John Fitzgerald León Ocampo

November 19, 2020

Contents

Abstract

Chapter 1

Introduction

1.1 Introduction

Interesting direct manifestations of helium electronic correlations have been found in certain highly asymmetric doubly excited states which are associated with highly asymmetric classical configurations. The most interesting one is the frozen planet configuration (FPC), that is characterized by having a stable dynamics against autoionization. These regions can support quantum states called frozen planet states, that can transform under external electromagnetic periodic driving in quantum states that propagate along the classical trajectory without dispersion. Previous studies suggest that when a static electric field is added to the periodic driving an improvement on the stablization of the nondispersive wave packets can be achieved, this is a consequence of the effect this perturbation has on helium classical dynamics, confining its motion to a vicinity of the field polarization axis.

In this project, we will propose a method to investigate highly doubly excited states of helium under periodic driving and an external static electric field. The eigenvalues calculated from the Schrödinger equation for this system allow us to analize the stabilization properties of the states, such as their energies and their decay rates, from which the lifetime of the resonance states can be computed. The localization of the states can be analized using electronic projection of phase space, Husimi's densities. The latter properties will be tested as function of the static electric field.

This research work is intended to investigate the effect of a static electric field perturbation on frozen planet states and to analize their corresponding localization properties and lifetimes. Another pretention of this research project is to develop a similar analysis on nondispersive wave states, which are special states that emerge in the Floquet spectrum of the Hamiltonian, distinguished by their localization properties and by the fact that they follow concrete classical trajectories. Thus, Husimi's densities and stabilization properties for these wave packets will be studied in an external static perturbation.

1.2 Background

During the last decade, new techniques were created to develop coherent light sources that permitted to create ultra short pulses [**?**], down to a few tens of attoseconds ($1\,\mathrm{as} = 10^{-18}\,\mathrm{s}$), in the extreme ultraviolet spectral region. These pulses allowed

us to access a very fast dynamics, such as electron motion induced by light-matter interactions. Among many of the vast range of applications this impressive technologies has brought, one can mention the observation of the valence electron motion [?], the caracterization of electron wave packets in helium, monitoring the birth of autoionizing resonances [?] or the decay of a core vacancy [?]. In [?], it was shown that the photoionization delay, this is, the time for the photoelectron emission [?], depends on the excitation energy and the underlying ionic core structure in the attosecond time scale. On the other side the capacity to control electron dynamics is very important for some areas in chemistry, the reason for this is that time dependent electronic density is responsible for bond formation and bond breaking in molecules, this new research field is sometimes called attosecond chemistry [?, ?]. The full understanding of these experimental results requires background theoretical interpretations that can explain the intrinsic phenomenology and also be able to perform predictions. The interaction of light with the helium atom seems a promising way to follow this path, since it is the simplest example of a multielectron atom, but its dynamics is nonintegrable from both, classically and quantum mechanically [?].

The helium atom is a special case of a three body problem with a Coulomb interaction between the two electrons. The classical dynamics of this system is non-integrable, then its phase space is characterized by being chaotic in general [?], with only small regions of regular motion. An important consequence of the loss of integrability is the failure of first quantization attempts on the basis of Niels Bohr's postulates [?, ?, ?]. The correspondence between the chaotic-regular classical dynamics and the non-integrability of the quantum system [?] could be understood after the development of modern semiclassical theory in the second half of the 20th century [?, ?] and the subsequent semiclassical quantization of helium [?, ?].

Early observations made by Madden and Codling [?] demonstrated that two-electrons doubly excited states were highly correlated, and they cannot be described by a model based on independent particle quantum numbers, therefore doubly and highly excited states of helium are of great interest among experimentalists and theoreticians.

Strong electronic correlation have been found, specifically, in highly asymetrically excited states of unperturbed helium which are well localized around the classical frozen planet configuration [?, ?]. These states transform under near-resonantly periodic driving into nondispersive wave packets (NDWP), which are quantum objects that allow the possibility to localize the electronic population in phase space [?, ?], stablishing therefore a connection between a quantum wave function and its corresponding classical system [?]. The existence of two-electron NDWP was theoretically proven for one dimensional [?, ?], planar [?, ?, ?], and recently for the full three dimensional helium atom [?].

However, the localization properties of NDWP strongly depend on the stability of the dynamics of electron in the FPC, this is, the time that the configurations take before the atom ionization. This stabilization is rapidly broken when a small deviation of collinearity is present [?]. As it was proven in [?] a static electric field applied in the direction of the driving polarization axis can be used to enforce the stabilization of the driven classical configuration in all three spatial dimensions.

The nondispersive wave packets dynamics was already explained for the collinear helium case in [?]. There, it was shown that applying a static electric field with

a strength of up to 20 percent of the driving field amplitude, it was possible to modify the ionization rates in an apprecciable manner. Certain specific values for the static perturbation allow to modify the wavepacket's lifetime significantly, for example, from 1.8×10^6 to 1.0×10^4 field cycles . Nevertheless, ionization rates don't always decrease with increasing static field, instead, an erratic dependence between ionization rates and applied dc fields has been found, this behavior has not been explained yet.

It is important to realize the fact that even though bound states of any atom become quasi bound under the influence of a dc field, we are only working on the region of highly doubly excited states. Thus, on the basis of the previous cited phenomenona present on the classical FPC, we try to find appropiate values of the static field, such that ionization rates and other localizations properties get substantially modified.

The purpose of this research work is to investigate the effect of a static electric field perturbation on frozen planet states and to analize their localization properties and lifetimes. Thus we can exhibit the propiate values for the static field to get the best lifetimes of the resonances. These same properties will be determined for NDWP in the full three dimensional case in presence of the static perturbation.

