Ionization of Rydberg Atoms by Subpicosecond Half-Cycle Electromagnetic Pulses

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We have ionized Rydberg atoms using subpicosecond half-cycle electromagnetic pulses. The threshold electric field required to ionize a Rydberg state with effective quantum number n^* is found to scale as n^{*-2} for states with $n^* > 13$ in contradistinction to the n^{*-4} threshold scaling for static field ionization and high order multiphoton ionization. This novel result is explained using a classical model.

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The ionization of atoms by pulsed electromagnetic radiation has been studied using pulsed lasers [1], microwaves [2,3], and ramped dc electric fields [4]. However, until very recently, the temporal width of available pulses was quite large compared to the internal time scales of the atoms under study. In the last decade, however, ultrashort (< 100 fs) laser pulses have been produced, which have durations of only tens of optical cycles [5,6]. Ionization in these laser fields can differ drastically from ionization with longer pulses because the large laser bandwidth may coherently excite several atomic states [7].

We have extended the study of ionization by coherent broadband laser pulses to its logical limit by employing ultrashort, half-cycle electromagnetic pulses. Half "optical-cycle" pulses with widths $\tau < 200$ fs have been created in photoconducting semiconductor switches illuminated by 100 fs laser pulses [8]. These freely propagating electromagnetic pulses have central frequencies around 1 THz (33 cm^{-1}). We have succeeded in increasing the available pulse energy by more than an order of magnitude, and have demonstrated that peak fields in excess of 100 kV/cm can be produced in a nearly unipolar 500 fs electric field pulse [9]. While these fields are still insufficient to ionize the ground state of atoms, they are quite capable of ionizing high-lying Rydberg states.

We have studied the ionization of Na Rydberg atoms using freely propagating, 500 fs single-polarity pulses. Complete ionization of Rydberg states with principle quantum number $n > 13$ is observed. We show that Rydberg states subjected to these short pulses begin to ionize when the peak electric field is proportional to their binding energies. In contrast, ionization of atoms by relatively long (μs) microwave pulses and ramped dc electric fields requires peak fields which scale as n^{-5} and n^{-4}, respectively [3,4]. The novel threshold field scaling can be explained by classical mechanics.

In the experiment, two tunable dye lasers excite ground state Na atoms to a Rydberg state. The half-cycle pulse (HCP) is then weakly focused on the Rydberg atoms. Any ions formed by the HCP are collected using a microchannel plate (MCP) detector. We record the number of ions (or electrons) which are produced as a function of the peak electric field in the HCP.

The source of the HCP [10] is a thin (0.5 mm) GaAs semiconductor wafer with a surface area of ~ 3 cm^2. An electric field ($F < 10$ kV/cm) is applied parallel to the surface of the GaAs. The electric field is then shorted across the semiconductor surface by illuminating one side of the wafer with 100 fs, 770 nm laser pulse, which drives the GaAs into conduction. A substantial fraction of the radiated energy from the rapidly accelerating electrons in the photoconductor is transmitted through the wafer in the form of a spatially coherent electromagnetic pulse. At least 80% of the energy in the coherent radiation resides in a $\tau_{HCP} \sim 500$ fs, single-polarity pulse. This HCP is polarized in the direction of the bias field in the wafer and has a bandwidth of ~ 1 THz. The peak electric field in the HCP is proportional to the bias field. An electric field autocorrelator [8] allows us to determine the bandwidth of the HCP, and cross correlation techniques are used to infer temporal pulse shapes [9]. A typical interferogram, frequency spectrum, and fitted electric field shape are shown in Fig. 1 [9].

The ultrashort laser system used to generate the HCPs is quite involved, and has been described elsewhere [6].

