

Time evolution and multiphoton ionization of Rydberg wavepacket in microwave fields

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Received 10 April 1995; in final form 3 May 1995

Abstract

We present a theoretical study of the time evolution of Rydberg wavepacket, initially localized in radial space and mimicking the classical particle, undergoing microwave multiphoton ionization. The dynamical behavior of the Rydberg wavepacket is found to be different from that of Rydberg atoms initially prepared at given Rydberg eigenstates. It is seen that the Rydberg wavepacket can exhibit collapse and revival behavior in the short time regime. Further, both the microwave ionization rates and the quantum phase space trajectories of the Rydberg wavepackets exhibit qualitatively different behavior in small scaled-frequency ($\omega_s (\equiv \omega n_0^3) \ll 1.0$) and large scaled-frequency ($\omega_s \geq 1.0$) regimes.

1. Introduction

Recently it has been demonstrated experimentally [1,2] that it is feasible to create a Rydberg wavepacket from the ground state by a short pulse laser excitation. In this case, a non-stationary state is obtained which is a coherent superposition of stationary states. The Rydberg wavepacket so generated can be localized in radial space and will mimic the classical radial motion of a charged particle in a Coulomb field. The time evolution of the wavepacket exhibits effects like dispersion and revivals [3,4]. As such the study of the dynamics of Rydberg wavepacket provides a unique opportunity, both theoretically and experimentally, for probing the correspondence be-

tween classical and quantum mechanics. Most experimental studies so far have been focused on the time evolution of the field-free Rydberg wavepacket [1–4].

In this Letter, we present the first theoretical study of the time evolution of Rydberg wavepacket driven by microwave fields. Historically there has been considerable interest in the past decade in the study of microwave ionization of Rydberg H atoms initially prepared in highly excited eigenstates [5,6]. This is a non-integrable system which exhibits classically chaotic motion and has served as one of the paradigms for ‘quantum’ chaos study [7]. In this Letter, we report some interesting new features of microwave ionization from the initial Rydberg wavepacket whose behavior is shown to be qualitatively different from that of ionization from the highly excited eigenstates.

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2. Time evolution of Rydberg wavepacket in microwave fields

A prototype system that has been widely used in the study of microwave ionization of Rydberg atoms (initially prepared in given highly excited eigenstates) is the 1D hydrogen atom driven by the external fields [7]. The dynamical evolution of such Rydberg atoms satisfies the time-dependent Schrödinger equation (in atomic units),

$$i\partial/\partial t \psi(x, t) = \hat{H}(t) \psi(x, t), \quad (1)$$

where

$$\hat{H} = \hat{P}^2/2m + \hat{V}(x, t) \quad (2a)$$

and

$$\hat{V} = -1/x - Fx \sin(\omega t). \quad (2b)$$

Here we are interested in the time evolution of the Rydberg atoms initially prepared in the (Gaussian) wavepacket form,

$$\psi(x, 0) = (\pi\sigma^2)^{-1/4} \exp[-(x-x_0)^2/2\sigma^2], \quad (3)$$

where $x_0 = 2\bar{n}_0^2$, $\sigma = (20/\pi)\bar{n}_0$, and \bar{n}_0 is the mean quantum number of the wavepacket.

We shall extend the fast Fourier transformation (FFT) method [8–10] to numerically integrate the time-dependent Schrödinger equation in space (x) and time (t). We have previously shown that this method is capable of providing reliable and efficient treatment of the time evolution of Rydberg eigenstates in microwave fields [10]. It also allows an accurate treatment of the continuum states. To propagate the wavefunction, we use the split-operator algorithm [8],

$$\begin{aligned} \psi(t + \Delta t) \approx & \exp[-i(\hat{P}^2/4m)\Delta t] \exp(-i\hat{V}\Delta t) \\ & \times \exp[-i(\hat{P}^2/4m)\Delta t] \psi(t) \\ & + O[(\Delta t)^3], \end{aligned} \quad (4)$$

where the effect of applying the operator $\exp[-i(\hat{P}^2/4m)\Delta t]$ is evaluated in terms of discrete Fourier transforms and the FFT algorithm.