Chapter 2

Theoretical Framework and numerical treatment of the problem

2.1 Hamiltonian

The helium atom can be considered as a three body Coulomb problem composed of two electrons and a massive nucleus. Neglecting relativistic effects and in the infinite nucleus mass approximation, the Hamiltonian in atomic units *Bulechleitner* for such a system can be described as

$$H_0 = \frac{\vec{P}_1^2}{2} + \frac{\vec{P}_2^2}{2} - \frac{2}{|\vec{r}_1|} - \frac{2}{|\vec{r}_2|} + \frac{1}{|\vec{r}_1 - \vec{r}_2|}, \tag{2.1}$$

where $\vec{r}_i = (x_i, y_i, z_i)$ and $\vec{P}_i = (p_{ix}, p_{iy}, p_{iz})$ are the position and momentum of particle $i = 1, 2$ respectively.

Since the Hamiltonian is time independent a solution of the form $\Psi(1,2) = \phi(x_1, x_2)\chi(s_1, s_2)$ is expected, this is, the product of a coordinate function and a spin state. This wave function has to be antysimmetric because of the Pauli exclusion principle.

For two electron atoms the spin basis has a four states basis $|S, M_s\rangle$, where S denotes the total and M_s its projection on the quantization axis. Therefore, we have three symmetric spin $S = 1$ states

$$|1,1\rangle = |\uparrow\uparrow\rangle\,, \quad |1,0\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle\right), \quad |1,-1\rangle = |\downarrow\downarrow\rangle\,, \tag{2.2}$$

and one antysimmetric spin $S = 0$ singlet state

$$|0,0\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right) \tag{2.3}$$

In order of the total wave function to be antysimmetric under exchange, we have two alteratives, spin singlet state must have symmetric spatial wave function; spin triplet states have antysimmetric spatial wave function.

2.2 Helium spectrum

When the interaction betweent the two electrons is neglected, one can write H as the sum of two one-particle Hamiltonians. Then, the Schrodinger equation can be posed as

$$[H(1) + H(2)] |\Psi\rangle = (E_{n_1} + E_{n_2}) |\Psi\rangle . \tag{2.4}$$

Therefore, the energy spectrum by continuum states above the single ionizations thresholds and a series of Rydberg bound states with energy given by

$$E_{n_1} + E_{n_2} = -\frac{2}{n_1^2} - \frac{2}{n_2^2}. \tag{2.5}$$

The Rydberg series labeled by the principal quantum number N converges to the single ionization threshold, $I_N = -\frac{2}{N^2}$, and all of these series converge to the double ionization threshold (DIT) at zero energy.

Once the electron electron interaction is taken into account some features in the spectrum are found. First, the energies of ionizanization thresholds remain unaffected by this term. The reason for this is the fact that when an electron gets ionized, their separation becomes significant enough to make the interaction between them negligible. Despite this, all other energies get considerable modified, and, particularly, bound states transforms into autoionizing resonance states embedded in the continua above the first ionization threshold.

2.3 Complex rotation method

´ To extract the energies and decay rates of resonance states, we use the complex coordinate rotation method [**?**, **?**].The complex rotation of any operator by an angle θ is given by the non-unitary operator

$$R(\theta) = \exp\left(-\theta \frac{\vec{r}.\vec{p} + \vec{p}.\vec{r}}{2}\right). \tag{2.6}$$

The transformations of the position and momentum operators are given by

$$\vec{r} \to H(\theta)\vec{r}H(-\theta) = e^{i\theta}\vec{r}, \tag{2.7}$$
$$\vec{p} \to H(\theta)\vec{p}H(-\theta) = e^{-i\theta}\vec{p}. \tag{2.8}$$

As a consequence, the rotated Hamiltonian operator is no longer Hermitian, therefore its eigenvalues are, in general, complex. The spectrum of the Hamiltonian has the following properties [**?**]:

- The bound spectrum of H_0 is invariant under the complex rotation.
- The continuum states are rotated by an angle of -2θ with the real axis, around the ionization threshold of the unrotated Hamiltonian.
- Provided large enough θ the resonance states are "exposed" in the lower half plane. The corresponding complex eigenvalues are $E_{i\theta} = E_i - i\Gamma_i/2$, where the real part corresponds to the energy of the resonance, and the imaginary part contains the decay rate Γ_i, which is the inverse of the resonant lifetime.

2.4 Expansion of the wave function

The quantum dynamics of the system is governed by the time independent Schrödinger equation

$$H_0\Psi(\vec{r}_1,\vec{r}_2) = E\Psi(\vec{r}_1,\vec{r}_2), \tag{2.9}$$

where H_0 is the unperturbed Hamiltonian. The solutions of this equation are expanded in a configuration interaction (CI) basis [**?**, **?**] for a given value of the total angular momentum L and its projection M on the Z-axis,

$$\Psi^{L,M}(\vec{r}_1,\vec{r}_2) = \sum_{l_1,l_2}\sum_{s}\sum_{n_1,n_2} \psi^{l_1,l_2,L,M,\epsilon_{12}}_{k_{1s},k_{2s},n_1,n_2}\beta^{l_1,l_2}_{n_1,n_2}\mathcal{A}F^{l_1,l_2,L,M}_{\kappa_{1s},\kappa_{2s},n_1,n_2}(\vec{r}_1,\vec{r}_2), \tag{2.10}$$

with

$$F^{l_1,l_2,L,M}_{\kappa_{1s},\kappa_{2s},n_1,n_2}(\vec{r}_1,\vec{r}_2) = \frac{S^{k_{1s}}_{n_1,l_1}(r_1)}{r_1}\frac{S^{k_{2s}}_{n_2,l_2}(r_2)}{r_2}\Lambda^{n_1,n_2}_{l_1,l_2}(\theta_1,\phi_1,\theta_2,\phi_2), \tag{2.11}$$

where $\psi^{l_1,l_2,L,M,\epsilon_{12}}_{k_{1s},k_{2s},n_1,n_2}$ are the expansion coefficients and $\beta^{l_1,l_2}_{n_1,n_2}$ controls the redundancy that might occur within the basis due to simmetrization. The symmetrization operator $\mathcal{A}$ takes the form

$$A = \frac{1+(-1)^{l_1+l_2-L}\epsilon_{12}P}{\sqrt{2}}, \tag{2.12}$$

where ϵ_{12} is the two dimensional levi civita symbol, and P exchanges simultaneously $(\lambda_1,\kappa_1,\mu_1,\nu_1)$ to $(\lambda_2,\kappa_2,\mu_2,\nu_2)$ therefore this operator allows to transform onto either singlet or tripet states.

