# Microwave Ionization of an Atomic Electron Wave Packet

Michael W. Noel,* Lung Ko, and T. F. Gallagher
*Department of Physics, University of Virginia, Charlottesville, Virginia 22901*
(Received 22 December 2000; published 5 July 2001)

A short microwave pulse is used to ionize a lithium Rydberg wave packet launched from the core at a well-defined phase of the field. We observe a strong dependence on the relative phase between the motion of the wave packet and the oscillations of the field. This phase dependent ionization is also studied as a function of the relative frequency. Our experimental observations are in good qualitative agreement with a one-dimensional classical model of wave packet ionization.

DOI: 10.1103/PhysRevLett.87.043001 PACS numbers: 32.80.Rm, 42.50.Hz

While the physical pictures painted by classical and quantum mechanics are in stark contrast, the study of highly excited atoms has brought into focus many of the connections between the two. Of particular interest is the study of a quantum system whose classical counterpart exhibits chaotic dynamics. This situation is realized by a Rydberg atom interacting with a strong oscillating field. Studies of this system have identified the connection between the onset of chaos in the classical system and the onset of ionization [1,2], revealed regions of mixed regular and chaotic dynamics which lead to local regions of stabilization against ionization [3–7], and observed quantum localization effects where classical descriptions break down completely [8–10]. Although much of the theoretical work has been done using classical mechanics, until now the experimental work on microwave ionization has all been done starting from stationary quantum states, either Rydberg eigenstates or incoherent mixtures of Rydberg states. Comparing the results of such experiments to classical calculations requires averaging over large ensembles of initial conditions, blurring to some extent the physics.

An attractive alternative is to examine the ionization of a wave packet, which approximates a classical atom with well-defined initial conditions. Using a wave packet, it should be straightforward to probe several ideas which have emerged from the theoretical work. For example, calculations have shown that nondispersive wave packets with nearly infinite lifetimes can exist in strong microwave fields [11–13]. Of more immediate interest, there are predictions of stable orbits occupying small regions of phase space where the microwave frequency is equal to or a subharmonic of the classical Kepler frequency. By studying the ionization of wave packets launched along well-defined trajectories at well-defined phases of the microwave field, we have the opportunity to study these effects in more detail. In this Letter, we report the first experiment of this kind in which we have measured the phase dependence of microwave ionization of a wave packet localized in the radial coordinate and launched from the core at a well-defined phase of the microwave field.

Our experimental technique consisted of exciting an atomic electron wave packet in the presence of the microwave field at a well-defined phase. After $\sim$1 ns, the microwave field was switched off. Finally, we measured the number of atoms that remained bound after the microwave interaction by ionizing them with a slowly ($\sim 1\ \mu$s) rising electric field pulse and collecting the resulting electrons on a microchannel plate detector.

Laser excitation of the lithium Rydberg wave packet was done in three steps. The $2s \rightarrow 2p$ and $2p \rightarrow 3s$ transitions were driven with a pair of 5 ns pulsed dye lasers. The Rydberg wave packet was then excited by driving the $3s \rightarrow \bar{n}p$ transition with a 5 ps dye laser pulse. Here, $\bar{n}$ is the average principal quantum number of the wave packet. This pulse was generated by amplifying, in a three stage dye amplifier running at 20 Hz, the pulses generated in a mode-locked dye laser running at 76 MHz. The central frequency of this laser could be tuned to excite wave packets between $\bar{n} = 53$ and 86, and the laser had a bandwidth sufficient to excite between 4 and 13 states over this range. All of the lasers were polarized in the same direction as the microwave field.

The arrival time of the mode-locked laser pulses at the interaction region was synchronized with the 11 GHz microwave field oscillations by phase locking the microwave oscillator to the 145th harmonic of the 76 MHz mode-locked pulse train. We detected part of the unamplified mode-locked pulse train with a photodiode with 100 ps response time. The signal produced was a comb of frequencies at harmonics of 76 MHz and extending beyond 11 GHz, which was mixed with the continuous wave 11 GHz output of the microwave oscillator. The intermediate frequency signal from the mixer was low-pass filtered to produce an error signal which was fed into the frequency modulation port of the microwave oscillator, phase locking it to the 145th harmonic of the mode-locked train.