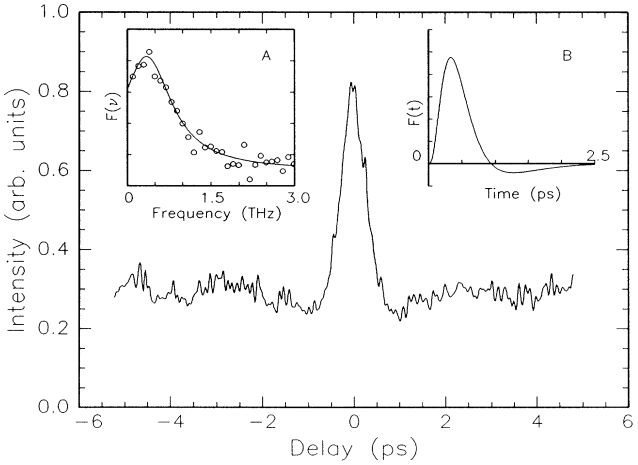

FIG. 1. Electric field autocorrelation as a function of delay in one leg of the Michelson interferometer. Inset (a): Frequency spectrum of the HCP. Inset (b): Inferred temporal pulse shape of the HCP.

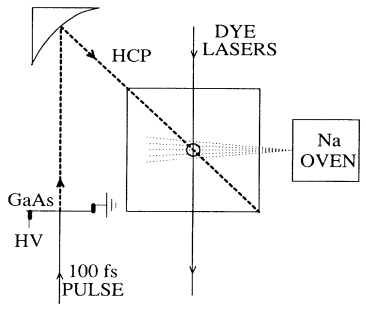

FIG. 2. Schematic diagram of the interaction region shown from above. The HCP is produced by applying a 100 fs laser pulse to the GaAs wafer which shorts the high voltage (HV) across the photoconducting surface. The transmitted radiation is weakly focused with a parabolic mirror. The two pulsed dye lasers and the HCP interact with the Na beam between two parallel conducting plates.

Briefly, the 100 fs, 770 nm laser pulses originate in a self-mode-locked Ti:Al_2O_3 oscillator and are amplified in a chirped pulse amplifier (CPA). Pulse energies of 10 mJ are available; however, we find that GaAs generates HCPs with maximum field amplitudes for a laser fluence on the wafer surface of only 40 $\mu J/cm^2$ [9].

Na atoms are produced in *ns* or *nd* Rydberg states via two photon excitation through the $3p_{1/2}$ resonance line. The frequency of the second photon is tuned to excite a single Rydberg state between $n=12$ and 50. Both photons are linearly polarized in the same direction, and we excite states with $|m_j| = \frac{1}{2}$ only, where m_j is the quantum number associated with the projection of the total angular momentum along the laser polarization axis. The thermal Na beam originates in a resistively heated oven in a vacuum chamber with a base pressure of 5×10^{-7} Torr. The HCP is generated in the vacuum chamber and the orientation of the various beams and components is shown in Fig. 2. The HCP enters the interaction region $\sim$50 ns after the Rydberg states have been excited. The pulse, which is linearly polarized in the same direction as the dye lasers, is weakly focused by a gold parabolic mirror to a 6 mm waist at the laser-atom interaction region between two parallel capacitor plates. 100 ns after the HCP, a small ($\sim$50 V/cm) electric field is applied between the capacitor plates forcing any ions formed by the HCP through a 2 mm diam hole in the upper plate towards the MCP. The small extraction hole allows us to ignore any spatial variation in the HCP. The change in the ion current is recorded as a function of peak field in the HCP as the bias field on the GaAs wafer is varied.

Several typical electric field scans are shown in Fig. 3. Here, the dye lasers are tuned to excite the 35*d*, 20*d*, or 15*d* state. Note that the ionization thresholds are quite broad. This effect is not due to experimental artifacts, but instead is inherent in the ionization process as discussed below.