The angular part of the wave function is an expansion in terms of bipolar spherical harmonics [**?**]

$$\Lambda^{n_1,n_2}_{l_1,l_2}(\theta_1,\phi_1,\theta_2,\phi_2) = \sum_{m_1,m_2}\langle l_1m_1l_2m_2|LM\rangle\, Y_{l_1,m_1}(\theta_1,\phi_1)Y_{l_2,m_2}(\theta_2,\phi_2) \tag{2.13}$$

The radial part of the wave function $S^k_{n,l}$ are the one-electron Coulomb-Sturmian functions which are the solution of the Sturm-Liouville eigenvalue problem [**?**]. The solutions are given by

$$S^{(k)}_{n,l}(r) = N^{(k)}_{n,l}r^{l+1}e^{-\kappa r}L^{(2l+1)}_{n-l-1}(2\kappa r), \tag{2.14}$$

where κ is a dilation parameter and $L^{(2l+1)}_{n-l-1}(2\kappa r)$ are Laguerre polynomials.

The reason for taking this particular basis is that the dilation parameter κ can be adjusted in order to improve convergence in a determined region of the spectrum, large values of this parameter imply Sturm-Coulomb functions with short extent in space, and small κ corresponds to large extent in space, respectively. The frozen planet states, which is the scope of reaserch as will be indicated, are highly asymmetric excited states, in this way the configuration is better described using large dilation parameter for the inner electron and a small parameter for the outer one. Using this approach allows us to reduce the basis size compared to other CI approaches because every electron must be described using a different set of dilaton parameter and Coulomb Sturmian functions, so it is possible to include just the fundamental basis elements.

2.5 Matrix form of the Schrodinger equation

The Hamiltonian in () can be split in order to write the Schrodinger equation in the form

$$(T+V+U)\left|\Psi\right\rangle = E\left|\Psi\right\rangle, \tag{2.15}$$

where T is the kinetic energy, T is the nucleus-electron interaction and U the electron-electron interaction. These quantities are

$$T = \frac{\vec{P}_1^2}{2} + \frac{\vec{P}_2^2}{2}, \quad V = -\frac{2}{|\vec{r}_1|} - \frac{2}{|\vec{r}_2|}, \quad U = \frac{1}{|\vec{r}_1 - \vec{r}_2|}. \tag{2.16}$$

To get the matrix elements of H we take the inner product between each side of () and the wave function (). This is equivalent to multiplicating from the left with $\beta^{l_1',l_2'}_{n_1',n_2'}\mathcal{A}'F^{l_1',l_2',L',M'}_{\kappa_{1s}',\kappa_{2s}',n_1',n_2'}(\vec{r}_1,\vec{r}_2)$ and then integrating over the whole space. This results in

$$\sum_{l_1,l_2}\sum_{s}\sum_{n_1,n_2}\beta^{l_1,l_2}_{n_1,n_2}\beta^{l_1',l_2'}_{n_1',n_2'}\psi^{l_1,l_2,L,M}_{k_{1s},k_{2s},n_1,n_2}(T+V+U)^{l_1,l_2,L,M,l_1',l_2',L',M'}_{k_{1s},k_{2s},n_1,n_2,\kappa_{1s}',\kappa_{2s}',n_1',n_2'} \tag{2.17}$$

$$= \sum_{l_1,l_2}\sum_{s}\sum_{n_1,n_2} ES^{l_1,l_2,L,M,l_1',l_2',L',M'}_{k_{1s},k_{2s},n_1,n_2,\kappa_{1s}',\kappa_{2s}',n_1',n_2'} \tag{2.18}$$

where the matrix elements of the Hamiltonian constituyents are

$$T^{l_1,l_2,L,M,l_1',l_2',L',M'}_{k_{1s},k_{2s},n_1,n_2,\kappa_{1s}',\kappa_{2s}',n_1',n_2'} = \int d\vec{r_1}d\vec{r_2}\mathcal{A}'F^{l_1',l_2',L',M'}_{\kappa_{1s}',\kappa_{2s}',n_1',n_2'}(\vec{r}_1,\vec{r}_2)T\mathcal{A}F^{l_1,l_2,L,M}_{\kappa_{1s},\kappa_{2s},n_1,n_2}(\vec{r}_1,\vec{r}_2), \tag{2.19}$$

$$V^{l_1,l_2,L,M,l_1',l_2',L',M'}_{k_{1s},k_{2s},n_1,n_2,\kappa_{1s}',\kappa_{2s}',n_1',n_2'} = \int d\vec{r_1}d\vec{r_2}\mathcal{A}'F^{l_1',l_2',L',M'}_{\kappa_{1s}',\kappa_{2s}',n_1',n_2'}(\vec{r}_1,\vec{r}_2)V\mathcal{A}F^{l_1,l_2,L,M}_{\kappa_{1s},\kappa_{2s},n_1,n_2}(\vec{r}_1,\vec{r}_2), \tag{2.20}$$

