

## IONIZATION OF RYDBERG ATOMS BY SUBPICOSECOND ELECTROMAGNETIC PULSES

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Theoretical analysis of the recently observed ionization of Rydberg atoms by ultra-short half-cycle single-polarity electromagnetic pulses is presented. It is shown that the observations are consistent with the general classical scaling for the hydrogen atom in a microwave field, but the quantitative explanation of the results is possible only within the framework of quantum mechanics. In other words, the photonic basis approach for quantum dynamics and the multiphoton ionization theory yield the measured threshold fields. This is an example of classical scaling of a non-classical effect.

### 1. Introduction

Studies of the ionization of highly excited atoms by static and dynamic fields have received a great deal of attention in recent years. Investigations are usually carried out for excited atoms with the principal quantum number  $n_0 \leq 100$  in microwaves or dc electric fields (for a review see [1-4] and references therein). These atoms serve as important simple examples of non-linear systems strongly driven by an external field and showing stochastic behaviour. Also they provide a unique opportunity to explore quantum manifestations of the classical chaos and the quantum-classical correspondence for chaotic systems [3,4]. However, until now temporal resolution and duration of electromagnetic pulses generated were rather large compared to the internal time of the atoms under study. Recently [5], in photoconducting switches, ultrashort half-cycle single-polarity electromagnetic pulses (HCP) with the width of  $\tau_{HCP} \approx 500$  fs and central frequency around 1 THz have been created, and they have been used for ionization of Na Rydberg atoms with the principal quantum number  $n_0 > 13$ . The authors [5] emphasize that the threshold electric field of the short pulses necessary for ionizing a

Rydberg state with the effective principal quantum number  $n_0$  is found to scale as  $n_0^{-2}$  in contradiction to the  $n_0^{-4}$  threshold scaling for static field ionization and high order multiphoton ionization. This is also in contrast with the  $n_0^{-5}$  threshold scaling for ionization by relatively long ( $\mu$ s) microwave pulses.

The purpose of this report is to show that the observations [5] are consistent with the general scaling relations for the hydrogen atom in a microwave field [6] and with the multiphoton ionization theory [1]. The threshold field strength may simply be evaluated via the photonic basis approach for chaotic dynamics. Moreover, it is likely that the quantitative description of the observations [5] is possible only on the basis of quantum mechanics although the threshold field scales classically. This may be in analogy with the classical scaling of non-classical stability in microwave ionization of Rydberg atoms found in [7].

### 2. Scaling relations

The Hamiltonian (in atomic units, a.u.,  $e = \hbar = m = 1$ ) for the hydrogen atom in a microwave field of the frequency  $\omega$  and field

strength  $F$  is

$$H = \mathbf{p}^2/2 - 1/r + zF\sin(\omega t + \varphi), \quad (1)$$

where  $\mathbf{r}$  and  $\mathbf{p}$  are the position and momentum of the electron, respectively, and  $\varphi$  is the initial phase. Measuring the time of the microwave field action on the hydrogen atom in field periods, one can introduce the scale transformation [6]

$$\begin{aligned} t &= \omega \tilde{t}, & \mathbf{r}_s &= \omega^{2/3} \mathbf{r}, & \mathbf{p}_s &= \mathbf{p}/\omega^{1/3}, \\ F_s &= F/\omega^{4/3}, & H_s &= H/\omega^{2/3}, \end{aligned} \quad (2)$$

where the scaled Hamiltonian is

$$H_s = \mathbf{p}_s^2/2 - 1/r_s + z_s F_s \sin(t + \varphi). \quad (3)$$

The scaled time-dependent Schrodinger equation can be expressed as

$$i\omega^{1/3} \frac{\partial \Psi}{\partial t} = (H_s^0 + V_s) \Psi,$$

where

$$\begin{aligned} H_s^0 &= \frac{\omega^{2/3}}{2} \left[ -\frac{1}{r_s^2} \frac{\partial}{\partial r_s} \left( r_s^2 \frac{\partial}{\partial r_s} \right) + \frac{l(l+1)}{r_s^2} \right] - \frac{1}{r_s}, \\ V_s &= z_s F_s \sin(t + \varphi). \end{aligned} \quad (5)$$

The scaled energy spectrum of the unperturbed hydrogen atom is

$$E_s = -1/2\omega^{2/3}n^2 = -1/2s^{2/3}, \quad (6)$$

where  $s = \omega/(-2E)^{3/2}$  is the relative field frequency, i.e. the ratio between the microwave frequency  $\omega$  and the Kepler orbital frequency of the electron  $\Omega = (-2E)^{3/2}$ .