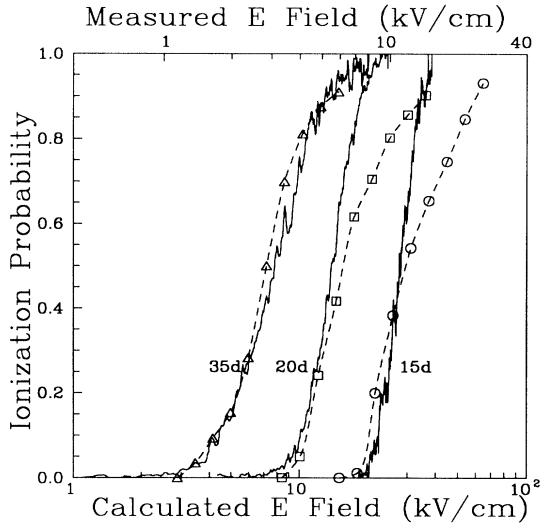

FIG. 3. Measured (solid curves) and calculated (dashed curves) ionization probabilities for the 30*d*, 20*d*, and 15*d* states as a function of the peak electric field in the HCP. The electric field scales for the numerical and experimental data differ by a factor of 2.5. Note the broad ionization thresholds which are intrinsic to the ionization process.

Three independent sets of threshold measurements were made using two different GaAs photoconductors in identical configurations. The energy and waist of the HCP at the laser-atom interaction region were measured outside of the vacuum chamber. The calibration of the peak electric field in the pulse assumed a Gaussian temporal profile with the width of the large half cycle shown in Fig. 1(b). The calibration gives a peak electric field in the interaction region which is 25% higher than the bias field applied across the GaAs surface with an estimated 90% of the radiated energy contributing to the HCP (see inset, Fig. 1).

Figure 4 shows the measured electric fields needed to ionize 10% and 50% of the Rydberg state population. The data are the average of the three runs. The field needed to ionize 10% of the *s* or *d* states follows a clear n^{*-2} scaling, where the effective quantum number n^* is defined in terms of the Rydberg state energy $E=-\frac{1}{2}n^{*2}$ a.u. The field needed to ionize 50% of the atoms scales as $n^{*-3/2}$.

The problem of ionization of Rydberg states by a 500 fs, single-polarity pulse is quite different from ionization by a similarly shaped 500 ns pulse. The long pulse amplitude varies slowly compared to the atomic time scale (i.e., the Kepler period of a classical electron, $\tau_K=2\pi n^{*3}$ a.u.). The energy levels of the atom during the pulse are approximately equal to the static field Stark states. With increasing electric field, population in a given Rydberg state moves out along one (adiabatic) or several (diabatic) discrete energy levels until the classical field ionization limit is reached. This field is proportional to n^{*-4}.

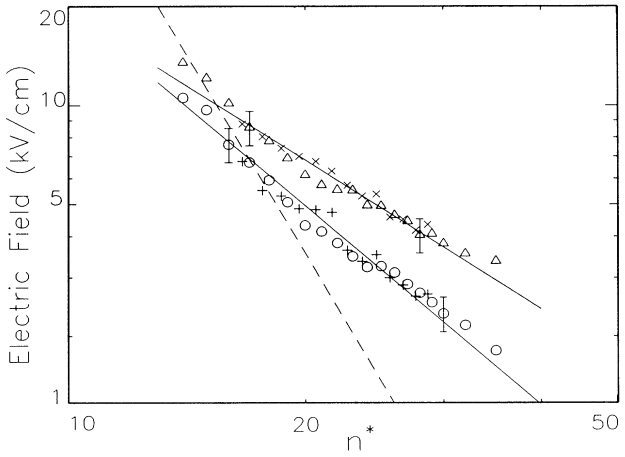

FIG. 4. Experimental ionization thresholds as a function of peak field in the HCP. (○: d states, 10% ionization; +: s states, 10% ionization; △: d states, 50% ionization; ×: s states, 50% ionization). The data clearly show the broadening of the thresholds at higher values of n^*. The solid lines drawn through the data have slopes of -2 and $-\frac{3}{2}$ for 10% and 50% ionization, respectively. The dashed line shows the "long pulse" diabatic field ionization limit $\frac{1}{9}n^{*4}$.

Very little energy is exchanged between the slowly changing field and the Rydberg electron (the small amount is due to the dc Stark shift) and ionization occurs because of the lowering of the Coulomb potential barrier at large radius on one side of the atom.