$$U^{l_1,l_2,L,M,l_1',l_2',L',M'}_{k_{1s},k_{2s},n_1,n_2,\kappa_{1s}',\kappa_{2s}',n_1',n_2'} = \int d\vec{r_1}d\vec{r_2}\mathcal{A}'F^{l_1',l_2',L',M'}_{\kappa_{1s}',\kappa_{2s}',n_1',n_2'}(\vec{r}_1,\vec{r}_2)U\mathcal{A}F^{l_1,l_2,L,M}_{\kappa_{1s},\kappa_{2s},n_1,n_2}(\vec{r}_1,\vec{r}_2), \tag{2.21}$$

and

$$S^{l_1,l_2,L,M,l_1',l_2',L',M'}_{k_{1s},k_{2s},n_1,n_2,\kappa_{1s}',\kappa_{2s}',n_1',n_2'} = \int d\vec{r_1}d\vec{r_2}\mathcal{A}'F^{l_1',l_2',L',M'}_{\kappa_{1s}',\kappa_{2s}',n_1',n_2'}(\vec{r}_1,\vec{r}_2)S\mathcal{A}F^{l_1,l_2,L,M}_{\kappa_{1s},\kappa_{2s},n_1,n_2}(\vec{r}_1,\vec{r}_2). \tag{2.22}$$

Thus, the Schrodinger equation () is formulated in its equivalent matrix form yields the generalized eigenvalue problem

$$\mathbf{H1} = E\mathbf{SSI}, \quad \mathbf{H} = \mathbf{T}+\mathbf{V}+\mathbf{U}, \tag{2.23}$$

where Si is the vector representation of the wave function and $\mathbf{H}$ is the matrix representation of the Hamiltonian.

Under complex rotation the Hamiltonian transforms as

$$\mathbf{H}_\theta = \mathbf{T}e^{-2i\theta} + \mathbf{V}e^{-i\theta} + \mathbf{U}e^{-i\theta}, \tag{2.24}$$

and the generalized eigenvalue problem as

$$\mathbf{H}_\theta \mathbf{1}_\theta = E_\theta \mathbf{S}\mathbf{1}_\theta, \tag{2.25}$$

where, again, $\mathbf{H}_\theta$ and $\mathbf{1}_\theta$ are the matrix and vector representations, respectively, of the Hamiltonian and the wave function in the complex rotated frame.

Despite integrals () can be computed analitically, these are evaluated using the Gauss-Laguerre integration method, since using Sturmian functions with several dilation parameters make the analitical calculation more difficult.

2.6 Helium atom under periodic driving and external static electric field

The Hamiltonian of the electromagnetically driven helium atom reads

$$H = H_0 + (F\cos\omega t + F_{\rm st})(\mathbf{e}_x).(\vec{r}_1 + \vec{r}_2), \tag{2.26}$$

where $F\cos\omega t$ is a linearly polarized driving at frequency ω and amplitude F ($\mathbf{e}_x$ represents the unit vector along the x-axis). Besides there is an additional static field strength $F_{\rm st}$.

A convenient way to handle a Hamiltonian with temporal periodicity $T = 2\pi/\omega$ is through Floquet theory [**?**, **?**]. In this formalism any solution of the Schrodinger equation can be expanded in a series of time periodic wave functions $|\phi_{\epsilon_i}(t)\rangle$,

$$|\psi(t)\rangle = \sum_i c_i e^{-\mathrm{i}\epsilon_\mathrm{i}\mathrm{t}} |\phi_{\epsilon_i}(t)\rangle , \tag{2.27}$$

with $|\phi_{\epsilon_i}(t+T)\rangle = |\phi_{\epsilon_i}(t)\rangle$. Here, the ϵ_i and $|\phi_{\epsilon_i}(t)\rangle$ are called quasienergy and floquet states, respectively. They satisfy the floquet eigenvalue equation

$$\left(H - \mathrm{i}\frac{\partial}{\partial \mathrm{t}}\right) |\phi_{\epsilon_i}(t)\rangle = \epsilon_i |\phi_{\epsilon_i}(t)\rangle . \tag{2.28}$$

Due to the time periodicity, Floquet states can be expanded in Fourier series

$$|\phi_{\epsilon_i}(t)\rangle = \sum_{k=-\infty}^{\infty} e^{-\mathrm{ik}\omega\mathrm{t}} \left|\phi_{\epsilon_i}^k\right\rangle . \tag{2.29}$$

When substituted this expansion into the eigenvalue equation (2.28) with H as in (2.26), one gets the eigenvalue problem

$$(H_0 + F_{\rm st}(x_1+x_2) - k\omega)\left|\phi_{\epsilon_i}^k\right\rangle + \frac{F}{2}(x_1+x_2)\left(\left|\phi_{\epsilon_i}^{k+1}\right\rangle + \left|\phi_{\epsilon_i}^{k-1}\right\rangle\right) = \epsilon_i \left|\phi_{\epsilon_i}^k\right\rangle , \tag{2.30}$$

in the position Gauge. Time dependence has been eliminated at the cost of adding a new quantum number k. For a classical field in the limit of large number of photons there is a one-to-one correspondence between the quasienergy spectrum of the Floquet eigenvalue equation and the energy spectrum on an atom dressed by a

quantum radiation field [**?**, **?**]. In the semiclassical limit the number k counts the number of photons exchanged between the atom and the field.

Equation (2.30) can also be written in matrix form given by

$$\mathbf{A}\phi_i = \epsilon_i\phi_i, \qquad \mathbf{A} = \mathbf{H}_0 - k\omega\mathbb{I} + \mathbf{F}\mathrm{st} + \mathbf{F}, \tag{2.31}$$

where ϕ_i is the column vector representing $\left|\phi^k_{\epsilon_i}\right\rangle$ and $\mathbf{F}$st and $\mathbf{F}$ are the matrix representations of the dipole parts associated to the static and periodic fields, respectively (2.30).

