Nondispersive Electronic Wave Packets in Multiphoton Processes

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We describe how the nonlinear coupling between the unperturbed Coulomb motion of a Rydberg electron and a linearly polarized microwave field may create an electronic wave packet of almost "eternal" lifetime, evolving according to the classical equations of motion *without dispersion*. This new quantum object owes its existence to the underlying mixed regular chaotic structure of classical phase space and is experimentally accessible.

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The excitation and/or ionization of atomic Rydberg states by linearly polarized microwave radiation is a highly nonlinear process involving multiphoton transitions of very high order within the Rydberg manifold and to the continuum. Since this strongly nonlinear coupling between the atom and the field induces chaotic motion of the electron in a classical picture, it is also one of the few domains of atomic physics where the quantum signatures of classical chaos can be probed theoretically, numerically, as well as by laboratory experiments, all in one system. Since chaos manifests itself in the temporal evolution of classical trajectories, the most appropriate way to search for these signatures is to directly analyze the long time evolution of a quantum wave packet initially placed on a classical trajectory [1–3].

Conventional experimental creation of wave packets relies on the coherent excitation of neighboring Rydberg states of the unperturbed atom by a short optical pulse [2]. However, the anharmonicity of the unperturbed Rydberg spectrum—induced by the nonlinearity of the Coulomb interaction—implies the dispersion of these wave packets on a time scale longer than $T \sim n^3$, with n the mean value of the principal quantum number of the unperturbed eigenstates spanning the wave packet, and T the associated Kepler period. Obviously, it is desirable to counterbalance the dispersion, in order to dispose of a quasiclassical quantum object for arbitrary experimental exposure times, and to launch the quantum wave packet along a well-defined classical trajectory. We shall outline how to achieve both of these aims in the present contribution.

As has been shown earlier [4,5], the Floquet eigenstates of one-dimensional, periodically driven systems with nonlinear classical dynamics may reflect faithfully the underlying invariant structures of classical phase space. In particular, for the hydrogen atom restricted to one spatial degree of freedom, the principal nonlinear resonance between the atomic and the driving frequency (i.e., atomic frequency nearly equal to and in phase with the driving frequency), which is the largest island of stability embedded in the "chaotic sea" of classical, mixed regular chaotic phase space [6], creates a *coherent eigenstate* of the atom in the field [5–7]. The purpose of the present paper is to demonstrate how such a robust structure of stable motion in mixed phase space creates a "quasiclassical" quantum eigenstate of the real quantum system, as, e.g., in 3D hydrogen exposed to a linearly polarized microwave field. This *eigenstate of the atom in the field*—which oscillates with the driving frequency—exhibits the characteristic properties of a wave packet evolving according to the classical equations of motion, but does *not* disperse. The nonlinear interaction between the atom and the field counterbalances the dispersion mechanism induced by the nonlinear Coulomb interaction and locks the oscillations of the Rydberg electron and those of the external field. Since eigenstates of a quantum system can in principle be excited by resonant excitation, the present study may open a new way to generate quantum wave packets with well-defined classical analogs.

The Hamiltonian describing the hydrogen atom in a monochromatic, linearly polarized microwave field of constant amplitude F and frequency ω reads, in the length gauge and in atomic units,

$$H = \frac{\mathbf{p}^2}{2} - \frac{1}{r} + Fz\cos(\omega t)\,. \tag{1}$$

As described in earlier work, a complex dilation of the Floquet-Hamiltonian associated with H allows us to obtain the energies, the widths, and the associated Floquet eigenstates of the atom in the field [8].

Since we are interested in comparing the quantum dynamics to the underlying classical motion, we shall characterize the interaction between the atom (prepared in an initial state of principal quantum number n_0) and the field by the values of the scaled variables ω_0 and F_0,

$$F_0 = n_0^4 F, \qquad \omega_0 = n_0^3 \omega\,, \tag{2}$$

that derive from the classical scaling properties of (1) and completely determine the structure of classical phase space [9]. Throughout the remainder of this paper we chose $\omega_0 = 1.304$, which allows comparisons with a typical situation in the microwave experiments performed by the Stony Brook group [10].

