A fresh look at 3D microwave ionization curves of hydrogen Rydberg atoms

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Abstract

Analytical arguments and numerical simulations suggest that the shapes of 3D microwave ionization curves measured by Koch and collaborators (see P. M. Koch and K. A. H. van Leeuwen, Phys. Rep. 255, 289 (1995)) depend only weakly on the angular momentum of the atoms in the initial microcanonical ensemble, but strongly on the principal quantum number and the magnetic quantum number. Based on this insight, coupled with the computational power of a high-end 60-node Beowulf PC cluster, we present the first 3D quantum calculations of microwave ionization curves in the experimentally relevant parameter regime.
When Bayfield and Koch first reported their microwave ionization experiments \[1\] the results were puzzling and appeared to be in complete contradiction with Einstein’s theory of the photoelectric effect: the experiments showed sharp ionization thresholds as a function of the field strength, not the frequency, as one might have expected. After initial confusion and unsuccessful attempts to explain the results within the framework of multiphoton theory \[2\], classical molecular dynamics calculations \[3\] and classical chaos theory \[4\] emerged as the first theories capable of providing physical insight into the most pertinent features of the ionization mechanism. Thus, about a decade after the seminal experiments by Bayfield and Koch the reason for the surprising behavior of the hydrogen atom in a strong microwave field was finally discovered: chaos. By now it is firmly established that the hydrogen atom in a strong microwave field is a classically chaotic system \[5,6\]. Since nowadays quantum systems chaotic in their classical limit are called “quantum chaotic” \[7\], the Bayfield and Koch experiments were the first quantum chaos experiments.

After an initial flurry of activity in the field of microwave ionization of hydrogen Rydberg atoms in the late 1970s and in the 1980s, progress slowed down considerably. This is mainly due to the fact that neither the original Bayfield and Koch experiment \[1\], nor the successor experiments by Koch and collaborators at Stony Brook \[8\] are fully quantum number resolved experiments. Only the principal quantum number of the Rydberg atoms is known before the atoms are irradiated by the microwave field \[8\]. This renders these experiments effectively three-dimensional (3D), which poses formidable problems for classical and quantum theory alike. Based on new insight into the ionization mechanism of 3D hydrogen atoms and aided by a 60-node Beowulf cluster computer built by the authors at Wesleyan University \[9\], it has only recently become possible to approach the 3D microwave ionization problem of hydrogen Rydberg atoms in the experimentally relevant parameter regime \[10\]. In this paper we report the results of the first three-way comparison between experiment, 3D classical and 3D quantum theory for microwave ionization curves of \(n_0 = 37\) hydrogen Rydberg atoms.

The experiments are done in the following way \[8\]: protons are generated in a plasma ion source and accelerated to an energy of about 14 keV. In a charge exchange cell they
pick up an electron which is subsequently excited into a high \( n_0 \) Rydberg state \((n_0 = 24, \ldots, 90)\). Following this preparation stage the atoms fly towards a microwave cavity. On their way to the cavity the atoms experience stray electric and magnetic fields. No controlled, state-defining electric or magnetic fields are switched on. The stray fields are not strong enough to change the principal quantum number of the atoms, but strong enough to produce a statistical mixture of the substates of the hydrogenic atomic shell defined by the principal quantum number \( n_0 \). This statistical mixture was demonstrated to be consistent with a micro-canonical ensemble \([8]\), i.e. all angular momentum quantum numbers \((l_0)\) and magnetic quantum numbers \((m_0)\) occur with equal probability in the initial ensemble.

Thus prepared the atoms fly into a cylindrical microwave cavity where they experience a total of about 500 cycles of a 9.92 GHz microwave field linearly polarized along the beam axis and operated in the TM\(_{020}\) mode. The linear polarization is an important boon for classical and quantum calculations. It means that \( m \) is a good quantum number. Angular momentum, however, is strongly mixed by the microwave field. This can be understood intuitively on the basis of the structure of the angular momentum coupling matrix \( \langle lm|z|l'm \rangle \sim \delta_{l',l+1} + \delta_{l',l-1} \). It is a tridiagonal matrix coupling nearest neighbors strongly with the same weights. Its eigenvectors are broad states, supporting the strong coupling argument. Thus we expect strong \( l \) mixing after only a few microwave cycles and consequently a microwave ionization probability that is independent of \( l_0 \) to a very good approximation. We confirmed this analytical argument with detailed numerical classical and quantum calculations. Both the analytical arguments and the numerical simulations lead us to the conclusion that the measured microwave ionization threshold fields depend only weakly on \( l_0 \), but strongly on \( n_0 \) and the magnetic quantum number \( m_0 \). This is the central physical insight that forms the basis of the computations reported in this paper.