The matrix $\mathbf{A}$ is partitioned in blocks labeled by its k components. The matrix H_0 has a block diagonal structure, and since we are working in the Sturmian basis, this matrix is very dense. The matrices $\mathbf{F}$ and $\mathbf{F}_{st}$ are symmetric in the gauge we have chosen and they have a block structure defined by the selection rules $\Delta k = \pm 1$ and $\Delta L = \pm 1$.

For the numerical implementation the basis has to be truncated. This is done by

$$k_{\min} \leq k \leq k_{\max}, \qquad L = 0, 1, ..., L_{\max}. \tag{2.32}$$

Using the basis expansion (6) for the full Floquet system leads to huge matrices in the eigenvalue problem (16). Even for a small number of Floquet blocks and angular momenta the matrix representation will require storage space of hundres of Gb. We, therefore, propose to represent the eigenvalue problem of the full system in the atomic basis, that is, in the basis of eigenstates of $\mathbf{H}_0$,

$$\left\{\left|\phi_i^{L,k}\right\rangle\right\}, \qquad \left|\phi_i^{L,k}\right\rangle = \left|\phi_i^L\right\rangle \otimes |k\rangle\,. \tag{2.33}$$

The tensor product is motivated by the identification of the floquet quauntum number k with the classical number of photon exchanged by the atom and the field. The states $\left|\phi_i^L\right\rangle$ are the solutions of the time independent Schrödinger equation.

$$H_0\left|\phi_i^L\right\rangle = \epsilon_i^L\left|\phi_i^L\right\rangle. \tag{2.34}$$

Using this basis, equation (2.31) transforms into the matrix equation

$$\tilde{\mathbf{A}}\tilde{\phi}_i = \epsilon_i\tilde{\phi}_i, \qquad \tilde{\mathbf{A}} = \mathbf{h}_0 - k\omega\mathbb{I} + \mathbf{F}\mathrm{st} + \mathbf{F}, \tag{2.35}$$

where $\tilde{\phi}_i$ is the vector representation of $\left|\phi_i^{L,k}\right\rangle$, and $\mathbf{h}_0$ is the diagonal matrix containing the eigenvalues of $\mathbf{H}_0$. The great advantage here is that the matrix form of $\tilde{\mathbf{A}}$ is much sparser than $\mathbf{A}$, which is mandatory for a suitable numerical implementation as can be see in Figure **??**. The diagonalization of the matrix equation is performed using Lanczos algorithm [**?**].

Chapter 3

Unperturbed frozen planet states and non dispersive wave packets

3.1 The classical frozen planet configuration of Helium

The classical dynamics of the helium atom under the influence periodic and static electromagnetic perturbation is described with the Hamiltonian

$$H = \frac{\vec{P}_1^2}{2} + \frac{\vec{P}_2^2}{2} - \frac{2}{|\vec{r}_1|} - \frac{2}{|\vec{r}_2|} + \frac{1}{|\vec{r}_1 - \vec{r}_2|} + (F\cos\omega t + F_{\rm st})(\mathbf{e}_x).(\vec{r}_1 + \vec{r}_2), \qquad (3.1)$$

where $\vec{r}_i = (x_i, y_i, z_i)$ and $\vec{P}_i = (p_{ix}, p_{iy}, p_{iz})$ are the position and momentum of particle $i = 1, 2$ respectively. Besides $F\cos\omega t$ is a linearly polarized driving at frequency ω and amplitude F ($\mathbf{e}_x$ represents the unit vector along the x-axis). Besides there is an additional static field strength $F_{\rm st}$. The classical dynamics goberned by this Hamiltonian is invariant under the scaling transformations

$$\vec{r}_i \to N\vec{r}_i \quad (i = 1, 2), \qquad (3.2)$$

$$\vec{p}_i \to N^{-2}\vec{p}_i \quad (i = 1, 2), \qquad (3.3)$$

$$t \to N^3 t, \qquad (3.4)$$

$$\omega \to N^{-3}\omega, \qquad (3.5)$$

$$F \to N^{-4}F, \qquad (3.6)$$

$$F_{st} \to N^{-4}F_{st}, \qquad (3.7)$$

$$H \to N^{-2}H, \qquad (3.8)$$

where N is an arbitrary, real positive number. Due to this invariance the intrinsic quantities depend only on the action integral over one cycle of the Kepler oscillation of the inner electron, where x_2 and p_2 are its position and momentum, respectively.

Un this work we are interested in a particular solution of the classical equations of motion, which is called the frozen planet configuration (FPC) [**?**, **?**]. This is an asymmetric configuration where both electrons are located on the same side of the nucleus. While the inner electron oscillates rapidly in extremely eccentric Kepler

trajectories around the nucleus, the outer electron remains nearly frozen around some equilibrium distance. The importances of this configuration lies in the fact that it is classical stable under autoionization and defines a relatively large region of regular motion in the chaotic unperturbed phase space of helium.

The regularity of the classical FPC is visualized in a Poincaré surface section. The necesity of a method like this stems from the fact that the driven helium dynamics takes place in a five dimensional phase-space, spanned by the positions and momenta of the electrons, therefore a reduction of the dimensionality is a practical choice to be made. Here, each point (x_1, p_1) represents the position and momenta of the outer electron when the inner electron collides with the nucleus. In the case of collinear the phase-space contains a large region of bounded motion, as can be seen in figure.

When the driving field is switched on, the phase space turns mixed regular-chaotic. Now, the phase space exhibits an intrinsic regular region centered around the equilibrium position of the configuration [**?**], which comes from the interelectronic perturbation, besides there is an additional regular island immersed in the chaotic sea which is a consequence of the non linear resonances between the external driving and the unperturbed oscillation of the unperturbed oscillation of the outer electron. When the phase of the periodic driving is varied as time increases, the intrinsic island remains at rest, while the field-induced resonance island oscillates around the intrinsic island with the same period of the driving field.