In this calculation, the spatial x coordinate is discretized from 0 to 25000 a_0 by using 2048 mesh points. To prevent unphysical reflection from the

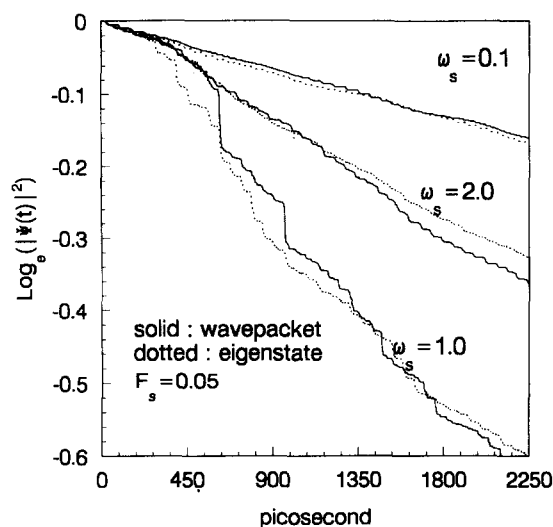


Fig. 1. Logarithm of $|\psi(t)|^2$ versus t (in ps), showing the decay of the wavefunctions with time due to microwave ionization. The scaled field strength is $F_s = 0.05$ au for all the three scaled frequency cases considered: (a) $\omega_s = 0.1$ au, (b) $\omega_s = 1.0$ au, and (c) $\omega_s = 2.0$ au. Solid curves for Rydberg atoms initially prepared in the Gaussian wavepacket form with $\bar{n}_0 = 72$. Dotted curves for Rydberg atoms initially prepared at $n_0 = 72$ eigenstate.

outer boundary, we adopt the absorbing boundary method [10] and place a slowly vanishing filter function ($b \approx 0.004$),

$$f(x) = \{1 + \exp[b(x - x_f)]\}^{-1}, \quad (5)$$

centered at $x_f = 22500 a_0$. The result is insensitive to the choice of b and x_f , provided that x_f is chosen large enough and b small enough.

Fig. 1 shows the comparison of the decay rate of the Rydberg H atoms initially prepared in the eigenstate $n_0 = 72$ (dotted curves) or in the localized wavepacket form, Eq. (3), with $\bar{n}_0 = 72$ (solid curves). Three different microwave frequencies have been considered, all at the same scaled field strength $F_s (= Fn_0^4) = 0.05$ au: (a) ω_s (scaled frequency $\equiv \omega n_0^3$) = 0.1 au, (b) $\omega_s = 1.0$ au, and (c) $\omega_s = 2.0$ au. The time scale plotted here is in terms of the real time (in ps). If expressed in terms of the number of optical cycles (oc), the time span in Fig. 1 is equivalent to $t = 0$ to 4 oc, for the $\omega_s = 0.1$ case, $t = 0$ to 40 oc, for the $\omega_s = 1.0$ case, and $t = 0$ to 80 oc for the $\omega_s = 2.0$ case. We see that the $\omega_s = 1.0$ case has the largest ionization (decay) rate overall and the $\omega_s = 0.1$ case has the smallest. This may be at-

tributed to the fact that for $\omega_s = 1.0$, the microwave frequency $\omega (\equiv \omega_s n_0^{-3})$ is close to the spacing ΔE of nearby levels and resonance excitation can occur,

resulting in the enhancement of ionization rate. For the small scaled frequency ($\omega_s = 0.1$) case, the microwave frequency ω is much smaller than the adja-