Let us first reconsider the coherent eigenstate of the hydrogen atom restricted to one degree of freedom, in a microwave field of amplitude $F = 2.36865 \times 10^{-9}$ a.u. = 12.18 V/cm and frequency $\omega = 5.47145 \times 10^{-6}$ a.u. = $2\pi \times 36$ GHz with (quasi)energy $\epsilon \simeq -1.299 \times 10^{-4}$ a.u. (corresponding to principal quantum number $n_0 \simeq 62$ and scaled variables $F_0 = 0.035$ and $\omega_0 = 1.304$). Figures 1(a)–1(c) show the temporal evolution of this time-periodic eigenstate of the atom in the field, which solves the Floquet eigenvalue equation

$$(H - i\partial_t)|\psi\rangle = \epsilon|\psi\rangle. \quad (3)$$

$|\psi\rangle$ is a wave packet trapped by the principal resonance in classical phase space [6,7] and evolves according to the regular solution of the classical dynamics, *without dispersion*. It mimicks a classical trajectory that evolves *in phase* with the field with the same period $T = 2\pi/\omega$. The observed localization of the wave packet can only be realized in a strongly nonperturbative situation. Indeed, the time evolution of the periodic wave function is not a simple oscillation at frequency ω but also involves higher harmonics. In a multiphoton language, this means that building such a wave packet requires contributions of multiphoton processes of various orders, with well-defined amplitudes and relative phases, a phenomenon not easy to explain without referring to the underlying classical dynamics.

Note that the apparent (but *transient*) "dispersion" of the wave packet on a time scale shorter than T [Fig. 1(c), and equally Figs. 1(d) and 2(c) further below] reflects nothing but the probing of the Coulomb singularity by the electron and is *completely* consistent with the classical dynamics [6]. It is the quantum manifestation of the strong acceleration of the electron in the immediate vicinity of the nucleus and therefore of *classical* origin. The usual dispersion known from wave packets created by the coherent superposition of *distinct* Rydberg states with different energies of unharmonic spacing *cannot* occur here since the observed wave packet is made up by *one single* (Floquet) eigenstate of the system, with one single (quasi)energy.

The width of this wave packet eigenstate (i.e., the field-induced continuum coupling) is insignificant with respect to the maximum precision of the numerical calculation, corresponding to an *eternal lifetime* of the electronic wave packet ($\Gamma^{-1} \geq 10^{13}$ a.u. $\equiv 242$ μs). Such a lifetime is comparable to the natural lifetime (due to spontaneous emission, a phenomenon neglected in the present approach) of Rydberg states with $n_0 \simeq 60$.

Another solution of (3) is shown in Figs. 1(d)–1(f), corresponding to a coherent state placed on the hyperbolic point associated with the principal resonance, but contaminated by the ingoing and outgoing probability flux along the stable and unstable manifolds crossing at this point in phase space [6,7]. This is why obvious interference structure persists at any phase of the field. The electronic density is strongly peaked at maximum distance from the nucleus, at a phase $\omega t = \pi$ of the microwave. Since the associated classical trajectory is highly unstable, the quantum wave packet formed by the Floquet state can trace the classical trajectory only for a short time, and yet it displays the important feature of evolving in *phase opposition* (out of phase) with respect to the driving field. It is exactly this dephasing by π that defines the hyperbolic point and hence the irregular classical solution. Finally, despite the strong instability of the classical trajectory, this "contaminated" or "irregular coherent state" was also