In atomic units and to a good approximation the Hamiltonian describing the experiments by Koch \textit{et al.} \([8]\) is given by

\[
\hat{H} = \frac{\hat{p}^2}{2} - \frac{1}{\hat{r}} + \epsilon \hat{z} f(t) \sin(\omega t),
\]

(1)
where $\hat{p}$ is the electron momentum, $\hat{r}$ is the distance of the electron from the proton, $\epsilon$ is the microwave field strength, $z$ is the electron coordinate with respect to the proton in beam direction, $\omega$ is the microwave frequency and

$$f(t') = \left[ 1 + \exp \left( -\frac{t' - 92.22}{13.35} \right) \right]^{-1} - \left[ 1 + \exp \left( -\frac{t' - 409.72}{15.86} \right) \right]^{-1}$$

(2)
is an envelope function that realistically models the switch-on and switch-off stages of the atom upon entering and leaving the microwave cavity [8]. The variable $t'$ is time measured in number of field cycles.

We perform quantum calculations in a discrete basis of normalizable Stark states. We choose the extremal Stark state to represent the $l$ manifold for a given $n_0$. The hydrogen wave function is expanded into a set of extremal Stark states and the coupled linear equations resulting from the time-dependent Schrödinger equation are propagated forward in time using a fourth-order Runge-Kutta integrator method.

Modeling the experimental situation, ionization is implemented via an absorbing boundary condition located at $n = n_c = 91$. This is the experimental cut-off $n$ beyond which the experimental detection apparatus counts atoms as being “ionized” [8]. For $n_0 = 37$, converged calculations with respect to basis size (see below) are obtained by using a basis of $n = 30\ldots100$. In order to avoid reflections from our basis end at $n = 100$ we set the high $n$ part of the wave function to 0 after each microwave cycle. Although the cut-off $n$ inside of the microwave cavity is considerably higher than $n_c = 91$, we checked explicitly with computations including up to 150 states that this procedure is permissible, since the chance of a highly excited state feeding back into low-$n$ states is very small. This is consistent with the results of early classical calculations [3]. The ionization probability is then calculated according to $P_I = 1 - P_b$, where $P_b$ is the probability to remain in the bounded states with $n = 30\ldots90$.

The ionization probability data taken by Koch et al. [8] are reported as a function of the electric field amplitude at the center of the microwave cavity. Over the radius of the beam the amplitude of the TM$_{020}$ mode in the microwave cavity drops approximately 7% [8]. This
is called radial droop. We account for the radial droop by integrating the atom density of the beam times the local microwave field over the cross section of the beam. Since we calculate the ionization probability in steps of 1 V/cm, the integration reduces to a weighted sum.

Figure 1 shows the experimental ionization curve together with radial droop corrected quantum mechanical ionization curves for eight different values of $m_0$ for $n_0 = 37$. We see that the ionization threshold fields increase with increasing $m_0$. This is expected since the Stark splitting of levels decreases with increasing $m$. Figure 1 shows that the ionization threshold depends strongly on $m_0$.

Taking the strong $m_0$ dependence into account is essential for accurately reproducing the experimental ionization curve. Since $m$ is a conserved quantum number and the experiment uses an ensemble of atoms with equi-distribution in $m$, our approach is to find the ionization probability for fixed $m_0$ and then average over all $m_0$. Assuming $l_0$-independence leaves us with 37 different calculations corresponding to $|m_0| = 0, \ldots, 36$. Each $|m_0|$ corresponds to $2(37 - |m_0|)$ different $l_0$ quantum numbers. Since the $|m_0|$-dependence of the ionization signal is rather smooth, we performed calculations for $|m_0| = 0, 5, 10, 15, 20, 25, 30, 35$ only. Each $|m_0| \neq 0, 35$ represents two ranges of $m$-values, $\{|m_0| - 2, \ldots, |m_0| + 2\}$ and $\{-|m_0| - 2, \ldots, -|m_0| + 2\}$. $|m_0| = 0$ represents $m = -2, \ldots, 2$ and $|m_0| = 35$ represents $m = \pm 33, \pm 34, \pm 35, \pm 36$. The corresponding weights for the eight $m_0$ values computed are then given by $(370 - 10|m_0|)/1369$ for $|m_0| \neq 0$ and $179/1369$ for $|m_0| = 0$. Although performed in parallel on our 60-node Beowulf cluster consisting of high-end PIII and Athlon processors the quantum computations reported in this paper still took several months of CPU time to complete. This is mainly due to the fact that we accurately included the experimental envelope function (2) and integrated over the full number of field cycles (about 500 per electric field value) used in the experiments.

In addition to the quantum mechanical calculations we performed classical molecular dynamics calculations for the $n_0 = 37$ case. We used the procedures and methods described in [3] and represented the microcanonical distribution with 25 classical trajectories.
per microwave field value.