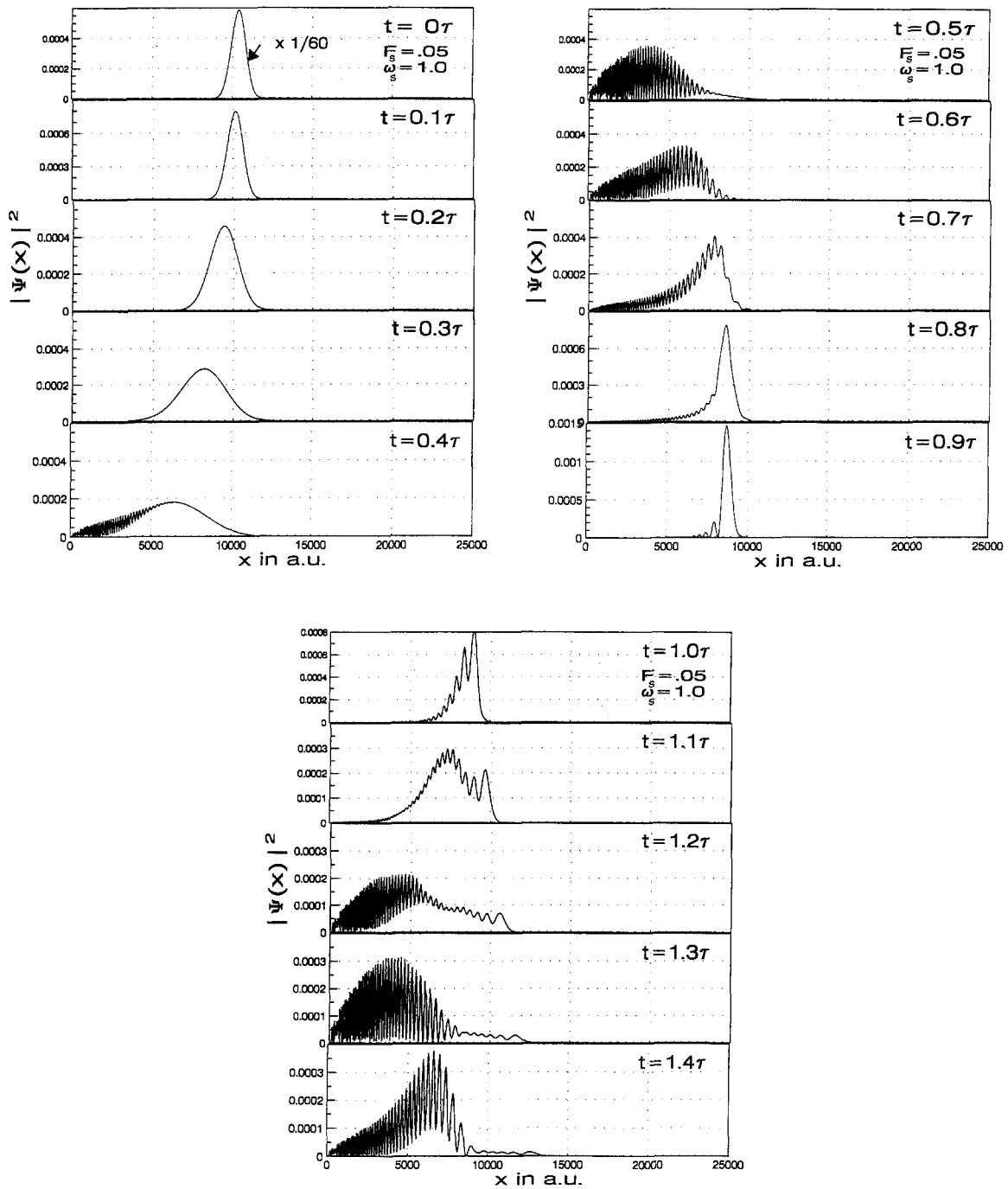


Fig. 2. The time evolution of Rydberg wavepacket ($\bar{n}_0 = 72$) in the microwave field in the short time period ($t = 0$ to 1.4 optical cycles). The field parameters are $F_s = 0.05$ au, and $\omega_s = 1.0$ au.