The phase-locked microwave field was turned off in $\sim$3 cycles by mixing with a square pulse whose trailing edge arrived $\sim$1 ns after the arrival of the amplified laser pulse. After three stages of amplification, the microwave pulse was sent into our vacuum chamber via coaxial cable and coupled into a short piece of WR90 waveguide. The microwave pulse was then coupled out of the opposite end of the waveguide and continuously monitored on a sampling

  

oscilloscope to ensure the quality of the phase locking. The waveguide had a small hole in one of its sides, through which the lithium atoms entered, opposed by a second small hole in the opposite side where the laser beams entered. The atoms passed above a thin copper horizontal septum in the waveguide, and the field ionization pulse was applied to the septum. After field ionization, electrons were extracted through a small hole in the top of the waveguide and detected with a pair of microchannel plates.

Data were taken by tuning the picosecond laser to excite a wave packet at the desired central frequency and adjusting the amplitude of the microwave field to ionize half of the atoms. The relative phase between the wave packet and the microwave field was then scanned by adjusting the arrival time of the laser pulse using an optical delay line. The delay was scanned over 130 ps, corresponding to 1.4 cycles of the microwave field.

Figure 1 shows delay scans for $\bar{n} = 75$, 67, and 61. The atoms are excited in the microwave field $E\cos\omega t = E\cos\phi$, and for reference the phase angle $\phi$ is also shown. The absolute phase is obtained by comparing one of the experimental curves to a calculation; however, the relative phase of the three experimental curves was not adjusted. The number of atoms ionized by the microwave field varies by up to 20% as this delay is scanned. The period of the oscillations in these scans is half that of the microwave period, consistent with the symmetry of the radial wave packet being excited. In essence, the wave packet does not care whether the field points up or down at a given time since it expands from the core as a shell with the angular distribution of a $p$ state.

In Fig. 1 it is also apparent that the depth of modulation varies with the tuning of the laser. At $\bar{n} = 75$ and 61, there is a large phase dependence, while at $\bar{n} = 67$ the ionization is relatively insensitive to the phase of the microwave field. This dependence is shown explicitly in Fig. 2 where we show the peak-to-peak modulation depth as a function of laser tuning. This plot is shown in scaled units $\omega_0 = \bar{n}^3\omega$, i.e., $\omega_0$ is simply the ratio of the microwave frequency $\omega$ to the classical Kepler frequency for the two body Coulomb problem, $1/\bar{n}^3$. It should be noted that although lithium is certainly not a two body system we believe that this scaling is appropriate since the $p$ states used in this experiment exhibit only a small quantum defect and therefore behave almost hydrogenically. Not surprisingly, the modulation depth, or phase dependence, displays structure associated with the positions of the classical subharmonic resonances at $\omega_0 = 1/3$, $1/2$, and 1. However, it appears that the ionization is least sensitive to phase at the classical resonances and most sensitive in between. In an effort to understand this behavior, we draw on insight gained from previous experiments on ionization of stationary Rydberg states, examine the phase dependence of the final state distributions for wave packet ionization, and compare the observed phase dependence with a simple classical model.

In Ref. [7] we reported ionization of Rydberg eigenstates with a $1/2$ ns, 11.5 GHz microwave pulse. The field amplitude necessary to ionize 50% of the atoms shows a series of flat steps near $\omega_0 = 1/3$, $1/2$, and 1, indicating that the ionizing field is insensitive to $\omega_0$ in these regions. In contrast, the amount of ionization at a given