There is a notable difference in the time scales for both electrons oscillation modes, being the Keppler oscillations of the inner electron almost 15 times faster than the slow oscillation of the outer electron around its equilibrium distance. The separation of time scales allows one to treat the outer electron dynamics in the framework of adiabatic approximations, and, consequently, to define an effective potential to describe its slow dynamics. From the shape of this potential, it is possible to obtain the natural scale for the field strength, which indicates the maximum field that can be applied to the configuration without ionizing it, and the frequency scale, that is given bye the curvature of the potential at its minimun. These scaled quantities in atomic units are given by

$$x_{\min} = 2.6S^2, \tag{3.9}$$

$$\omega_I = 0.3S^{-3}, \tag{3.10}$$

$$F_I = 0.3S^{-4}, \tag{3.11}$$

where

$$S = \frac{1}{2\pi}\int p_2 dx_2. \tag{3.12}$$

In a quantum description the action variable is replaced by the principal number of the inner electron, thus the scale quantitites adopt the form of

$$x_{\min} = 2.6N^2, \tag{3.13}$$

$$\omega_I = 0.3N^{-3}, \tag{3.14}$$

$$F_I = 0.3N^{-4}. \tag{3.15}$$

In this description of the problem, there are eigenstates of the spectrum that are well localized along the frozen planet orbit, this is due to the large phase space volume occupied by the stability region of the FPC . These states are the *frozen planet states* (FPS), whose existence has been demonstrated on 1D [**?**], 2D [**?**] and 3D helium [**?**]. FPS can be recognized by proper analysis of their spectral properties sucha as decay rates and the expectation value of $\cos\theta_{12}$. Since these states are localized along a collinear configuration the expectation value of $\cos\theta_{12}$ is expected to be approximately one. Besides, they ought to have long lifetime compared to other resonances in the same energy region which is the small decay rate condition. Nevertheless, none of these features allows to completely caractize FPS, then the localization properties of electronic density in configuration and phase space have to be considered.

3.1.1 Identification of frozen planet states

Frozen planet states are eigenstates in the helium spectrum that are localized in the stability region of the FPC. Hence, given a collinear configuration one main feature for a state to be considered a FPS is that the expectation value of the cosine of the angle θ_{12} between the radius vector $\vec{r_1}$ and $\vec{r_2}$ has to be close to unity. Moreover, since these resonance are expected to posses a large stability against autoionization, long lifetimes, or, equivalently, small decay rates are needed. Nevertheless, these two requirements are not sufficient to guarantee the FPS nature of a given state. Instead, frozen planet states can be identificated unambiguously by their localization properties in configuration and phase space. Nevertheless, this can´t be performed directly due to the dimension of the spaces involved (e.g six for configuration space). Then, visualization requires projection of the probability density. Thus, two projections of the electronic densities are used in configuration space, these are, conditional probability distributions for θ_{12} or one-electron probability densities, whereas, for phase space projection we use Husimi densities.

3.2 Frozen planet states in N=6 helium spectrum

In this section, we present the principal characteristics of frozen planet states lying below the $N = 6$ ionization threshold. These states are of major interest, as we will see later, in the determination and characterization of non dispersive wave packets.

In order to characterize the FPS we first have to determine the section of the spectrum corresponding to states converging to $N = 6$ ionization threshold. To find these states (figure), we diagonilize the hamilotinan in (), where we adjust the parameter, to find only states in the energy region of interest. FPS are characterized by their expectation value and their low decay rates. Figure () shows the resonance lying above $N = 6$ ionization threshold, with the potential FPS identified with red circles. It is seen that these states match the necesary conditions. Energies, decay rates and cos for the first members of these series are shown in table.

To completely categorize these states as FPS we still have observe their localization properties. In figure () the conditional probability densities for the first two states in the series are shown. For stae It is observed that the maximum probabily for the inner electron is close to the necleus at , whereas for the outer electron, labeled bye , the maximum probability is at .

On the other hand Figure 3.6 displays the Husimi distributions of the first four 1 S FPS converging to the 6th series for planar and three-dimensional helium. Comparing these plots with the classical configuration, we observe that the maximum of the probability density of the ground state is localized at the equilibrium position of the outer electron in the classical FPC given by Eq. (3.2). On the other hand, excited FPS are localized along periodic orbits with higher energy of the classical configuration.

Chapter 4

Non dispersive wave packets

Usually wave packets spread as time evolves. Nevertheless, it has been shown that, in some quantum systems such as one-electron Rydberg systems, this spreading could be overcome with non linear resonancses between the system and an external periodic driving. This procedure allow to create Non dispersive wave packets (NDWP), which are well localized and follow a classical periodic orbit without spreading. The dynamics of frozen planet states gives rise to wave packet that propagate along Keppler trajectories of arbitrry eccentricity and whose lifetime can be considerably long compared to any other state in the spectrum.

In section () we will review generalities of the frozen planet configuration under periodic driving. Then, section 2 reviews the characterization of non dispersive wave packets for helium. In the last section of this chapter, we shall review how an aditional static field perturbation can modify the fundamental properties of nondispersive wave packets

4.1 Classical frozen planet configuration under periodic driving

Now we consider the FPC in its driven case, this is, when the field frequency is chosen near resonant with the frequency of the periodic orbit.

