Rapid Note

Controlling nonspreading wavepackets

K. Sacha^{1,a}, J. Zakrzewski^{1,b}, and D. Delande^{2,c}

¹ Instytut Fizyki imienia Mariana Smoluchowskiego, Uniwersytet Jagielloński, ulica Reymonta 4, 30-059 Kraków, Poland
 ² Laboratoire Kastler-Brossel, Tour 12, Étage 1, Université Pierre et Marie Curie, 4 Place Jussieu, 75005 Paris, France

Received: 27 November 1997 / Accepted: 27 January 1998

Abstract. We show how a static electric field can be used to control the localization of nonspreading wavepackets of a hydrogen atom driven by a linearly polarized microwave field. This allows for creation of wavepackets localized on fully stable resonance islands which, at the same time, can easily be excited by a direct optical transition from a low lying state. A semiclassical analysis is used to predict the energies and properties of such states.

PACS. 03.65.Sq Semiclassical theories and applications - 05.45.+b Theory and models of chaotic systems - 32.60.+i Zeeman and Stark effects

Strongly localized wavepackets that follow classical trajectories are an extremely useful tool in studies of the dynamics of a quantum system, especially in the semiclassical regime. Remarkable progress has been achieved over the last several years in the understanding of their properties as well as in their experimental creation and use [1]. It has been a common knowledge that the wavepackets in nonlinear systems must disperse, limiting their application to short time physics only. A breakthrough has been the recent discovery of nonspreading wavepackets in atoms driven by external electromagnetic (microwave) fields of either circular [2] or linear [3] polarization. The electronic motion is locked to the microwave frequency via the classical primary nonlinear resonance. The wavepacket then follows the periodic orbit lying in the center of the stable resonance island, and is trapped within the island (which prevents spreading). It is a linear combination of usual electronic states, and at the same time a single eigenstate of the combined system: atom + electromagnetic field [3], *i.e.* a dressed state or Floquet state. By construction, such a state evolves periodically in time and therefore does not spread.

Interesting properties of Floquet states localized in a classical resonance island have been studied much earlier [4]. That they can be viewed simultaneously as nonspreading wavepackets has been realized later [3]. An independent approach [2] used a specific property of the hydrogen atom in a circularly polarized microwave field: in the frame rotating with the microwave field, the system is

time-independant. Hence, the periodic orbit at the center of the resonance island, found first by Klar [5], appears in the rotating frame as a stable equilibrium point. A local harmonic expansion of the potential yields the gaussian form of the wavepacket. For other externally driven systems, the time-dependence cannot be removed by changing the coordinate frame. This makes the circular polarization case not generic.

It has been shown [6] that an additional magnetic field makes the local harmonic expansion much better and can be also used to stabilize another periodic orbit and thus create another wavepacket. Both these wavepackets move around the nucleus on circular orbits making their direct optical excitation hardly possible.

The full exact quantum studies of wavepacket properties in circularly polarized microwaves [7] revealed a number of their interesting properties, in particular the fluctuating character of their residual decay due to ionization (for details see Ref. [8]). Importantly, the limitations of the model without magnetic field suggested in [6] seem to be unjustified or at least exagerated. The single Floquet states found do not disperse by construction, follow closely the classical orbit and are linear combinations of pure electronic (mostly circular) states. Thus they are genuine quantum wavepackets in the standard quantum textbook meaning of this word.

An experimental method for the creation of such wavepackets starts from the excitation of the electron in a circular state followed by a careful switch on of the microwave [7,9]. The method requires the preparation of the atom in an initial pure circular state, a possible but not trivial task [10]. It is of great importance to find more

^a e-mail: sacha@chaos.if.uj.edu.pl

^b e-mail: kuba@crazy.if.uj.edu.pl

^c e-mail: delande@spectro.jussieu.fr

easily accessible wavepackets with the same nonspreading character. The obvious candidate is the hydrogen atom in a linearly polarized microwave, the case discussed in [3]. In the principal resonance island, the electron moves back and forth along the polarization axis of the microwave exactly at the microwave frequency (being phase locked) and comes every period close to the nucleus Thus it is vulnerable to a direct optical excitation from low lying atomic states. However, the stable resonant island supporting the wavepacket states in the simplified one-dimensional (1D) hydrogen atom model where the motion is restricted to the polarization axis, becomes a separatrix for the three-dimensional (3D) realistic model: the stable periodic orbit in 1D turns angularly unstable in 3D [3].