cent level spacing ΔE . Thus, microwave ionization occurs through non-resonant processes, yielding smaller ionization rate. Fig. 1 also compares the wavefunction decaying behavior for the Rydberg eigenstate versus the Rydberg wavepacket. For the $\omega_s = 1.0$ case, it is seen that there is some initial time delay for ionization. After that, the eigenstate tends to have a larger decay rate at least in the short and intermediate time period. This is understandable in terms of the resonant excitation nature from the initial eigenstate. However, the decay of the wavefunction in time is not quite monotonic and in fact the wavepacket can have larger decay rate at later times. Similar behavior occurs for the $\omega_s = 2.0$ case. For the non-resonant ($\omega_s = 0.1$) case, the decay rate is comparable for the wavepacket and eigenstate excitation, both showing some exponential decay behavior at shorter times. At longer times (not shown in Fig. 1), we found the wavepacket tends to be ionized faster than the eigenstate, similar to the larger frequency cases.

It is instructive to examine the time evolution of the initial wavepacket in the microwave field. It has been observed experimentally that, in the field-free case, the initially localized Rydberg wavepacket can exhibit collapse, fractional revival and full revival [3,4]. In Fig. 2, we show the time evolution of the Rydberg wavepacket for the $\omega_s = 1.0$ and $\bar{n}_0 = 72$ case for a series of time interval $\Delta t = 0.1 \tau$, where τ is the period of field oscillation. The Rydberg (Gaussian) wavepacket, initially centered at $x_0 = 10368 a_0$ ($= 2\bar{n}_0^2$), is undergoing first the broadening and the drifting to the shorter distance from $t = 0$ to 0.3τ . At about $t \approx 0.4\tau$, the left branch of the wavepacket reaches the Coulomb center (nucleus), inducing photoabsorption and causing a rapid oscillatory behavior starting from the short distance. As the center of wavepacket continues to drift to the shorter distance, the oscillatory behavior spreads more and more across the wavepacket. At $t \approx 0.5\tau$, the center of the wavepacket reaches the closest distance of approach to the Coulomb center, and the whole wavepacket exhibits rapid oscillatory behavior, while still preserving the overall Gaussian envelope. After that, the wavepacket begins to bounce back and its center now drift to the larger distance. As time goes by, the oscillatory behavior gradually disappears starting from the right side (larger distance). Finally at $t \approx$

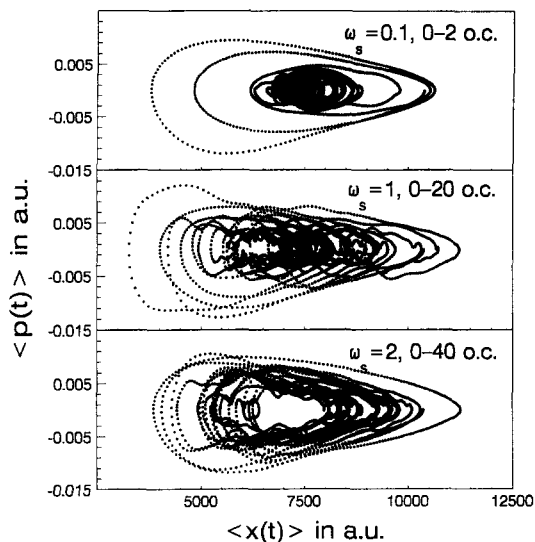


Fig. 3. Quantum phase space trajectories for the time evolution of the initial Rydberg wavepacket ($\bar{n}_0 = 72$) in the microwave field for $\omega_s = 0.1, 1.0$ and 2.0 au, respectively. The scaled field strength is $F_s = 0.05$ au.