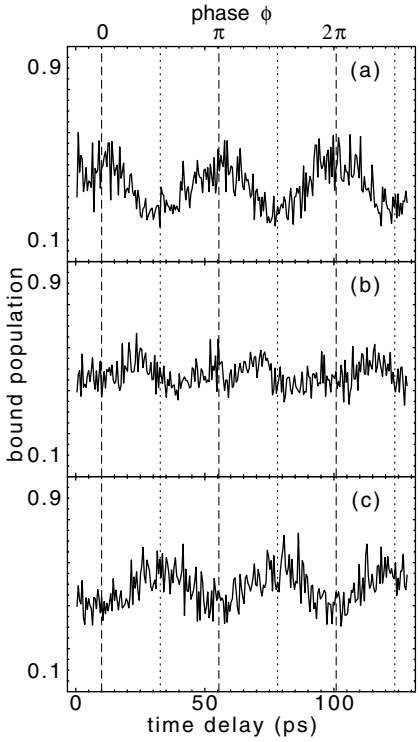


FIG. 1. Normalized remaining atom signals as a function of time delay of the mode-locked laser pulse for (a) $\bar{n} = 75$, (b) $\bar{n} = 67$, and (c) $\bar{n} = 61$.

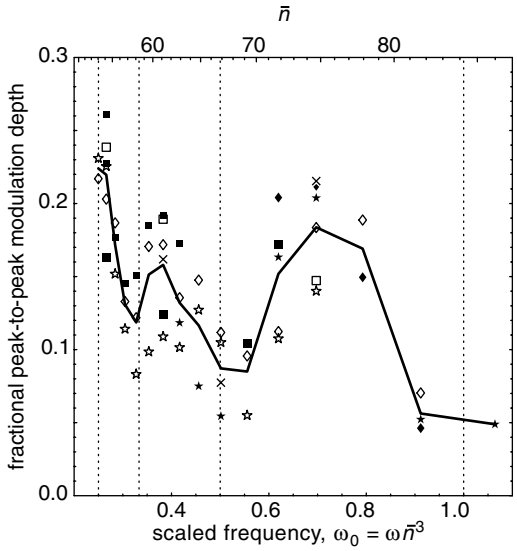


FIG. 2. Peak-to-peak modulation depth of the phase dependent ionization signals plotted versus scaled frequency. Different symbols represent data taken on different days and the solid line is the average of all data sets taken at a given laser tuning.

field amplitude depends strongly on $\omega_0$ for the regions between classical resonances. Furthermore, analysis of the final bound states showed that population in neighboring $n$ states was pulled to the resonances, the values of $n$ for which $\omega_0 = 1/3, 1/2$, and 1. Considering the wave packet experiment in light of these results leads us naturally to the following question. When the wave packet is excited at some microwave phases, is there more population pulling to the resonances than at other phases, leading to the observed phase variation in the ionization?

To address this question experimentally we have examined the final bound state distribution, and Fig. 3 shows time resolved electron signals for atoms that remained bound after the microwave interaction for $\bar{n} = 75$, 67, and 61. Since we use a slowly rising field ionization pulse, the arrival time of the electrons at the detector can be roughly mapped onto the final state distribution. These state distributions are for the initial wave packet and the two phases indicated in Fig. 1. In Fig. 3(a), for a wave packet initially excited to $\bar{n} = 75$, above $\omega_0 = 1/2$, one phase of the field, $\phi = 0$, leaves a significant amount of population near $\bar{n} = 67(\omega_0 = 1/2)$, while the population distribution for the other phase, $\phi = \pi/2$, is left near the initial state. For an initial state near $\omega_0 = 1/2$, Fig. 3(b), the width

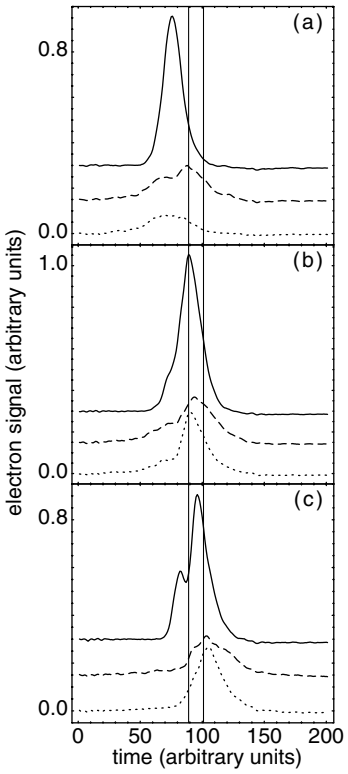


FIG. 3. State distributions for wave packets initially excited near (a) $\bar{n} = 75$, (b) $\bar{n} = 67$, and (c) $\bar{n} = 61$. The solid curve is the state distribution for the initially excited wave packet. The dashed (dotted) curve is for ionization at phases of $\phi = 0$ and $\pi/2$, indicated by the dashed (dotted) lines in Fig. 1. The vertical lines indicate the locations of $\omega_0 = 1/2$ (left) and $\omega_0 = 1/3$ (right).