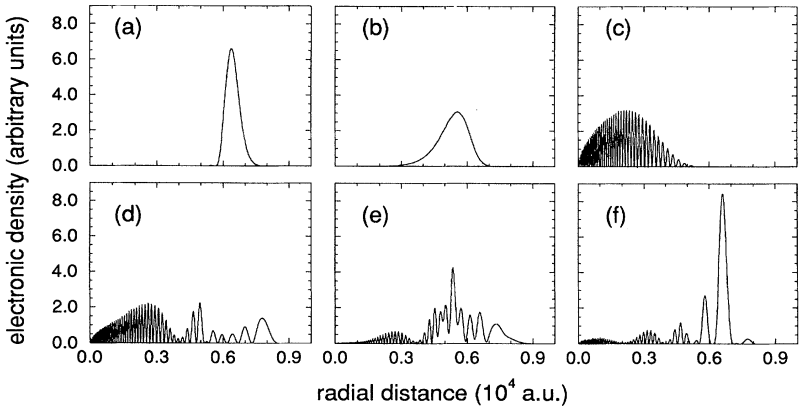

FIG. 1. (a)–(c) Coherent Floquet eigenstate of the hydrogen atom restricted to one spatial dimension in a linearly polarized microwave field of amplitude $F = 12.18$ V/cm and frequency $\omega/2\pi = 36$ GHz [corresponding to $F_0 = 0.035$ and $\omega_0 = 1.304$ for $n_0 = 62$, respectively, see Eq. (2)]. We show the periodically ($T = 2\pi/\omega$) evolving wave packet at phases $\omega t =$ (a) 0, (b) $\pi/2$, (c) π. The state evolves according to the regular solution of the classical dynamics, *in phase* with the microwave field, and is to be associated with the principal resonance between the atomic and the driving frequency (equality of microwave and Kepler frequency). It does not disperse and has eternal lifetime ($\tau > 242$ μs). Quasienergy $\epsilon \simeq -1.299 \times 10^{-4}$ a.u. (d)–(f) "Contaminated" coherent Floquet state, for the same field parameters and phases as (a)–(c). The state evolves according to the irregular solution of the classical dynamics, *in phase opposition* with the microwave field, and is to be associated with the principal hyperbolic point in classical phase space [6,7]. Even this quantum eigenstate associated with an unstable classical trajectory has an eternal lifetime ($\tau > 242$ μs). Quasienergy $\epsilon \simeq -1.293 \times 10^{-4}$ a.u.

found to have an "eternal" lifetime (as the "regular" coherent state discussed before), a feature that we attribute to its localization along a cantorus (acting as a barrier that inhibits quantal probability transport [11]) in classical phase space [5].

Do similar "classical" eigenstates exist for the real hydrogen atom in a microwave field? To answer this question, we investigated the Floquet spectrum of 3D atomic hydrogen in a field of amplitude $F \simeq 1.96 \times 10^{-7}$ a.u. = 1007.8 V/cm and frequency $\omega \simeq 1.07175 \times 10^{-4}$ a.u. = $2\pi \times 705$ GHz, in the vicinity of the $n_0 = 23$ energy shell, with $m_0 = 0$ the value of the (conserved) projection of the angular momentum onto the polarization axis of the field. This corresponds to $F_0 = 0.0549$ and $\omega_0 = 1.304$. Note that the only difference with the microwave experiments performed at Stony Brook [10] are the higher values of n_0 in the laboratory experiments. A numerical treatment of the 3D hydrogen atom in a microwave field at principal quantum numbers $n_0 \simeq 60$ and scaled frequencies $\omega_0 \simeq 1$ is not yet feasible due to computer memory limitations. However, we checked in our above 1D hydrogen calculations that the properties of the Floquet states we are discussing here do not change qualitatively from $n_0 \simeq 60$ to $n_0 = 23$ [5]. Classical structures that are resolved for $n_0 = 23$ will certainly prevail for $n_0 \simeq 60$, since the effective size of $\hbar$, i.e., of the quantum mechanical coarse graining of classical phase space *decreases* correspondingly from $\hbar/23$ to $\hbar/60$, as we approach the semiclassical limit by increasing n_0.