Stabilization of the angular motion is possible by adding an additional static electric field directed along the microwave polarization axis as shown in a different context by Leopold and Richards [11]. The same combination of microwave and static fields can be used to control the trajectories followed by strongly localized wavepackets, from a circular orbit in pure microwave field (such a wavepacket was already discussed in [12]) via elliptical orbits of different eccentricity to a straight line motion along the polarization axis.

To test this proposition, we consider a classical hydrogen atom in static electric and microwave fields along the same Oz direction with the Hamiltonian (in atomic units):

$$H = \frac{p_x^2 + p_y^2 + p_z^2}{2} - \frac{1}{r} + Fz \cos \omega t + Ez, \qquad (1)$$

where F is the amplitude of the microwave and E the static field. L_z is an exact constant of the motion, we take $L_z = 0$ in the following, the generalization of the results for arbitrary L_z being straightforward. We consider situations where both the static and the microwave fields can be considered as perturbations of the Kepler motion of the electron. We thus express the Hamiltonian in action-angle variables of the unperturbed hydrogen atom: these are the total action J (corresponding to the principal quantum number n) and the conjugate angle Θ describing the radial motion. To describe the angular motion, we use the total angular momentum L and its conjugate variable ψ , the angle between the Runge-Lenz vector (along the major axis of the unperturbed elliptical trajectory) and the z axis. In the absence of external fields, J, L and ψ are constants while Θ evolves at the classical Kepler frequency $\omega_K = 1/J^3$. We will consider the 1:1 resonance with the microwave frequency. The general case of m: kresonance will be discussed elsewhere [13]; recent experimental results [14] on this system have shown the importance of classical trajectories resonant with the microwave frequency: they contribute dominantly to the quantum dynamics, for example in the density of oscillator strengths. Similarly to the approach used in [11], we use the secular perturbation theory in the vicinity of the 1:1 resonance at first order in F. The fastest motion along the Θ coordinate is removed by passing to the "rotating" frame and averaging the Hamiltonian over the fast motion. Note that "rotating" frame means here a canonical transformation

removing the fast motion and not a frame rotating around the field axis. The next step is to expand the Hamiltonian around the center of the resonance island located at the principal action $J = n_0 = \omega^{-1/3}$ keeping terms quadratic in $\tilde{J} = J - n_0$ and linear in F and E. This yields the approximate resonance Hamiltonian:

$$H_{\rm res} = -\frac{1}{2n_0^2} - \omega n_0 - \frac{3\tilde{J}^2}{2n_0^4} + EV_0 \cos\psi + \frac{F}{2} [V_1 \cos(\Theta + \psi) + V_{-1} \cos(\Theta - \psi)], \quad (2)$$

 V_k are the Fourier coefficients of the perturbation; $V_0 = -3en_0^2/2$ and

$$V_k(n_0, L) = \frac{1}{k} \Big[\mathcal{J}'_k(ke) + \frac{L}{n_0 e} \mathcal{J}_k(ke) \Big] n_0^2, \text{ for } k = \pm 1,$$
(3)

with $e = \sqrt{1 - L^2/n_0^2}$ being the eccentricity of the classical elliptical trajectory and $\mathcal{J}_k(x)$ and $\mathcal{J}'_k(x)$ standing for the ordinary Bessel function and its derivative, respectively. A shorthand notation

$$\Gamma = \frac{1}{2}\sqrt{V_1^2 + 2V_1V_{-1}\cos 2\psi + V_{-1}^2}$$

$$V_{-1} - V_1$$

and $\tan \beta = \frac{v_{-1} - v_1}{V_1 + V_{-1}} \tan \psi$ allows us to express (2) as

$$H_{\rm res} = -\frac{1}{2n_0^2} - \omega n_0 - \frac{3J^2}{2n_0^4} + EV_0 \cos\psi + F\Gamma(e,\psi)\cos(\Theta - \beta).$$
(4)