of the population distribution changes with phase, but remains localized near $\bar{n} = 67$, or $\omega_0 = 1/2$. The results of Fig. 3(c), for an initial wave packet excited between the $\omega_0 = 1/2$ and $1/3$ subharmonic resonances, are somewhat less clear because the states are beginning to field ionize along both adiabatic as well as diabatic paths. Although population seems to have moved toward $\omega_0 = 1/3$, to lower $\bar{n}$, for both phases, $\phi = 0$ and $\phi = \pi/2$, it is clear that more moves for one phase, $\phi = \pi/2$, than the other. Overall, the behavior of the state distributions is consistent with the speculation drawn from the results of the eigenstate experiment.

The observations of Fig. 3 are consistent with the following simple physical picture. If a Rydberg atom is placed in a microwave field which is turned on and off slowly, it is likely that there will be no net energy transfer to the Rydberg electron. However, if the field is turned on suddenly, and then turned off slowly, it is clear that there is likely to be net energy transfer and that it will be determined by the phase of the field at which it is turned on. Exciting the wave packet at a given phase is equivalent to turning on the field with that phase. Energy transfer from the field is largest when the Rydberg electron's velocity is highest, which occurs when it is at the core, so it seems that excitation of the wave packet at $\phi = 0$ or $\pi$ will lead to the largest energy transfer, or change in $\bar{n}$. The change in $\bar{n}$ should be up in half the atoms and down in half the atoms since the electron leaves the core going in and opposite to the field direction. On the other hand, excitation at $\phi = \pi/2$ or $3\pi/2$ leads to minimal energy transfer since there is no microwave field when the electron is moving rapidly. Returning to Fig. 3(a), we can see that it is reasonable for population to be largely unaffected for $\phi = \pi/2$ and be more broadly distributed for $\phi = 0$. In particular, the sudden turn on for $\phi = 0$ has moved a significant fraction of the population to the location of the $\omega_0 = 1/2$ subharmonic resonance where it is trapped. The ionization curve in Fig. 1(c) shows more ionization at $\phi = 0$ than at $\phi = \pi/2$, in apparent contradiction with Fig. 1(a). However, the state distributions of Fig. 3(c) paint a consistent picture. For the $\phi = 0$ interaction, the remaining bound population is more broadly distributed than for $\phi = \pi/2$. From this we can conclude that the low $n$ subharmonic resonances are less effective at trapping population, and that the atoms which initially gain energy ionized leading to the net increase in ionization for the sudden turn on.

While this simple picture provides insight, it is equally useful to carry out quantitative calculations, and we present the results of a simple classical model for wave packet ionization. We integrate the equations of motion for a one-dimensional model atom for an ensemble of electrons initially placed near the atomic core. After interacting with the ten cycle field which turns off smoothly in three cycles, we count the number of atoms that remain bound as a function of the initial phase of the field. The amplitude of the field was adjusted to ionize an average of 10%

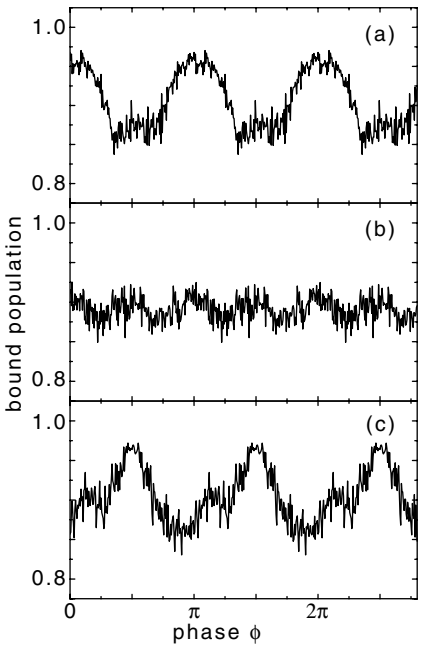


FIG. 4. One-dimensional model calculations for phase dependent ionization of wave packets near (a) $\bar{n} = 75$, (b) $\bar{n} = 67$, and (c) $\bar{n} = 61$. The average ionization level is set to 10%.

