multiplet structure that is rich in both even and odd harmonics (inset of Fig. 4.10(b)). Interestingly, it has the characteristic features of HHG spectra of atoms and molecules, viz. a rapid initial decrease in signal intensity followed by a plateau followed by a sharp fall in intensity.
Fig. 4.10 HHG spectra in a.u. plotted against harmonic order ($\omega/\omega_l$) for (a) the two-dimensional harmonic oscillator at $I = 2 \times 10^{17}$ W cm$^{-2}$ and the Henon-Heiles oscillator at (b) $I = 2 \times 10^{17}$, (c) $I = 5 \times 10^{16}$, (d) $I = 5 \times 10^{13}$ W cm$^{-2}$ respectively. The inset of (b) shows both even and odd harmonics. (c) shows only the 1st and 2nd harmonics while (d) shows only the first harmonic.
4.6 Conclusion

The present chapter demonstrates that an answer to the question whether a system exhibits quantum chaos is not straightforward. Since the HHO is a classically chaotic system, it has been shown by studying various time-dependent quantities that quantum chaos makes its presence felt when the system has reached the continuum, as speculated in Section 4.1. The electronic moving in an HH potential under intense laser fields requires a threshold intensity $2 \times 10^{17}$ Wcm$^{-2}$ in order to reach the continuum. This is in parallelism with atoms and molecules which also require a threshold intensity for ionization to occur. After 5 optical cycles, the electron moving in the HH potential in laser field intensity $2 \times 10^{17}$ Wcm$^{-2}$ reaches the continuum when the maximum value of laser electric field is attained. At the threshold intensity, various diagnostics like autocorrelation function, quantum “phase space” volume and quantum “phase space” trajectory give a consistent interpretation of quantum chaos. Below the threshold intensity quantum chaos is not manifested even in a nonintegrable system like the HH oscillator. Note that the two-dimensional HO does not display quantum chaos at the intensity $2 \times 10^{17}$ Wcm$^{-2}$.

The statistical analysis of the power spectrum is not found to be applicable in the quantum dynamical evolution of the time-dependent wavefunction in intense laser fields because of lack of statistical divergence. While *time-dependent signatures* indicate the presence of quantum chaos, *time-independent signatures* such as nearest neighbour spacing distribution and KS entropy do not yield unambiguous answers. Since both quantum and classical chaos are dynamical phenomena, more time-dependent (dynamical) signatures need to be devised.

On the basis of the distance function, overlap integral and divergence of “phase space” trajectories, the present work establishes the greater sensitivity of quantum motion to the Hamiltonian rather than to the initial quantum state for the classically chaotic HH oscillator.
Another outcome of the present work is that two-dimensional nonlinear oscillators are suitable model systems for studying laser-matter interactions as they mimic the behaviour of atoms and molecules in intense laser fields. The power and HHG spectra show similar characteristic features as atomic spectra under intense laser fields. The present study strengthens the speculation that atoms and molecules in intense laser fields should show quantum chaos.
References