This classical Hamiltonian has a scaling property: the n_0 dependences of the various terms in equation (4) can be factored out, resulting in a classical dynamics depending only on the scaled quantities $E_0 = En_0^4$, $F_0 = Fn_0^4$ and $L_0 = L/n_0$. The crucial point noticed in [11] is that, for moderate F values $(F_0 \ll 1)$, the radial motion (in \tilde{J}, Θ) and the angular motion (in L, ψ) have very different frequencies: $\simeq \sqrt{F_0}\omega$ for the fast radial motion and $\simeq F_0\omega$ for the slow angular motion. This has been used in [11] to estimate the ionization thresholds using the Chirikov overlap criterion (for similar treatments for other microwave polarizations see Refs. [16,17]). We can go beyond this analysis and use an approximate adiabatic analysis to semiclassically quantize the levels of interest. Such an approach has been shown in excellent agreement with exact quantum data for E = 0 [15] providing further justification for the results shown.

The fast radial (\tilde{J}, Θ) motion is described by a pendulum Hamiltonian, see equation (4), with the potential part being dependent on ψ and L (via the eccentricity e). The semiclassical quantization of the pendulum Hamiltonian is easily performed. As we are interested in the ground state of the radial motion, an harmonic approximation for the pendulum Hamiltonian is appropriate. This allows us to write the Hamiltonian for the slow angular motion as

$$H_{\text{slow}} = -\frac{1}{2n_0^2} - \Omega I + F\Gamma(e,\psi) + EV_0\cos\psi \qquad (5)$$



Fig. 1. Contours of the classical Hamiltonian, equation (5), describing the slow angular motion of the electron of an hydrogen atom exposed to collinear static and resonant microwave electric fields, in the (L_0, ψ) plane $(L_0$ is the scaled angular momentum and ψ the angle between the major axis of the elliptical trajectory and the field axis). The plot is for principal quantum number $n_0 = 60$, scaled microwave field $F_0 = 0.03$ and scaled static field $E_0 = 0.12F_0 < E_c$ (a), and $E_0 = 0.25F_0 > E_c$ (b). The lighter the background, the higher the energy. The contours are plotted at the semiclassically quantized values of the energy and thus represent the 60 states of the hydrogenic manifold. Observe the motion of the stable island along the $\psi/\pi = 1$ line corresponding to the highest lying state in the manifold.

where I is the action variable of the harmonic motion in the resonance island at frequency $\Omega = \sqrt{3F\Gamma}/n_0^2$ (the term $-\omega n_0$ has cancelled out by passing back to the laboratory frame). The standard WKB quantization of this motion yields half-integer quantized values of the action I, *i.e.* I = 1/2 for the minimal wave packet we are interested in.

The slow angular (L, ψ) motion, generated by H_{slow} , takes place along curves of constant $H_{\text{slow}}(L, \psi)$, see Figure 1. It is easy to check that:

- For E₀ = 0 (microwave only) two stable fixed points exist in the (L₀, ψ) plane, corresponding to stable motion along two resonant periodic orbits. The first one corresponds to a maximal angular momentum L₀ = 1, e = 0, *i.e.* a circular orbit. There, the angle ψ is a dummy variable since the direction of the major axis is not defined. The nonspreading wavepackets localized on such an orbit have been discussed in [12]. The other stable fixed point corresponds to L₀ = 0, ψ = π/2, representing a motion along a straight line in the plane perpendicular to the polarization axis. The corresponding wavepackets are only weakly radially localized since Γ(e, ψ) vanishes at the fixed point, leading to a small (J̃, Θ) resonance island in its vicinity.
- For $0 < E_0 < E_c$ (with E_c given below) a stable fixed point exists for $\psi = \pi$ and L_0 decreasing with increasing E_0 ; it corresponds to a stable resonant elliptical trajectory with major axis along the microwave polarization axis. Thus, by adding a static field, the wavepacket found in [12] can be, in a controlled way, smoothly "moved" from a circular to an elliptical trajectory. Simultaneously the fixed point at $L_0 = 0$ moves from $\psi = \pi/2$ to lower ψ values; it corresponds to a straight linear orbit tilted with respect to the polarization axis.
- For $E_0 > E_c$, the fixed points reach their extremal positions. Now, the stable points are at $L_0 = 0$, $\psi = 0$, π corresponding to straight linear orbits elongated along the polarization axis. The motion is fully stable in all directions and the approximate one-dimensional description of the system possible [11]. The angular motion is very similar to the pure static Stark case.

The critical field value is

$$E_{\rm c} = \frac{2}{3} \left| F_0 \mathcal{J}_1'(1) - \frac{\sqrt{3F_0 \mathcal{J}_1'(1)}}{4n_0} \right| \approx 0.217F_0 - 0.164 \frac{\sqrt{F_0}}{n_0} \cdot$$
(6)

In the semiclassical limit $n_0 \to \infty$, we recover the purely classical value for the stability of the 1D motion found in [11].

The semiclassical quantization of the slow motion is done using the standard semiclassical prescription [18], *i.e.* $\oint Ld\psi = 2\pi(p+1/2)$ with p an integer. It has to be carried out numerically because of the complicated functional dependence on L, ψ in equation (5). The contours corresponding to the quantized values for $n_0 = 60$ are shown in Figure 1. Note the strong localization in L, ψ for states lying in the vicinity of stable fixed points.

Figure 2 represents the semiclassical energy levels obtained for a fixed microwave amplitude $F_0 = 0.03$ while increasing E_0 from 0 to $2E_c$ for $n_0 = 60$. Note that, because of the explicit time-dependence of the Hamiltonian, these are rather quasi-energies of the Floquet states (dressed states). For large E_0 the levels are practically equidistant - the manifold is similar the one observed for the Stark effect. The separatrices dividing the phase space appear



Fig. 2. The semiclassical energy levels of the $n_0 = 60$ resonant hydrogenic manifold as a function of the ratio of the static electric field to the microwave amplitude for $F_0 = 0.03$. The insert shows the scaled angular momentum L_0 of the stable fixed point $(L_0, \psi = 0)$ as a function of the same variable. The corresponding trajectory evolve from a circular orbit containing the polarization axis to a straight linear orbit along this axis *via* intermediate elliptical orbits. Above $E_0 = E_c$ the stable fixed point is located at $L_0 = 0$. The corresponding wavepacket state, localized in the vicinity of the fixed point, is the highest lying state in the main figure. For $E_0 > E_c$, it is a completely localizad wavepacket in the 3 dimensions of space, which propagates back and forth along the polarization axis without spreading.

clearly in the plot as a series of avoided crossings. The uppermost state - an analogue of the blue shifted Stark state - is the wavepacket localized on a circular orbit for $E_0 = 0$, then on a elliptical trajectory with eccentricity increasing with E_0 , and finally, for $E_0 > E_c$ on a straight linear trajectory.

Such a blue shifted wavepacket should be easily accessible to experiments. A direct optical excitation from low lying states is possible. Another way of preparing such a wavepacket would consist of the excitation of a blue shifted Stark state followed by a smooth turn on of the microwave. Then, to reach the wavepackets localized on elliptical trajectories, one may decrease the static field below E_c sufficiently slowly to pass adiabatically the avoided crossings around E_c .