of the atoms for each case. This choice of ionization level is motivated by extensive studies of eigenstate ionization using one-dimensional models [5,14]. Figure 4 shows the results of these classical calculations for $\bar{n} = 75$, 67, and 61, which are above, at, and below the $\omega_0 = 1/2$ subharmonic resonance. The essential features of the data in Fig. 1 agree quite well with these calculations, and it is from these calculations that we determined the absolute phases shown in Figs. 1 and 3. A relatively large phase dependence is observed for $\bar{n} = 75$ and 61, and very little phase dependence is seen at $\bar{n} = 67$. The experimentally observed 90° shift of the phase dependent response between $\bar{n} = 75$ and 61 is also reproduced by this model. At $\bar{n} = 67$, the $\omega_0 = 1/2$ subharmonic resonance, the residual phase dependent modulation appears to have doubled in frequency in both the classical calculations and the experimental data.

In conclusion, we have measured the phase and frequency dependence of wave packet ionization by a short microwave pulse. A one-dimensional classical model is presented, which agrees qualitatively with most of the features of our data. These results verify the importance of synchronization in both phase and scaled frequency between a localized initial state and an ionizing microwave field. We believe this line of experimentation will open the door to answering fascinating open questions such as the formation of localized states by the microwave field itself, as suggested in Ref. [11], and the dynamics of the initially localized state in this experiment [15].

This work was supported by the Air Force Office of Scientific Research and the National Science Foundation. We thank W. M. Griffith for useful discussions.

*Current address: Department of Physics, Bryn Mawr College, Bryn Mawr, PA 19010.

[1] Roderick V. Jensen, Phys. Rev. A **30**, 386 (1984).
[2] K. A. H. van Leeuwen, G. v. Oppen, S. Renwick, J. B. Bowlin, P. M. Koch, R. V. Jensen, O. Rath, D. Richards, and J. G. Leopold, Phys. Rev. Lett. **55**, 2231 (1985).
[3] R. C. Jensen, Phys. Scr. **35**, 668 (1987).
[4] James E. Bayfield and David W. Sokol, Phys. Rev. Lett. **61**, 2007 (1988).
[5] R. V. Jensen, S. M. Susskind, and M. M. Sanders, Phys. Rep. **201**, 1 (1991).
[6] P. M. Koch and K. A. H. van Leeuwen, Phys. Rep. **255**, 289 (1995).
[7] Michael W. Noel, W. M. Griffith, and T. F. Gallagher, Phys. Rev. A **62**, 063401 (2000).
[8] E. J. Galvez, B. E. Sauer, L. Moorman, P. M. Koch, and D. Richards, Phys. Rev. Lett. **61**, 2011 (1988).
[9] J. E. Bayfield, G. Casati, I. Guarneri, and D. W. Sokol, Phys. Rev. Lett. **63**, 364 (1989).
[10] R. Blumel, A. Buchleitner, R. Graham, L. Sirko, U. Smilansky, and H. Walther, Phys. Rev. A **44**, 4521 (1991).
[11] Andreas Buchleitner and Dominique Delande, Phys. Rev. Lett. **75**, 1487 (1995).
[12] Andreas Buchleitner and Dominique Delande, Chaos Solitons Fractals **5**, 1125 (1995).
[13] Andreas Buchleitner, Dominique Delande, Jakub Zakrzewski, Rosario N. Mantegna, Markus Arndt, and Herbert Walther, Phys. Rev. Lett. **75**, 3818 (1995).
[14] D. Richards, in *Classical Dynamics in Atomic and Molecular Physics,* edited by T. Grozdanov, P. Grujić, and P. Krstić (World Scientific, Singapore, 1988) p. 269.
[15] Tsin-Fu Jiang and Shih-I Chu, Chem. Phys. Lett. **240**, 278 (1995).