The result of our investigation is plotted in Fig. 2. Figure 2(a) shows the one-cycle averaged electronic density of a Floquet state of the 3D hydrogen atom in a microwave field, with properties analogous to those of the eigenstates shown in Fig. 1. This state extends only along the z axis, hereby retaining a completely one-dimensional character. Its effective one dimensionality makes this state a perfect candidate for the comparison to the 1D dynamics of the wave packets shown in Fig. 1. The periodic time evolution in both half spaces is completely independent, and this is actually due to the spatial degeneracy of the problem with respect to the $z = 0$ plane. The latter can be most easily seen by investigating the interaction part of the Hamiltonian (1), $Fz\cos(\omega t)$. Replacing z by $-z$ is equivalent to a phase shift of π, i.e., to the replacement $\omega t \rightarrow \omega t + \pi$. Thus, the temporal evolution in the $z > 0$ half space is symmetric to the evolution in the $z < 0$ half space *delayed* by a phase shift π. This is shown in Figs. 2(b)–2(d), and helps us to identify the classical motion mimicked by this eigenstate. Take the electronic density at phase $\omega t = 0$ of the field, Fig. 2(b). At this instant, there are two distinct, strongly localized maxima of the probability density, each in one of the two half spaces $z > 0$ and $z < 0$, the former one approaching the nucleus a little closer than the latter. Because of the symmetry argument given above, we know that the solution for $z > 0$ must equally appear for $z < 0$, with a phase delay of π, and vice versa. This is what we actually observe in Fig. 2(d), which is the symmetrical counterpart of (b). Hence, there are *two* wave packets moving forth and back from the nucleus in *each* half space. The two wave packets in the same half space are mutually phase shifted by π, evolving in phase with or in phase opposition to the driving field. Therefore they are the perfect analogs of the regular and the irregular states shown in Fig. 1, but now formed by *one single* Floquet state of the real atom. We surmise that the superposition of the two distinct 1D Floquet states in the 3D eigenstate shown here indicates a qualitative change in the topology of classical phase space due to the extension from three to five dimensions.

The fact that for any sign of z there is always one wave packet approaching the nucleus and another approaching the apocenter makes the actual dynamics of the electronic density relatively complicated and ensures that the distinct wave packets are most pronounced when they reach their outer turning points at phases $\omega t = 0$ and $\omega t = \pi$, respectively. However, a weak static electric field should be sufficient to lift the spatial degeneracy and isolate one out of two wave packets on either side.

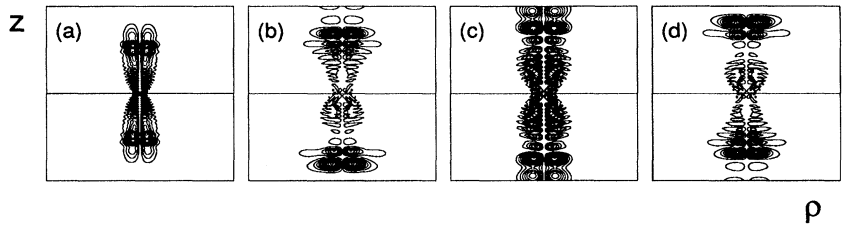

FIG. 2. (a) Contour representation of the one-cycle average of the electronic density $\rho|\psi(\mathbf{r},t)|^2$ of the Floquet eigenstate of the real hydrogen atom analogous to the regular and irregular eigenstates shown in Fig. 1, in cylindrical coordinates, $z, \rho = \pm 1400$ a.u. $F = 1007.8$ V/cm, $\omega = 2\pi \times 705$ GHz, corresponding to $F_0 = 0.0549$ and $\omega_0 = 1.304$ for $n_0 = 23$, respectively, see (2). Quasienergy $\epsilon \simeq -9.363 \times 10^{-4}$ a.u., lifetime $\tau \simeq 3.4$ μs (corresponding to about 10^6 unperturbed Kepler orbits of a Rydberg electron with $n_0 \simeq 20$). (b)–(d) Snapshots of the wave packet eigenstate of (a), at phases $\omega t =$ (b) 0, (c) $\pi/2$, (d) π of the driving field, on a smaller scale $z, \rho = \pm 950$ a.u. There are four distinct wave packets formed by the same Floquet state, two in each half space $z < 0$ and $z > 0$, respectively. These two pairs of wave packets are mutually identical, but delayed in phase by π [compare (b) to (d)]. In every half space, one wave packet shows exactly the same one-dimensional evolution of the regular eigenstate of Figs. 1(a)–1(c) (see the probability maximum at $\omega t = 0$ and $z < 0$), the other evolving as the irregular eigenstate of Figs. 1(d)–1(f) (see the probability maximum at $\omega t = \pi$ and $z < 0$).

