Resonant dynamics of H atom in elliptically polarized microwave field

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The dynamics of Rydberg states of atomic hydrogen driven by elliptically polarized microwaves of frequency fulfilling 2:1 classical resonance condition is investigated both semiclassically and quantum mechanically in a simplified two-dimensional model of an atom. Semiclassical results for quasienergies of the system are shown to be in a good agreement with exact quantum data. The structures of the quantum states are found to reflect the underlying classical dynamics; especially we show the existence of nonspreading wavepackets propagating on elliptical trajectories.

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The pioneering experiment on Rydberg hydrogen (H) atoms ionization in microwave fields [1] and a subsequent interpretation of its results in terms of the chaotic classical dynamics [2] opened up a new possibility to study dynamics of atomic systems whose classical counterpart reflects a chaotic behavior (for review see [3]). Quantum mechanically a simplified 2D model of an atom. Semiclassically, all of them are supported by the principal resonance island, i.e. \( \omega/\omega_K = 1 \). The wavepackets built inside a higher primary resonance island have been discussed by Holthaus [1] in model one dimensional (1D) systems and to a realistic H atom in LP microwaves [2].

Theoretical detection of such interesting states obviously needs reliable semiclassical methods. In the LP case the 1D approximation to the dynamics allowed to quantize resonant states exploring Mathieu equation [13]. In the CP case passing to the rotating frame removes explicit time dependence of the system and the harmonic approximation around the stable fixed point gives the appropriate semiclassical predictions [4]. For the realistic LP problem as well as for arbitrary elliptical polarization (EP) in two dimensional (2D) model of an atom the semiclassical method based on the Born–Oppenheimer approximation allows to describe resonant dynamics [12,14].

This brief report extends the previous analysis of the EP case [14], limited to the principal resonance island, to a higher primary resonance. We describe the full dynamics of quasienergies as a function of the microwave ellipticity for the 2:1 resonance case, concentrating in particular on nonspreading wavepackets. The experimental data for the EP problem are available in the range of the scaled frequency, \( \omega_0 = \omega/\omega_K \), up to 1.4 [15]. Thus a theoretical description of the system for \( \omega_0 = 2 \) should be able to be compared with an experiment probably in the immediate future.

As previously [14], we treat semiclassically and quantum mechanically a simplified 2D model of an atom. Study of such simplified models have been most successful in the past both for the LP problem (where 1D model has been a main source of quantum results for a long time [3]) and for the CP one where also the 2D, polarization plane restricted model has been utilized [8] (and references therein).

The Hamiltonian of the hydrogen 2D model atom driven by an elliptically polarized electromagnetic field reads in the dipole approximation (in atomic units)

\[
H = \frac{p_x^2 + p_y^2}{2} - \frac{1}{r} + F(x \cos \omega t + \alpha y \sin \omega t),
\]

where \( r = \sqrt{x^2 + y^2} \) while \( F \) and \( \omega \) denote the amplitude and the frequency of the microwave field, respectively. \( \alpha \) defines the ellipticity of the microwaves, with \( \alpha = 0 \) (\( \alpha = 1 \)) corresponding to a LP (CP) limiting case.
Using the Floquet theorem \[16\], the solution of the quantum problem is equivalent to diagonalizing the Floquet Hamiltonian, \( \mathcal{H} = H - i\partial_t \) with time-periodic boundary conditions, getting the eigenvalues (quasienergies) and time periodic eigenstates (Floquet states). The semiclassical quantization of resonant dynamics, based on a prescription of \[11\], closely resembles the similar procedure applied by us recently for the LP and EP case \[12,14\]. The method bases on passing to the extended phase space by defining the momentum \( p_t \) conjugate to the \( t \) (time) variable which yields the new Hamiltonian, \( \mathcal{H} = H + p_t \). The quasi-energies of the system will be then the quantized values of \( \mathcal{H} \). As the next step we express the Hamiltonian in action-angle variables of the unperturbed Coulomb problem \[13\]. For the 2D model atom those are e.g. the canonically conjugate pairs \((J, \theta)\) and \((L, \phi)\). \( J \) is the principal action (corresponding to the principal quantum number, \( n_0 \)). The conjugate angle, \( \theta \), determines the position of the electron on its elliptic trajectory and depends linearly on time, \( \theta \sim \omega_K t \), for an unperturbed atom. \( L \) is the angular momentum (equal to \( L_z \) for the 2D motion in the \( x-y \) plane) while \( \phi \) is the conjugate angle (the angle between the Runge-Lenz vector and the \( x \)-axis, i.e. the main axis of the polarization ellipse).

Considering the case of the resonant driving, i.e. \( \omega_0 = s \), we apply the secular perturbation theory \[17\] to average over the nonresonant terms which yields the approximate resonant Hamiltonian of the form

\[
\mathcal{H}_r = -\frac{1}{2sJ^2} - \omega J + \text{FT}(L, \phi; \alpha) \cos[\hat{\theta} - \beta(L, \phi; \alpha)] + \hat{p}_t
\]

(2)

where

\[
\hat{\theta} = s\theta - \omega t, \quad \hat{J} = \frac{J}{s}, \quad \hat{p}_t = p_t + \omega \hat{J}.
\]

(3)

The explicit form of \( \Gamma(L, \phi; \alpha) \) and \( \beta(L, \phi; \alpha) \) is given by Eq. (2.17) of \[13\].