To conclude, we have shown by a semiclassical analysis of the slow secular motion for the 3D hydrogen atom that the presence of a static electric field collinear with a linearly polarized microwave field allows for the full control of trajectories on which the nonspreading wavepacket moves. For sufficiently large static fields (above E_c , about 22% of the microwave amplitude) the quasi-1D motion along the electric field direction is stable enabling the creation of nonspreading wavepackets, strongly localized in all directions. These wavepackets should be much easier to excite experimentally than the wavepackets moving on circular orbits [2,6,7,12]. Finally let us point out that absorption/emission properties of such a wavepacket should be quite fascinating from the quantum optics point of view. The enhancement of the absorption/emission probability occurs when the electron is close to the nucleus. Thus one may expect a time-dependent rate of such processes and non-lorentzian spectral profiles. Study of absorption/emission properties of such wavepackets is under progress.

Laboratoire Kastler Brossel de l'UPMC et de l'ENS is unité associée 18 du CNRS. Support of KBN under project 2P03B 03810 (KS and JZ), of the bilateral collaboration scheme (JZ and DD) no.76209 and the Programme International de Coopération Scientifique (CNRS) no.408 is acknowledged.

References

- 1. J.A. Yeazell, C.R. Stroud, Phys. Rev. A 43, 5153 (1991).
- I. Białynicki-Birula, M. Kaliński, J.H. Eberly, Phys. Rev. Lett. 73, 1777 (1994).
- A. Buchleitner, Ph. D. thesis, Université Pierre et Marie Curie, Paris VI, 1993 (unpublished); D. Delande, A. Buchleitner, Adv. At. Mol. Opt. Phys. 35, 85 (1994); A. Buchleitner, D. Delande, Chaos, Solitons and Fractals 5, 1125 (1995); Phys. Rev. Lett. 75, 1487 (1995).
- J. Henkel, M. Holthaus, Phys. Rev. A 45, 1978 (1992); M. Holthaus, Chaos, Solitons and Fractals 5, 1143 (1995) and references therein.
- 5. H. Klar, Z. Phys. D 11, 45 (1989).
- D. Farrelly, E. Lee, T. Uzer, Phys. Rev. Lett. **75**, 972 (1995); E. Lee, A.F. Brunello, D. Farrely, Phys. Rev. A **55**, 2203 (1997).
- D. Delande, J. Zakrzewski, A. Buchleitner, Europhys. Lett. 32, 107 (1995); J. Zakrzewski, D. Delande, A. Buchleitner, Phys. Rev. Lett. 75, 4015 (1995).
- J. Zakrzewski, D. Delande, A. Buchleitner, Phys. Rev. E 57, 1458 (1998).
- J. Zakrzewski, D. Delande, J. Phys. B: Atom. Mol. Opt. Phys. **30**, L87 (1997).
- R.G. Hulet, D. Kleppner, Phys. Rev. Lett. **51**, 1430 (1983);
 D. Delande, J.C. Gay, Europhy. Lett. **5**, 303 (1988);
 J. Hare, M. Gross, P. Goy, Phys. Rev. Lett. **61**, 1938 (1988);
 R.J. Brecha, G. Raithel, C. Wagner, H. Walther, Opt. Commun. **102**, 257 (1993).
- J.G. Leopold, D. Richards, J. Phys. B: Atom. Mol. Opt. Phys. 19, 1125 (1986); *ibid.* 20, 2369 (1987).
- 12. M. Kalinski, J.H. Eberly, Phys. Rev. A 52, 4285 (1995).
- 13. K. Sacha, J. Zakrzewski, D. Delande (unpublished).
- 14. N. Spellmeyer et al., Phys. Rev. Lett. 79, 1650 (1997).
- A. Buchleitner, D. Delande, Phys. Rev. A 55, R1575 (1997); A. Buchleitner, K. Sacha, D. Delande, J. Zakrzewski (unpublished).
- G. Casati, I. Guarneri, D.L. Shepelyansky, IEEE J. Quant. Electron. 24, 1420 (1988).
- K. Sacha, J. Zakrzewski, Phys. Rev. A 55, 568 (1997); *ibid.* 56, 719 (1997).
- 18. I.C. Percival, Adv. Chem. Phys. 36, 1 (1977).