Equation (3) shows that classical motion of the electron under the definite initial conditions depends only upon the scaled field strength. On the other hand, the scaled threshold field strength  $F_s^{\text{th}}$  for the onset of classical chaos or for the ionization of the Rydberg atom is a function of the initial scaled energy or initial relative field frequency  $s_0 = \omega n_0^{3/2}$ , i.e.  $F_s^{\text{th}} = f(s_0)$ . A particular form of the function  $f(s_0)$  depends upon the ionization mechanism, which differs for low, intermediate, or high relative frequency  $s_0$ . Namely, the static field ( $s_0 \rightarrow 0$ ) ionization threshold is  $F_s^{\text{st}} \approx 0.130/s_0^{4/3}$ , while the threshold at the onset of classical chaos in the high-frequency ( $s_0 \gg 1$ )

microwave field is  $F_s^{\text{MW}} \approx 1/49s_0^{5/3}$  [1-3, 6].

According to Eqs. (4) and (5), the motion of the quantum hydrogen atom in a monochromatic field is governed by the scaled field and also by the scaled Planck constant  $\hbar = \omega^{1/3}$  (in a.u.).

Note the possibility of another scaling when time is measured in Kepler periods of the initial Rydberg state with the effective principal quantum number  $n_0$  [7]:

$$\begin{aligned} t_0 &= \tilde{t}/n_0^3, \\ \mathbf{r}_0 &= \mathbf{r}/n_0^2, & \mathbf{p}_0 &= \mathbf{p}n_0, \\ F_0 &= Fn_0^4, & H_0 &= Hn_0^2. \end{aligned} \quad (7)$$

Here the scaled Hamiltonian  $H_0$  is

$$H_0 = \mathbf{p}_0^2/2 - 1/r_0 + z_0 F_0 \sin(s_0 t_0 + \varphi), \quad (8)$$

while the effective Planck constant is  $\hbar = 1/n_0$  a.u., and the scaled energy spectrum of the unperturbed atom is  $E_0 = En_0^2 = -(1/2)(n_0/\hbar)^2$ . This means that the initial scaled energy is  $E_0^i = -1/2$ , the classical dynamics determined by the Hamiltonian in Eq. (8) depends on  $F_0$  and  $s_0$ , and that the threshold field  $F_0^{\text{th}}$  is a function of the relative frequency  $s_0$ :  $F_0^{\text{th}} \equiv F_s^{\text{th}} s_0^{4/3} = f_0(s_0)$ . But in our problem, the scaling according to Eqs. (2)-(5) is more convenient.

Moreover, we can introduce into the scaled Hamiltonian (3) the relative field strength  $F_0 = Fn_0^4 = F_s s_0^{4/3}$ :

$$\begin{aligned} H &= \omega^{2/3} H_s, \\ H_s &= \mathbf{p}_s^2/2 - 1/r_s + s_0^{-4/3} z_s F_0 \sin(t + \varphi). \end{aligned} \quad (9)$$

The threshold values of  $F_0$  for ionization onsets at different mechanisms depend weaker upon the initial effective principal quantum number  $n_0$  and relative frequency  $s_0$ . Thus, for the above relative field strength, the static field ionization threshold is  $F_0^{\text{st}} \approx 0.130$ , the onset of classical chaos in the microwave field is at  $F_0^{\text{MW}} \approx 1/49s_0^{1/3}$ , while the threshold field for HCP ionization, according to finding [5], is  $F_0^{\text{HCP}} \approx 0.3/n_0^2 \tau_{\text{HCP}}^{2/3}$  if  $s_0 \geq 1$ , i.e.  $F_0^{\text{HCP}} \approx 0.14s_0^{2/3}$ .

Now let us refer to the theory of HCP ionization.

### 3. Ionization theory

#### 3.1 Classical ionization

In classical terms, ionization occurs when HCP gives to the electron an energy increase which is greater than its binding energy. From the equation of motion  $\dot{\mathbf{E}} = \mathbf{F}\dot{\mathbf{z}}$  it follows that the equation for the threshold field is

$$\frac{1}{2s_0^{2/3}} = F_s^{\text{th}} \max_{t_0} \int_0^\pi \dot{z}_s(t + t_0) \sin t dt, \quad (10)$$

where  $t_0$  is the initial phase of electron motion in the Rydberg atom. The energy gained by the electron is a maximum at small distances from the nucleus, where its velocity is maximal and independent of the electron energy:  $\dot{z}_s = (4/3)^{1/3}$ .

$(t + t_0)^{-1/3}$ . Therefore, Eq. (10) can be expressed as

$$\frac{1}{2s_0^{2/3}} = \frac{2^{2/3}}{3^{1/3}} F_s^{\text{th}} \max_{t_0} \int_0^\pi \frac{\sin t}{(t + t_0)^{1/3}} dt. \quad (11)$$

The integral in Eq. (11) may be simply evaluated analytically by modelling HCP as an inverted parabola [5]. The numerical analysis yields a maximum of the integral in Eq. (11) to be at  $t_0 \approx -0.15$  and equals 1.9. Thus,  $F_0^{\text{th}} = F_s^{\text{th}} s_0^{4/3} \approx \approx 0.24s_0^{2/3}$ , which is by a factor of 1.7 higher than the experimental results. From numerical integration of classical equations of motion for an electron in the Rydberg orbit, which is subjected to a HCP, the observed scaling also follows, but theoretical and experimental thresholds differ by a factor of 2.5 [5], so that agreement of the classical theory with the experimental results is only qualitative.