The last stage is to quantize the system using the approximate Hamiltonian, Eq. (5). Trivial quantization of \( \hat{p}_t \), exploring the time periodicity the system (note that in the rotating frame, defined by Eq. (3)), the time period is \( \tau = 2\pi/\omega \), yields additive terms \( k\omega/s \) to quasienergies, where \( k \) is an integer number \[11,12\]. Thus the spectrum associated with states localized in the \( s:1 \) resonance island repeats itself along the energy axis at distances \( \omega/s \) \[11,12\]. The radial motion in \((J, \theta)\) space is much faster than the angular motion in \((L, \phi)\) space \[14,15\]. Hence, in the spirit of the Born-Oppenheimer approximation, one may first quantize the fast radial motion, keeping \( L \) and \( \phi \) fixed, then pass to the quantization of the slow angular motion. In fact, because of the specific form of Eq. (2), the order of the quantizations does not matter, one may quantize first the slow motion yielding discrete values of \( \Gamma \) and then go to quantization of the fast motion \[12,14\]. We consider below the \( s = 2 \) resonance as a generic example.

We are interested in strongly localized, wavepacket-like states lying close to the center of the resonance island in fast \((\hat{J}, \hat{\theta})\) variables. Previously \[12,14\] we have used pure WKB quantization for that motion which, however, works poorly when the island size is small. Such is a case for \( s > 1 \) resonances. Thus we improve the procedure and expand the principal action to the second order around the center of the resonance island defining \( I = \hat{J} - (s^2\omega)^{-1/3} \), Eq. (6) gives then a standard pendulum Hamiltonian with the island size given by \( \sqrt{IT} \). To ensure a maximal radial localization of the electron we consider then a ground state of the fast motion by taking appropriate quantum eigenvalues (as given by Mathieu equation solutions \[20\]), see similar treatment for 1D systems \[14,13\].

The slow angular motion is determined by constant values of \( \Gamma(L, \phi; \alpha) \). To compare the semiclassical predictions to the exact quantum calculations, we consider the \( n_0 = 42 \) manifold of our 2D model atom. For resonant driving we take the microwave frequency to be \( \omega = 2\omega_K = 2/(n_0 + 1/2)^2 \), i.e. \( \omega_0 = 2 \). Note that in the 2D model the effective principal quantum number is half integer. Fig. 4 shows values of \( \Gamma \) as a function of the scaled angular momentum, \( L_0 = L/(n_0 + 1/2) \), and the \( \phi \) angle for two different values of the field ellipticity, \( \alpha \). Semiclassically quantized contours, reflecting slow evolution of the electronic ellipse, are also presented in the figure. Those of them which are localized around extrema of \( \Gamma \) correspond to states with well defined electronic ellipse. Size of the resonance island in the \((J, \theta)\) space depends on value of \( \Gamma \), thus, only states localized around the maxima will show strong radial localization too. Note that \( \Gamma \) is equal to zero for circular orbits, i.e. for \( L_0 = \pm 1 \). It is obvious because circular motion is purely harmonic and no primary resonance exists except the 1:1 case.

Consider level dynamics with a change of the field ellipticity. Fig. 4 shows semiclassical and numerical results for quasienergies corresponding to the resonantly driven \( n_0 = 42 \) manifold as a function of \( \alpha \), for the scaled field amplitude \( F_0 = F/(n_0 + 1/2)^3 \approx 0.03 \). The nice quantitative agreement between semiclassics and numerics is achieved (except in the region of broad avoided crossings with other levels – partners in the crossing are not plotted for clarity) with significant improvement over earlier approach \[14\] for small \( \Gamma \) region.

This level dynamics is easy to understand by inspection of angular motion change with \( \alpha \). Fig. 5. Semiclassically, for \( \alpha = 0 \), all states are degenerate because of the symmetry of the \((L, \phi)\) space with respect to the \( L_0 = 0 \) axis. The highest degeneracy exists for librational states situated in elliptical islands around \( L_0 \approx \pm 0.6 \), \( \phi = 0, \pi \) because the islands are identical. With a small change of \( \alpha \), values of \( \Gamma \) corresponding to negative \( L_0 \) become
smaller while those corresponding to positive $L_0$ become greater. Quasienergy levels simply follow the increase or decrease of $\Gamma$, i.e. the greater value of $\Gamma$ the higher the corresponding quasienergy level. Thus, for $\alpha > 0$, the degeneracy of many states is removed. Still for $\alpha < 0.17$ there exist three pairs of identical elliptical islands which support identical semiclassical states, see Fig. 3.

With further increase of $\alpha$ the islands situated around $L_0 \approx -0.6, \phi = 0, \pi$ shrink and finally disappear — they support fewer and fewer librational states which during an increase of the field ellipticity vault over separatrix and become rotational ones. Also the islands situated, for $\alpha = 0$, around $L_0 = 0, \phi = \pi/2, 3\pi/2$ shrink with a change of the ellipticity. Additionally they move towards higher negative values of $L_0$. For $\alpha$ close to unity all elliptical islands are too small to support semiclassical states. Then all states are rotational.