The width of our 3D nondispersive wave packet(s) state is $\Gamma \simeq 7.2 \times 10^{-12}$ a.u. This gives a lifetime of $\tau \simeq 3.4\ \mu$s, which corresponds to $\tau/2\pi n_0^3 \sim 10^6$ Kepler periods of an unperturbed Rydberg electron with $n_0 \simeq 20$ and is again comparable to the natural lifetime due to spontaneous emission. Hence, as in the one-dimensional situation, this Floquet state has a very long lifetime, also compared to the typical interaction times in microwave ionization experiments (only the Garching experiments [12], on Rb Rydberg states, did scan the interaction time over several orders of magnitude), and is essentially *nondispersive* as compared to "conventional" Rydberg wave packets discussed in the introduction.

Let us finally address the experimental accessibility of our wave packet eigenstate. The form of the nodal lines as observed in Fig. 2 suggests a significant overlap with an extremal parabolic state $|n_1 = n_0 - 1, n_2 = 0, m_0 = 0\rangle$ (and, equivalently, $|n_1 = 0, n_2 = n_0 - 1, m_0 = 0\rangle$). We checked this assumption, knowing from a close inspection of the spectrum that the eigenstate of Fig. 2 originates from the $n_0 = 21$ manifold of the unperturbed atom [5]. Indeed, the eigenstate in the field has an overlap of approximately 15% with the initial state $|n_1 = 20, n_2 = 0, m_0 = 0\rangle$. For neighboring n_0 values ($n_0 = 19, \ldots, 24$), although some other parabolic states can contribute significantly, the overlap of the eigenstate in Fig. 2 with the extremal parabolic state is generally large. For example, it is still of approximately 8% with $|n_1 = 22, n_2 = 0, m_0 = 0\rangle$. Since $\omega_0 = \omega n_0^3 = 0.99$ for $n_0 = 21$, this signifies, in addition, that the principal resonance between driving field and unperturbed Kepler motion extends from $\omega_0 \simeq 0.74$ to $\omega_0 \simeq 1.49$. Note that this observation reinforces [5,13] the interpretation of Koch's local stability at $\omega_0 = 1.3$ as a consequence of the finite size of the classical principal resonance [14] rather than of some "scar" phenomenon [10,15]. It suggests the identification of our 3D eigenstate as the real separatrix state [14] recently invoked in the interpretation of these experiments. Because of the proximity of our 3D wave packet eigenstate to extremal parabolic states it should be the easiest accessible in Bayfield's [16] microwave experiments.

In conclusion, the wave packet eigenstate of the 3D hydrogen atom in the microwave field should be experimentally accessible via extremal parabolic states [16], possibly by adiabatic switching of the microwave field. Since irregular classical dynamics induce avoided crossings in the quantum spectrum, and as the state of Fig. 2 is an eigenstate of the atom dressed by the microwave field, a more appropriate way of accessing this state would consist of its resonant excitation from the atomic ground state or weakly excited states, *in the presence* of the microwave field. Because of the prominant role nonlinear resonances play in the transition from regular to chaotic motion, for any nonlinear system, we expect quasiclassical quantum eigenstates as shown in Fig. 2 to be a generic feature of (time dependent) quantum systems [4] with mixed regular chaotic classical counterparts. Note that, recently, the stability regions of classical mechanics associated with the Trojan asteroids in celestial mechanics [17,18] have been shown to support properly placed quantum wave packets in a numerical study of the bound state dynamics of a two-dimensional hydrogen atom exposed to a circularly polarized microwave field [17]. Our present work and earlier studies [4–7] on the case of linear microwave polarization let us conjecture that there exist associated "Trojan eigenstates" of the atom in the circularly polarized field [19].

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