#### 3.2 Multistep ionization in photonic basis

Direct first-order quantum ionization of the Rydberg atom by electromagnetic pulses is relatively weak and exhibits no threshold field dependence, since it is proportional to  $F^2$ . Therefore, we consider multistep and multiphoton ionization processes. Calculation of transition probabilities between a great number of unperturbed atomic states and, as a result, the ionization process itself is a complex problem. For simplification, one can introduce the photonic basis and calculate the one-photon transition in the

strongly perturbed spectrum of the Rydberg atom [2,3,6,8,9]. Scaled equations for the transition amplitudes  $b_N$  between photonic states can be written as [6]

$$\frac{db_{N'}}{dt} = -iF_q \sin t \sum_N \langle N' | z_s | N \rangle e^{i\Delta N t} b_N, \quad (12)$$

where  $F_q = F_s/\omega^{1/3} = F/\omega^{5/3}$  is the quantum-scaled field strength. Evaluating the effective dipole matrix element between photonic states, we take into account the fact that such matrix elements for short electromagnetic pulses consist of matrix elements for transitions to atomic states lying in the energy gap equal to the photon energy. Using the known expressions for the dipole matrix elements between excited atomic states [10]

$$\langle n' | z | n \rangle = -b/(nn')^{3/2} |E' - E|^{5/3}, \quad b \approx 0.411,$$

we thus obtain the effective scaled dipole matrix elements for transitions between the photonic states

$$\begin{aligned} \langle N \pm \Delta N | z_s | N \rangle \approx & - (3b/2^{1/3}) (|2\Delta N - 1|^{-2/3} - \\ & - |2\Delta N + 1|^{-2/3}). \end{aligned} \quad (13)$$

Since the most important are matrix elements between neighbouring photonic levels, transition probabilities  $P_{N,N'}$  between photonic states may be evaluated by means of the Presnyakov and Urnov model [11]

$$P_{N,N'} = J_{N'-N}^2(K),$$

$$K = \pi F_q |\langle N \pm 1 | z_s | N \rangle| \approx \pi F_q / 2. \quad (14)$$

Here  $J_N(K)$  is the Bessel function. For the ionization, the number  $N' - N = N_i \approx 1/2n_0^2\omega$  of photons is required, and, consequently, the onset of ionization may be evaluated from

$$P_i \approx J_{N_i}^2(K) \approx (eK/2N_i)^{2N_i/2\pi N_i}, \quad (15)$$

where  $e = 2.718\dots$ . Thus, ionization is appreciable if  $eK/2N_i \approx 1$ , i.e.  $F_s^{\text{phb}} \approx 2/e\pi s_0^{2/3}$  or  $F_0^{\text{phb}} \approx 0.23s_0^{2/3}$ . Thus, this simple evaluation results in the threshold field, higher than the experimental observations only by a factor of 1.5.

#### 3.3 Multiphoton ionization

According to analytical evaluations [1], the rate of multiphoton ionization of Rydberg atoms is:

$$w_i \propto (7.05n_0^2 F/\omega^{2/3})^{2N_i}, \quad N_i \gg 1. \quad (16)$$

Therefore, the ionization is appreciable if  $7.05n_0^2 F/\omega^{2/3} \approx 1$ , i.e.  $F_s^{MPh} \approx 1/7.05s_0^{2/3}$  or  $F_0^{MPh} \approx 0.14s_0^{2/3}$ , which is in agreement not only with the observed scaling but with the absolute experimental values [5] as well.

#### 4. Conclusions

Observations on HCP ionization of Rydberg atoms [5] are consistent with the general classical scaling theory for the hydrogen atom in a microwave field [6]. The same scaling and close threshold fields result from the multiphoton ionization theory [1] and may be simply evaluated in the photonic basis approach for quantum dynamics. A simple classical model explains the observed scaling but such theoretical and experimental ionization thresholds differ by a factor of 2-2.5. Therefore, this effect is similar to the classical scaling of non-classical stability in the microwave ionization of Rydberg atoms [7].

Systematic investigations of ionization dependence upon the pulse duration and the number of field oscillations are desired for a deeper insight into ionization mechanisms of the quasi-classical Rydberg atoms and the quantum-classical correspondence of strongly driven non-linear systems in general. Also some resonance features in interactions of highly excited atoms with short electromagnetic pulses are expected.

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