0.9τ , the wavepacket reaches the 'revival', nearly resuming its initial Gaussian shape. After that time, the wavepacket begins to drift to the left again and repeats the oscillatory behavior and so on as described before. Such revival phenomenon, however, will not last long. After a few such revivals, the wavepacket breaks up into several pieces and the time propagation begins to lose coherence. Similar behavior has also been observed for the $\omega_s = 0.1$ and $\omega_s = 2.0$ cases, although the revival time is different.

Fig. 3 depicts the quantum phase space trajectories corresponding to $\omega_s = 0.1$ ($t = 0-2$ oc), $\omega_s = 1.0$ ($t = 0-20$ oc), and $\omega_s = 2.0$ ($t = 0-40$ oc) cases. The initial state is prepared at $\bar{n} = 72$ Rydberg wavepacket and the scaled field strength is $F_s = 0.05$ au for all the three cases. Here $\langle x(t) \rangle$ and $\langle p(t) \rangle$ are, respectively, the expectation values of the electronic coordinate (x) and momentum (p). The three phase space diagrams span the same (real) time period and each contains 4000 phase space points. Several salient features are noticed: (i) For the non-resonant small frequency ($\omega_s = 0.1$) case, the phase space structure reveals a denser and more chaotic pattern, whereas the larger frequency ($\omega_s = 1.0$ and 2.0) cases show more regular patterns. (ii) As time goes longer, all the phase space volumes shrink. This

is attributed to the fact that we have placed a filter at a large distance and it has filtered out the ionized portion of the wavefunctions.

It will be instructive to compare the time evolution of the quantum Rydberg wavepacket and the classical dynamics in microwave fields. It is known from the previous studies of microwave ionization of eigenstates [5–7] that classical calculations can provide rather good agreement with both the experimental and the quantum results in the small scaled frequency regime ($\omega_s \ll 1.0$). However, quantum suppression of classical chaos can occur at larger scaled frequencies ($\omega_s \geq 1.0$) and significant discrepancies exist between classical and quantum results. In the present Rydberg wavepacket excitation, the wavepacket evolves more closely to the motion of the classical particle. Thus a detailed exploration of the classical-quantum correspondence will be of special interest. Work in this direction is in progress.

Acknowledgements

This work was partially supported by the Division of Chemical Sciences, Office of Basic Energy Sciences of the US Department of Energy. The authors

also acknowledge the partial support of the US–Taiwan International Cooperation Program through the US-NSF and the NSC (83-0501-I-009-063-B12) in Taiwan.

References

- [1] A. ten Wolde, L.D. Noordam, A. Lagendijk and H.B. van Linden van den Heuvell, *Phys. Rev. Letters* 61 (1988) 2099.
- [2] J.A. Yeazell, M. Mallalieu, J. Parker and C.R. Stroud, *Phys. Rev. A* 40 (1989) 5040.
- [3] J.A. Yeazell, M. Mallalieu and C.R. Stroud, *Phys. Rev. Letters* 64 (1990) 2007.
- [4] D.R. Meacher, P.E. Meyler, I.G. Hughes and P. Ewart, *J. Phys. B* 24 (1991) L63.
- [5] J.E. Bayfield and D.W. Sokol, *Phys. Rev. Letters* 61 (1988) 2007.
- [6] K.A.H. van Leeuwen, G.V. Oppen, S. Renwick, J.B. Bowlin, P. M. Koch, R.V. Jensen, O. Raht, D. Richards and J.G. Leopold, *Phys. Rev. Letters* 55 (1985) 2231.
- [7] G. Casati, B.V. Chirikov, D.L. Shepelyansky and I. Guarneri, *Phys. Rept.* 154 (1987) 77; R. Jensen, *Phys. Rept.* 201 (1991) 1.
- [8] M.D. Feit, J.A. Fleck Jr. and A. Steiger, *J. Comput. Phys.* 47 (1982) 412.
- [9] R. Kosloff and D. Kosloff, *J. Chem. Phys.* 79 (1983) 1823.
- [10] S.I. Chu and T.F. Jiang, *Comput. Phys. Comm.* 63 (1991) 482.