Figure 2 shows the experimental ionization curve (broken line) together with the results of our 3D classical (dots) and 3D quantum (dashed line) calculations. As far as the main rise of the ionization signal is concerned both the classical and the quantum calculations agree very well with the experimental results. In particular, all three methods find the onset of the main rise of the ionization signal to occur very close to 345 V/cm. The classical curve does not capture the pre-threshold ionization structure in the experimental data. But this is a known and therefore expected result. While the quantum mechanical computations are in satisfactory agreement with the experimental results as far as the main rise of the ionization signal is concerned, the quantum computations overestimate the ionization signal in the pre-threshold region. We checked explicitly that while \( l_0 \)-independence holds to a very good approximation in the field region of the main rise of the ionization signal, it does not hold at all in the pre-threshold region. This explains the discrepancy of the two ionization signals and (i) calls for quantum calculations with explicit \( l_0 \)-dependence in the pre-threshold region and (ii) points to the fact that the ionization mechanism in the pre-threshold region is very different from the mechanism active in the region of the main rise of the ionization signal. Realistic quantum computations including explicit \( l_0 \)-dependence, however, are currently beyond our computational means.

Discussing our results we note that the choice of basis end is not critical. We chose it small enough to make the problem tractable, but large enough to achieve convergence. This is illustrated in Figure 3, which shows the ionization probability for \( m = 0 \) in bases of \( n = 30 \ldots 100 \) (full line) and \( n = 30 \ldots 150 \) (broken line). Both bases yield comparable results.

We do not include any continuum states in our quantum calculations. All ionization occurs via excitation of high \( n \) states through the absorbing boundary condition at \( n = n_c \). In reality, of course, all bounded states are also directly coupled to the continuum by the microwave field and at first glance neglecting these couplings seems like a bad approximation. This is certainly true in a situation where the main ionization channels are via direct low-
order multiphoton transitions to the continuum. This is not the case here where chaotic diffusion and stochastic ionization \[5,6\] are the dominant processes. In this case ionization mainly proceeds through high-lying Rydberg states adequately modeled by our absorbing boundary at \(n = n_c\). A further argument is the following. In retrospect, assuming that the experimental results are accurate, this picture should be true to a good approximation. Otherwise our ionization curves would lie well below the experimental curves, the difference being “true ionization” via direct transitions to the continuum, not captured by our model. We acknowledge, however, that determining the branching ratio between these two ionization channels is an important but challenging task.

The \(n_0 = 37\) experiments at 9.92 GHz fall into the low-frequency or adiabatic regime (Regime-II in \[8\]). This regime is characterized by structure of quantum mechanical origin, here manifesting itself as a prominent pre-threshold bump. For \(n_0 \geq 70\) (approximately) these features disappear and all the experiments are in impressive agreement with classical 3D calculations (Regime-III in \[8\]). As such, the intermediate \(n_0\) are the most interesting to explore in the model presented here. Still, the \(l_0\)-independence argument holds for all \(n_0\). Therefore we expect to be able to model the experimental data for all initial principal quantum numbers.

In this paper we discussed a new model for reproducing measured microwave ionization curves of hydrogen Rydberg atoms. We presented the first 3D quantum mechanical calculations capable of reproducing experimental ionization curves in the experimentally relevant parameter regime. In the case of \(n_0 = 37\) we are able to accurately reproduce the ionization threshold and the shape of the ionization curve following the onset of the main rise of the ionization signal. In addition we qualitatively capture the pre-threshold ionization structure.

We expect that our model works for all of the 9.92 GHz microwave ionization data so far reported in the literature \[8\]. Preliminary calculations show that equally satisfying results are obtained for principal quantum numbers different from \(n_0 = 37\).

We suggest that experiments be carried out with initial selection of the magnetic quantum number. We predict ionization curves from such experiments to closely follow the curves
shown in Figure 1. This would lend credibility to the model presented here and allow further studies of the details of the ionization mechanism.

An open question is whether narrow “spikes” present in the computed ionization signals [10] are real. Indeed, much of our enthusiasm for performing realistic 3D quantum calculations draws from the desire to settle this question. The question of the spikes may, of course, also be attacked experimentally. In current microwave ionization experiments, however, these narrow features are washed out by the radial droop in the cavity field and by the presence of many $m$ quantum numbers in the initial state. Therefore experiments addressing the spikes should include $m$ selection and work with a narrow beam in order to reduce the radial droop of the microwave field. According to our calculations [10] a radial droop of less than 1 V/cm may be necessary.

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Figure Captions

**Fig. 1:** Ionization probability of microwave-driven hydrogen Rydberg atoms initially prepared in $n_0 = 37$ as a function of microwave field strength. Broken line: experimental result. Full lines: quantum mechanical extremal Stark state calculations for $m_0 = 0, 5, 10, 15, 20, 25, 30, 35$ with radial droop correction. The curves occur in sequence from the left-most ($m_0 = 0$) to the right-most ($m_0 = 35$).

**Fig. 2:** Ionization probability of microwave-driven hydrogen Rydberg atoms initially prepared in $n_0 = 37$ as a function of microwave field strength. Full line: experimental result; broken line: extremal Stark state calculation; dots: classical calculation. Both classical and quantum calculations are $m$ averaged and corrected for radial droop of the microwave field in the microwave cavity.

**Fig. 3:** Ionization probability of microwave-driven hydrogen Rydberg atoms initially prepared in $n_0 = 37$ and $m_0 = 10$ as a function of microwave field strength. Full line: basis $n = 30 \ldots 100$ (same as Figure 1). Broken line: basis $n = 30 \ldots 150$. 