The evolution of this system takes place in a five dimensional phase space spanned by the positions and momenta of the electron and the ωt of the driving field. Therefore, a complete visualization is not possible. However, for w and , the separation between the rapid Keppler oscillations of the inner electron and the slow oscillation of the outer electron, makes possible to map the phase space structure onto a two dimensiones surface, by a two-step section Poincaré method. In the first step, the position and momenta (x_1, p_1) of the outer electron are plotted every time the inner electron reaches the nucleus, this eliminates the fast Kepler oscillations. Then, these points are connected by cubic interpolation to yield a continues trajectory that is used to perform a second Poincaré section by fixing the phase of the driving field $\omega t = \phi_0$.

Figure () shows the two step Poincaré section obtained for fixed field frequency $\omega = 0.3N^3$a.u., and field amplitude $F = 0.005N^{-3}$a.u., and field phases $\omega t = 0$, $\omega t = \frac{\pi}{2}$ and $\omega t = \pi$.

Chapter 5

Conclusions

5.1 Section 1

Lorem ipsum dolor sit amet, consectetuer adipiscing elit. Ut purus elit, vestibulum ut, placerat ac, adipiscing vitae, felis. Curabitur dictum gravida mauris. Nam arcu libero, nonummy eget, consectetuer id, vulputate a, magna. Donec vehicula augue eu neque. Pellentesque habitant morbi tristique senectus et netus et malesuada fames ac turpis egestas. Mauris ut leo. Cras viverra metus rhoncus sem. Nulla et lectus vestibulum urna fringilla ultrices. Phasellus eu tellus sit amet tortor gravida placerat. Integer sapien est, iaculis in, pretium quis, viverra ac, nunc. Praesent eget sem vel leo ultrices bibendum. Aenean faucibus. Morbi dolor nulla, malesuada eu, pulvinar at, mollis ac, nulla. Curabitur auctor semper nulla. Donec varius orci eget risus. Duis nibh mi, congue eu, accumsan eleifend, sagittis quis, diam. Duis eget orci sit amet orci dignissim rutrum.

Nam dui ligula, fringilla a, euismod sodales, sollicitudin vel, wisi. Morbi auctor lorem non justo. Nam lacus libero, pretium at, lobortis vitae, ultricies et, tellus. Donec aliquet, tortor sed accumsan bibendum, erat ligula aliquet magna, vitae ornare odio metus a mi. Morbi ac orci et nisl hendrerit mollis. Suspendisse ut massa. Cras nec ante. Pellentesque a nulla. Cum sociis natoque penatibus et magnis dis parturient montes, nascetur ridiculus mus. Aliquam tincidunt urna. Nulla ullamcorper vestibulum turpis. Pellentesque cursus luctus mauris.

Nulla malesuada porttitor diam. Donec felis erat, congue non, volutpat at, tincidunt tristique, libero. Vivamus viverra fermentum felis. Donec nonummy pellentesque ante. Phasellus adipiscing semper elit. Proin fermentum massa ac quam. Sed diam turpis, molestie vitae, placerat a, molestie nec, leo. Maecenas lacinia. Nam ipsum ligula, eleifend at, accumsan nec, suscipit a, ipsum. Morbi blandit ligula feugiat magna. Nunc eleifend consequat lorem. Sed lacinia nulla vitae enim. Pellentesque tincidunt purus vel magna. Integer non enim. Praesent euismod nunc eu purus. Donec bibendum quam in tellus. Nullam cursus pulvinar lectus. Donec et mi. Nam vulputate metus eu enim. Vestibulum pellentesque felis eu massa.

Quisque ullamcorper placerat ipsum. Cras nibh. Morbi vel justo vitae lacus tincidunt ultrices. Lorem ipsum dolor sit amet, consectetuer adipiscing elit. In hac habitasse platea dictumst. Integer tempus convallis augue. Etiam facilisis. Nunc elementum fermentum wisi. Aenean placerat. Ut imperdiet, enim sed gravida sollicitudin, felis odio placerat quam, ac pulvinar elit purus eget enim. Nunc vitae tortor. Proin tempus nibh sit amet nisl. Vivamus quis tortor vitae risus porta

vehicula.

Fusce mauris. Vestibulum luctus nibh at lectus. Sed bibendum, nulla a faucibus semper, leo velit ultricies tellus, ac venenatis arcu wisi vel nisl. Vestibulum diam. Aliquam pellentesque, augue quis sagittis posuere, turpis lacus congue quam, in hendrerit risus eros eget felis. Maecenas eget erat in sapien mattis porttitor. Vestibulum porttitor. Nulla facilisi. Sed a turpis eu lacus commodo facilisis. Morbi fringilla, wisi in dignissim interdum, justo lectus sagittis dui, et vehicula libero dui cursus dui. Mauris tempor ligula sed lacus. Duis cursus enim ut augue. Cras ac magna. Cras nulla. Nulla egestas. Curabitur a leo. Quisque egestas wisi eget nunc. Nam feugiat lacus vel est. Curabitur consectetuer.

Suspendisse vel felis. Ut lorem lorem, interdum eu, tincidunt sit amet, laoreet vitae, arcu. Aenean faucibus pede eu ante. Praesent enim elit, rutrum at, molestie non, nonummy vel, nisl. Ut lectus eros, malesuada sit amet, fermentum eu, sodales cursus, magna. Donec eu purus. Quisque vehicula, urna sed ultricies auctor, pede lorem egestas dui, et convallis elit erat sed nulla. Donec luctus. Curabitur et nunc. Aliquam dolor odio, commodo pretium, ultricies non, pharetra in, velit. Integer arcu est, nonummy in, fermentum faucibus, egestas vel, odio.

Sed commodo posuere pede. Mauris ut est. Ut quis purus. Sed ac odio. Sed vehicula hendrerit sem. Duis non odio. Morbi ut dui. Sed accumsan risus eget odio. In hac habitasse platea dictumst. Pellentesque non elit. Fusce sed justo eu urna porta tincidunt. Mauris felis odio, sollicitudin sed, volutpat a, ornare ac, erat. Morbi quis dolor. Donec pellentesque, erat ac sagittis semper, nunc dui lobortis purus, quis congue purus metus ultricies tellus. Proin et quam. Class aptent taciti sociosqu ad litora torquent per conubia nostra, per inceptos hymenaeos. Praesent sapien turpis, fermentum vel, eleifend faucibus, vehicula eu, lacus.