As mentioned before strong localization in both angular and radial motion, supporting the existence of non-spreading wavepackets is expected around maxima of $\Gamma$. Thus one expect nonspreading wavepacket character for states localized around $L_0 \approx \pm 0.6, \phi = 0, \pi$ for $\alpha < 0.17$. For the greater field ellipticity only the islands around $L_0 \approx 0.6, \phi = 0, \pi$ could support nonspreading wavepackets. However, single wavepacket propagating along the 2:1 resonance periodic orbit could not fulfill the periodicity of Floquet states because the period of the orbit is twice longer than the microwave period. So one expects Floquet states being linear combinations of two wavepackets shifted in $t$, i.e. a coordinate variable in the extended phase space, by $2\pi/\omega$ which exchange their positions after the microwave period [11].

As a representative of such wavepackets propagating on an elliptical trajectory we have chosen to plot the states localized around $L_0 \approx -0.6, \phi = 0, \pi$ for $\alpha = 0.1$. Because of the tunneling effect a single quantum eigenstate contains a symmetric or antisymmetric combination of two semiclassical solutions corresponding to the ellipses with the Runge-Lenz vector directed parallel or antiparallel to the $x$-axis. Linear combinations of two such eigenstates allow to remove one of the ellipses. The resulting state is localized on a single ellipse but still it consists of symmetric combination of two wavepackets shifted in microwave phase by $2\pi$ (for the 2:1 resonance case [11]). To separate a single wavepacket one has to find another state localized on the same ellipse but containing antisymmetric combination of the wavepackets. The desired state is prepared using eigenstates coming from the similar manifold shifted by $\omega/2$ [12]. The resulting wavepacket moving on an elliptical trajectory is shown in Fig. 3. This is the single wavepacket rotating in the opposite direction to the direction of the field vector rotation. It propagates along the periodic orbit supported by the 2:1 resonance, thus, the period of the motion is twice longer than the microwave period.

In conclusion we would like to stress that the wavepacket presented in Fig. 3 is not an eigenstate of the system. Tunneling effects between the stable classical islands will be changing the shape of the packet but, at least in the example considered, the corresponding time scale is of order of a few hundreds of the microwave period. Another mechanism of its destruction is a slow ionization [12].

The analysis presented is restricted to the 2D model, its validity for the real three dimensional atom is an open question. Certainly, in the limiting LP case, due to the azimuthal symmetry, the wavepackets appear as doughnuts shaped localized functions moving up and down (assuming a vertical polarization of LP microwaves) [2]. For the CP case, on the other hand, the wavepacket motion was found to remain essentially 2D [2]. The interesting problem how the third dimension affects the dynamics for the general EP case is left for future work.

Finally we would like to note that the 2:1 resonance is an example of a general $s : 1$ resonance as the angular motion generated by $\Gamma$ is topologically the same for $s \geq 2$.

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FIG. 1. Two dimensional hydrogen atom illuminated by resonant, $\omega_0 = 2$, elliptically polarized microwaves. The effective scaled perturbation $\Gamma_0 = \Gamma \omega^{2/3}$ is plotted as a function of the scaled angular momentum, $L_0$, and the angle, $\phi$, between the Runge-Lenz vector and the main axis of the polarization ellipse – upper row. Bottom row shows equipotential curves of the angular part $\Gamma$ of the Hamiltonian $H_r$, Eq. (2), representing the slow evolution of the Kepler ellipse. The curves correspond to semiclassical states originating from the $n_0 = 42$ hydrogenic manifold. Columns correspond to the different ellipticity of the microwaves $\alpha = 0.1$ (left) and $\alpha = 0.5$ (right).

FIG. 2. Two dimensional hydrogen atom driven by resonant, $\omega_0 = 2$, elliptically polarized microwaves. Level dynamics, versus $\alpha$ (i.e. the degree of the field ellipticity), of the semiclassical quasi-energies [panel (a)] of the states originating from the $n_0 = 42$ hydrogenic manifold for $F_0 = 0.03$ compared with the exact quantum results [panel (b)].

FIG. 3. Wavepacket, being a linear combination of 4 eigenstates of the hydrogen atom plus elliptically polarized microwaves system with the field amplitude $F_0 = 0.03$, frequency $\omega_0 = 2$ for $n_0 = 42$ and the ellipticity $\alpha = 0.1$. Temporal evolution is plotted at times $\omega t = 0$ (top left), $\pi/2$ (top center), $\pi$ (top right), $3\pi/2$ (bottom left), $2\pi$ (bottom right). This wavepacket rotates on an elliptical orbit in the opposite direction to the rotation of the field vector and essentially repeats its periodic motion with period $4\pi/\omega$. It slowly disperses, either because the 4 building states are not exactly degenerate (tunneling effect) or because it ionizes. The size of each box is $\pm4000$ Bohr radii in both $x$ and $y$ directions.
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