In contrast, the 500 fs HCPs have a pulse duration $\tau_{\mathrm{HCP}} < \tau_K$ for all Rydberg states with $n^* > 14$. Therefore, the quasistatic ionization model fails and the ionization process must proceed through a significant exchange of energy between the Rydberg electron and the rapidly changing electric field. The lowering of the Coulomb barrier occurs briefly, but negative energy electrons remain bound to the nucleus when the electric field amplitude returns to zero. In classical terms, ionization occurs when the HCP gives the electron an energy "kick" which is greater than its binding energy in the absence of the pulsed field.

A simple classical model explains the observed energy scaling of the ionization threshold. The energy gained by a bound electron which is exposed to a time-varying electric field pulse is

$$\Delta E = -\int \mathbf{F}(t)\cdot\mathbf{v}(t)dt\,, \tag{1}$$

where $\mathbf{F}(t)$ is the electric field in the pulse, and $\mathbf{v}(t)$ is the velocity of the electron (atomic units are used unless otherwise noted). The minimum field at which ionization occurs is obtained by setting the energy gain equal to the binding energy of the atom and maximizing the integral in Eq. (1). Obviously, the integral is maximized if the velocity of the electron is antiparallel to the direction of the electric field throughout the HCP. Hence, the threshold field required to ionize an electron in a one-dimensional orbit sets a lower limit on the field required to ionize an electron in three dimensions.

At small distances from the nucleus, the velocity of a Rydberg electron is a maximum and is virtually independent of its total energy ($v=\sqrt{2/x}\,[1-O(x/n^{*2})]$). Therefore, the velocity is also independent of energy shifts comparable to the binding energy due to the HCP and Eq. (1) simplifies to

$$1/2n^{*2} = \int F(t)\sqrt{2/x(t)}\,dt\,. \tag{2}$$

Clearly, the smallest value of the peak field F_0 which satisfies Eq. (2) is obtained if the electron is located at the nucleus at the beginning or end of the HCP. Using this condition and modeling the HCP as an inverted parabola, Eq. (2) gives

$$F_0 \approx 2/5n^{*2}\tau_{\mathrm{HCP}}^{2/3}\,. \tag{3}$$

For a 500 fs HCP, $F_0 = 2.7\times10^6 n^{*-2}$ V/cm, which is in good agreement with the experimental 10% ionization result of $F_0 = 2.0\times10^6 n^{*-2}$ V/cm.

A more sophisticated analysis can be performed by numerical integration of the classical equations of motion for an electron in a Rydberg orbit which is subjected to a HCP. The formulation proceeds as follows. An electron is placed at some position in a Rydberg orbit of given energy and angular momentum. The position and velocity of the electron are tracked as the HCP turns on and then off. After the external electric field has been removed, the energy and angular momentum of the electron are recorded. The procedure is then repeated for a different initial position in the orbit. The theoretical data are normalized according to the classical probability for finding the electron at the position which is the starting point of the integration. This normalization accurately reproduces the quantum mechanical expectation values of $\langle r^2\rangle$, $\langle r\rangle$, $\langle r^{-1}\rangle$, and $\langle r^{-2}\rangle$. In this way we can evaluate the probability for ionizing a quantum electron using the classical equations of motion.

The results of the calculation exhibit the same $n^{*-2}\tau^{-2/3}$ threshold field scaling that was obtained from the one-dimensional model. The good agreement between the measured and calculated ionization probabilities is evident in Fig. 3. Note that the experimental and theoretical electric field scales in Fig. 3 differ by a factor of 2.5, so that the agreement is only qualitative. The classical result predicts that the n^{*-2} scaling is valid even for states where $\tau_{\mathrm{HCP}} \approx \tau_K$, a property which is clearly exhibited in the data.