Appendices

Lorem ipsum dolor sit amet, consectetuer adipiscing elit. Ut purus elit, vestibulum ut, placerat ac, adipiscing vitae, felis. Curabitur dictum gravida mauris. Nam arcu libero, nonummy eget, consectetuer id, vulputate a, magna. Donec vehicula augue eu neque. Pellentesque habitant morbi tristique senectus et netus et malesuada fames ac turpis egestas. Mauris ut leo. Cras viverra metus rhoncus sem. Nulla et lectus vestibulum urna fringilla ultrices. Phasellus eu tellus sit amet tortor gravida placerat. Integer sapien est, iaculis in, pretium quis, viverra ac, nunc. Praesent eget sem vel leo ultrices bibendum. Aenean faucibus. Morbi dolor nulla, malesuada eu, pulvinar at, mollis ac, nulla. Curabitur auctor semper nulla. Donec varius orci eget risus. Duis nibh mi, congue eu, accumsan eleifend, sagittis quis, diam. Duis eget orci sit amet orci dignissim rutrum.

Nam dui ligula, fringilla a, euismod sodales, sollicitudin vel, wisi. Morbi auctor lorem non justo. Nam lacus libero, pretium at, lobortis vitae, ultricies et, tellus. Donec aliquet, tortor sed accumsan bibendum, erat ligula aliquet magna, vitae ornare odio metus a mi. Morbi ac orci et nisl hendrerit mollis. Suspendisse ut massa. Cras nec ante. Pellentesque a nulla. Cum sociis natoque penatibus et magnis dis parturient montes, nascetur ridiculus mus. Aliquam tincidunt urna. Nulla ullamcorper vestibulum turpis. Pellentesque cursus luctus mauris.

Nulla malesuada porttitor diam. Donec felis erat, congue non, volutpat at, tincidunt tristique, libero. Vivamus viverra fermentum felis. Donec nonummy pellentesque ante. Phasellus adipiscing semper elit. Proin fermentum massa ac quam. Sed diam turpis, molestie vitae, placerat a, molestie nec, leo. Maecenas lacinia. Nam ipsum ligula, eleifend at, accumsan nec, suscipit a, ipsum. Morbi blandit ligula feugiat magna. Nunc eleifend consequat lorem. Sed lacinia nulla vitae enim. Pellentesque tincidunt purus vel magna. Integer non enim. Praesent euismod nunc eu purus. Donec bibendum quam in tellus. Nullam cursus pulvinar lectus. Donec et mi. Nam vulputate metus eu enim. Vestibulum pellentesque felis eu massa.

Quisque ullamcorper placerat ipsum. Cras nibh. Morbi vel justo vitae lacus tincidunt ultrices. Lorem ipsum dolor sit amet, consectetuer adipiscing elit. In hac habitasse platea dictumst. Integer tempus convallis augue. Etiam facilisis. Nunc elementum fermentum wisi. Aenean placerat. Ut imperdiet, enim sed gravida sollicitudin, felis odio placerat quam, ac pulvinar elit purus eget enim. Nunc vitae tortor. Proin tempus nibh sit amet nisl. Vivamus quis tortor vitae risus porta vehicula.

Fusce mauris. Vestibulum luctus nibh at lectus. Sed bibendum, nulla a faucibus semper, leo velit ultricies tellus, ac venenatis arcu wisi vel nisl. Vestibulum diam. Aliquam pellentesque, augue quis sagittis posuere, turpis lacus congue quam, in hendrerit risus eros eget felis. Maecenas eget erat in sapien mattis porttitor. Vestibulum porttitor. Nulla facilisi. Sed a turpis eu lacus commodo facilisis. Morbi

fringilla, wisi in dignissim interdum, justo lectus sagittis dui, et vehicula libero dui cursus dui. Mauris tempor ligula sed lacus. Duis cursus enim ut augue. Cras ac magna. Cras nulla. Nulla egestas. Curabitur a leo. Quisque egestas wisi eget nunc. Nam feugiat lacus vel est. Curabitur consectetuer.

Suspendisse vel felis. Ut lorem lorem, interdum eu, tincidunt sit amet, laoreet vitae, arcu. Aenean faucibus pede eu ante. Praesent enim elit, rutrum at, molestie non, nonummy vel, nisl. Ut lectus eros, malesuada sit amet, fermentum eu, sodales cursus, magna. Donec eu purus. Quisque vehicula, urna sed ultricies auctor, pede lorem egestas dui, et convallis elit erat sed nulla. Donec luctus. Curabitur et nunc. Aliquam dolor odio, commodo pretium, ultricies non, pharetra in, velit. Integer arcu est, nonummy in, fermentum faucibus, egestas vel, odio.

Sed commodo posuere pede. Mauris ut est. Ut quis purus. Sed ac odio. Sed vehicula hendrerit sem. Duis non odio. Morbi ut dui. Sed accumsan risus eget odio. In hac habitasse platea dictumst. Pellentesque non elit. Fusce sed justo eu urna porta tincidunt. Mauris felis odio, sollicitudin sed, volutpat a, ornare ac, erat. Morbi quis dolor. Donec pellentesque, erat ac sagittis semper, nunc dui lobortis purus, quis congue purus metus ultricies tellus. Proin et quam. Class aptent taciti sociosqu ad litora torquent per conubia nostra, per inceptos hymenaeos. Praesent sapien turpis, fermentum vel, eleifend faucibus, vehicula eu, lacus.