Resonant dynamics of the H atom in an elliptically polarized microwave field

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The dynamics of Rydberg states of atomic hydrogen driven by elliptically polarized microwaves of frequency fulfilling the 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 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 nonscattering wave packets propagating on elliptical trajectories. [S1050-2947(99)06902-4]

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The pioneering experiment on Rydberg hydrogen (H) atom 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 the behavior of atoms in microwave fields is associated with multiphoton processes, which makes the quantum perturbation calculations hardly possible (many atomic states are strongly coupled) while, on the other hand, allowing one to apply a semiclassical or even purely classical description. Signatures of local structures in classical phase space have been observed in the experimental ionization thresholds for ionization [4]. The stable structures are suitable for a semiclassical quantization. The largest of them are created by the primary resonances between the driving field and the unperturbed Kepler motions of the Rydberg electron. In terms of the principal quantum number \( n \) of the unperturbed atomic state, the ratio of the microwave frequency, \( \omega \), to the Kepler frequency, \( \omega_K = 1/n_0^2 \), has to fulfill \( \omega / \omega_K = s \), where \( s \) is an integer number. Some of the states supported by the resonance islands reveal interesting properties from the quantum-classical correspondence principle point of view. Their time dynamics may be viewed as a motion of nondispersive wave packets as shown for both circular polarization (CP) [4] and linear polarization (LP) [5] of microwaves. These new states attracted a lot of attention. Their decay via ionization [6] or spontaneous emission [7] as well as ways to populate such states in experiments [8] have been studied. The ways of modifying wave-packet properties by additional static fields were also considered [9,10]. Semiclassically, all of them are supported by the principal resonance island, i.e., \( \omega / \omega_K = 1 \). The wave packets built inside a higher primary resonance island have been discussed by Holthaus [11] in model one-dimensional (1D) systems and in a realistic H atom in LP microwaves [12].

Theoretical detection of such interesting states obviously requires reliable semiclassical methods. In the LP case the 1D approximation to the dynamics enabled the quantization of resonant states exploring the Mathieu equation [13]. In the CP case, passing to the rotating frame removes the explicit time dependence of the system and the harmonic approximation around the stable fixed point gives the appropriate semiclassical predictions [4,8]. For the realistic LP problem as well as for arbitrary elliptical polarization (EP) in the two-dimensional (2D) model of an atom, the semiclassical method based on the Born-Oppenheimer approximation allows us 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 non-scattering wave packets. 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 done previously [14], we treat semiclassically and quantum mechanically a simplified 2D model of an atom. Studies of such simplified models have been most successful in the past both for the LP problem (where the 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 [4,8] (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, \( H = \hat{H}(t) = i\hbar / \partial t \) 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 is based on passing to the extended phase space by defining the momentum \( p_t \), conjugate to the \( t \) (time) variable that yields the new Hamiltonian, \( \mathcal{H} = H + p_t \) [17]. The quasienergies of the system will then be the quantized values of \( \mathcal{H} \). As the next step we express the Hamiltonian in action-angle variables of the unperturbed Coulomb problem [18]. 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, \( \theta = \omega_0 t \), 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

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

where

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

The explicit form of \( \Gamma(L, \phi; \alpha) \) and \( \beta(L, \phi; \alpha) \) is given by Eq. (2.17) of [18].

The last stage is to quantize the system using the approximate Hamiltonian, Eq. (2). Trivial quantization of \( \hat{p}_t \), exploring the time periodicity of the system [note that in the rotating frame, defined by Eq. (3), the time period is \( \tau = 2\pi/\omega [12]) \), 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 \( (\hat{J}, \hat{\theta}) \) space is much faster than the angular motion in \( (L, \phi) \) space [19,18]. 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, wave-packet-like states lying close to the center of the resonance island in \( (\hat{J}, \hat{\theta}) \) 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 second order around the center of the resonance island defining \( I = \hat{J} - (s^2\omega)^{-1/3} \). Equation (2) gives then a standard pendulum Hamiltonian with the island size given by \( \sqrt{F}\Gamma \). 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 the similar treatment for 1D systems [11,13].

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 \( \omega = 2\omega K = 2/(n_0 + 1/2) \), i.e., \( \omega_0 = 2 \). Note that in the 2D model the effective principal quantum number is half-integer. Figure 1 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. The size of the resonance island in the \( (\hat{J}, \hat{\theta}) \) space depends on the 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., \( L_0 = \pm 1 \). 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. Figure 2 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 \( F_0 = F(n_0 + 1/2)^4 = 0.03 \). The nice quantitative agree-
ment 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 the earlier approach [14] for small $\Gamma$ region.

This level dynamics is easy to understand by inspection of angular motion change with $\alpha$. Fig. 1. 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 the 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. 1.

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. 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 nonspreading wave packets, is expected around maxima of $\Gamma$. Thus one expects nonspreading wave-packet 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 $L_0=0.6$, $\phi=0, \pi$ could support nonspreading wave packets. However, a single wave packet propagating along the 2:1 resonance periodic orbit could not fulfill the periodicity of Floquet states because the period of the orbit is twice as long as the microwave period. So one expects Floquet states being linear combinations of two wave packets 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 [11,12].

As a representative of such wave packets 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 us to remove one of the ellipses. The resulting state is localized on a single ellipse but still it consists of a symmetric combination of two wave packets shifted in the microwave phase by $2\pi$ (for the 2:1 resonance case [11,12]). To separate a single wave packet one has to find another state localized on the same ellipse but containing an antisymmetric combination of the wave packets. The desired state is prepared using eigenstates coming from the similar manifold shifted by $\omega L/2$ [11,12]. The resulting wave packet moving on an elliptical trajectory is shown in Fig. 3. This is the single wave packet 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 as long as the microwave period.

In conclusion, we would like to stress that the wave packet 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 [6].

![Wave packet](image)

**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 quasienergies [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.** Wave packet, being a linear combination of four eigenstates of the hydrogen atom plus elliptically polarized microwave 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 wave packet 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 four building states are not exactly degenerate (tunneling effect) or because it ionizes. The size of each box is $\pm 4000$ Bohr radii in both $x$ and $y$ directions.
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 wave packets appear as doughnut-shaped localized functions moving up and down (assuming a vertical polarization of LP microwaves) [12]. For the CP case, on the other hand, the wave-packet motion was found to remain essentially 2D [6]. The interesting problem of 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 Ω is topologically the same for s ≳ 2.

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