

Realistic three-dimensional computations of microwave-ionization curves of hydrogen Rydberg atoms

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We report a realistic computation of experimentally measured microwave ionization curves of hydrogen Rydberg atoms. The experimental field envelope, the duration of the microwave irradiation, the radial droop of the field in the cavity, and the microcanonical distribution of experimental initial states is completely taken into account. We obtain a strong dependence of the ionization probability on the initial magnetic quantum number. Also, a complete 3D three-way comparison between experiment, quantum, and classical theory in the experimentally relevant parameter regime is presented.

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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 activities 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 co-workers at Stony Brook (see a review of these experiments in [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 a 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 that is subsequently excited into a high n_0 Rydberg state ($n_0=24, \dots, 90$). 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 microcanonical 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 and co-workers (see, e.g., [8]) is given by

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

where \hat{p} is the electron momentum, \hat{r} is the distance of the electron from the proton, ϵ is the microwave field strength, z is the electron coordinate with respect to the proton in beam direction, ω is the microwave frequency and

$$f(t') = \left[1 + \exp\left(-\frac{t' - 92.22}{13.35}\right) \right]^{-1} - \left[1 + \exp\left(-\frac{t' - 409.72}{15.86}\right) \right]^{-1} \quad (2)$$

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 cutoff 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 $n = 30, \dots, 100$. 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 cutoff 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 $n = 30, \dots, 90$.

The ionization probability data taken by Koch and co-workers 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

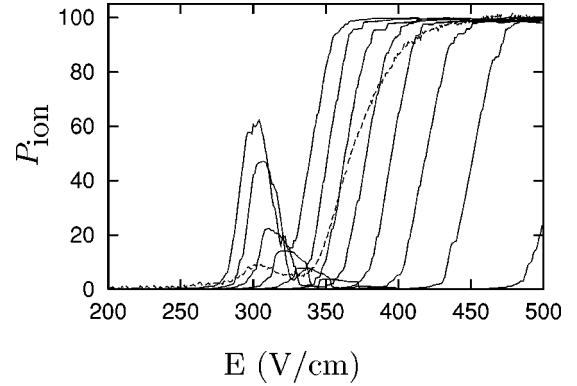


FIG. 1. Ionization probability of microwave-driven hydrogen Rydberg atoms initially prepared in $n_0 = 37$ as a function of microwave field strength. Broken line: experimental result. Full lines: quantum-mechanical extremal Stark state calculations for $m_0 = 0, 5, 10, 15, 20, 25, 30, 35$ with radial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

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 equidistribution in m_0 , 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 $|m_0| = 0, \dots, 36$. 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_0 values, $\{|m_0| - 2, \dots, |m_0| + 2\}$ and $\{-|m_0| - 2, \dots, -|m_0| + 2\}$, $|m_0| = 0$ represents $m = -2, \dots, 2$ 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), integrated over the full number of field cycles (about 500 per electric field value) used in the experiments, and that we have a fine enough grid in field values (1 V/cm) to be able to accurately model the radial droop. In addition, we address the problem of the microcanonical distribution by including the strong m_0 dependence of the ionization curves.

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.

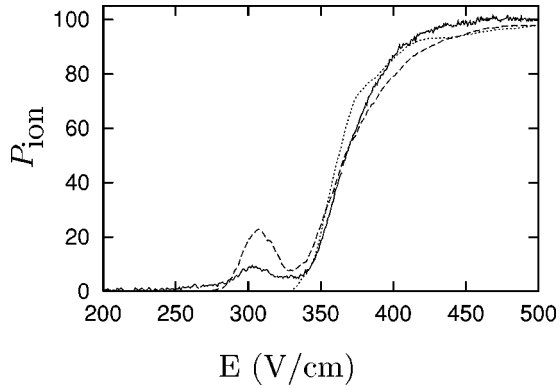


FIG. 2. Ionization probability of microwave-driven hydrogen Rydberg atoms initially prepared in $n_0=37$ as a function of microwave field strength. Full line: experimental result; dashed line: extremal Stark state calculation; dots: classical calculation. Both classical and quantum calculations are m_0 averaged and corrected for radial droop of the microwave field in the microwave cavity.

Figure 2 shows the experimental ionization curve (full 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 prethreshold ionization structure in the experimental data. But this is a known [8] 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 prethreshold 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 prethreshold region. This explains the discrepancy between the two ionization signals and (i) calls for quantum calculations with explicit l_0 dependence in the prethreshold region and (ii) points to the fact that the ionization mechanism in the prethreshold 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. We ran calculations with larger basis sizes (e.g., $n=30, \dots, 150$) and obtained final ionization curves differing negligibly from the ones presented here.

In our calculations we neglect direct coupling to the continuum. All ionization occurs via excitation of high n states through the absorbing boundary condition at $n=n_c$. This seems justified because 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 $n=n_c$.

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 prethreshold 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 approximately 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 model for reproducing measured microwave ionization curves of hydrogen Rydberg atoms. We presented 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 prethreshold 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]. We have performed preliminary calculations that show equally satisfying results for principal quantum numbers different from $n_0=37$. Our method allows us to compute ionization curves for microwave frequencies different from 9.92 GHz and for quantum numbers different from $n_0=37$ and thus allows us to predict ionization curves for parameter regimes not yet measured experimentally.

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 Fig. 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_0 quantum numbers in the initial state. Therefore experiments addressing the spikes should include m_0 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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- [1] J. E. Bayfield and P. M. Koch, *Phys. Rev. Lett.* **33**, 258 (1974).
- [2] P. M. Koch (private communication).
- [3] J. G. Leopold and I. C. Percival, *Phys. Rev. Lett.* **41**, 944 (1978); *J. Phys. B* **12**, 709 (1979).
- [4] B. I. Meerson, E. A. Oks, and P. V. Sasarov, *Pis'ma Zh. Eksp. Teor. Fiz.* **29**, 79 (1979) [*JETP Lett.* **29**, 72 (1979)].
- [5] G. Casati, B. V. Chirikov, I. Guarneri, and D. L. Shepelyansky, *Phys. Rep.* **154**, 77 (1987).
- [6] G. Casati, I. Guarneri, and D. L. Shepelyansky, *IEEE J. Quantum Electron.* **24**, 1420 (1988).
- [7] M. C. Gutzwiller, *Chaos in Classical and Quantum Mechanics* (Springer, New York, 1990).
- [8] P. M. Koch and K. A. H. van Leeuwen, *Phys. Rep.* **255**, 289 (1995).
- [9] <http://www.weswulf.org>.
- [10] Th. Clausen and R. Blümel, *Bull. Am. Phys. Soc.* **45**, 48 (2000); *Verh. Dtsch. Phys. Ges.* **35**, 1108 (2000).