Only those electrons which are near the nucleus during the HCP can ionize at the classical threshold. Hence, the n^{*-2} scaling is only observed for small ionization fractions due to the small probability for finding the electron near the ion core during the peak of the electric field pulse. However, unlike a short laser pulse, the HCP changes the energy of a *free* electron. Therefore, the

atomic electron can increase (or decrease) its energy at any distance from the nucleus. The extra energy needed to ionize the electron can be acquired over the entire classically allowed region and not just at small radii. In general, a higher peak field is needed to ionize electron probability which is at a large distance from the nucleus. Hence, there is an inherent width to the ionization probability due to the different energy boost that the electron receives depending on its location during the peak of the HCP. This broadening increases with n^* since the probability of finding the electron near the core decreases as n^{*-3}. Experimental verification of this effect is shown in Fig. 4 where the difference in the scaling of the 50% ionization data compared to the 10% ionization data is quite clear.

The numerical integration of the classical equations of motion also predicts a width to the ionization threshold. We can define a scaled width parameter

$$Q = 2\frac{F_{50\%} - F_{10\%}}{F_{50\%}},$$

where $F_{50\%}$ and $F_{10\%}$ are the fields at which 50% and 10% ionization occur, respectively. The experimentally determined Q parameters range from approximately 0.8 to 0.4 and are typically within 15% of the classical predictions. Figure 3 clearly shows that for the lowest values of n, the agreement is somewhat less impressive and the maximum deviation is 40%.

The results of the classical simulation reduce to those of diabatic field ionization for $\tau_{\mathrm{HCP}} > \tau_K$ and show a threshold field scaling of $\approx \frac{1}{9} n^{*4}$. Unfortunately, the maximum field amplitude which can be produced using the present apparatus is too low to observe total ionization of states with $n^* < 14$ ($\tau_{\mathrm{HCP}} \approx \tau_k$ for $n^* = 15$). Therefore, the prediction of the classical model cannot be stringently tested. However, the data are consistent with a $\frac{1}{9} n^{*4}$ scaling for the lower n values where $\tau_{\mathrm{HCP}} \sim \tau_K$.

Although the classical description of the ionization process seems to be in good agreement with our observations, a quantum description is still desirable. Unfortunately, an integration of the full time-dependent Hamiltonian is difficult because the number of states resonantly coupled to the initial state by the HCP is divergent. Single photon transitions are allowed between almost any pair of states with opposite parity. A Floquet description of the atom dressed by the time-varying HCP is completely invalid since the bandwidth of the pulse is greater than its central frequency. Of course, the features that make the quantum analysis so difficult (i.e., broad bandwidth and large density of essential states) may be responsible for the good agreement with the classical theory.

In conclusion, we have observed a new n^{*-2} threshold field scaling in the ionization of Rydberg atoms by half-cycle, subpicosecond radiation. The scaling which applies to states whose classical periods are greater than the pulse duration can be explained in terms of a classical picture. The classical model is also in good agreement with the observed width of the ionization thresholds.

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[1] G. Mainfray and C. Manus, Rep. Prog. Phys. **54**, 1333 (1991), and references therein.

[2] J. A. Bayfield and P. M. Koch, Phys. Rev. Lett. **33**, 258 (1974).

[3] P. Pillet, W. W. Smith, R. Kachru, N. H. Tran, and T. F. Gallagher, Phys. Rev. Lett. **50**, 1042 (1983); C. R. Mahon, J. L. Dexter, P. Pillet, and T. F. Gallagher, Phys. Rev. A **44**, 1859 (1991).

[4] R. F. Stebbings, C. J. Latimer, W. P. West, F. B. Dunning, and T. B. Cook, Phys. Rev. A **12**, 1453 (1975); T. F. Gallagher, L. M. Humphrey, R. M. Hill, and S. A. Edelstein, Phys. Rev. Lett. **37**, 1465 (1976); M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. Lett. **41**, 103 (1978); T. H. Jeys, G. W. Foltz, K. A. Smith, E. J. Beiting, F. G. Kellert, F. B. Dunning, and R. F. Stebbings, Phys. Rev. Lett. **44**, 390 (1980).

[5] R. L. Fork, B. I. Greene, and C. V. Shank, Appl. Phys. Lett. **38**, 671 (1981).

[6] J. Squier, F. Salin, G. Mourou, and D. Harter, Opt. Lett. **16**, 324 (1991).

[7] G. Alber, H. Ritsch, and P. Zoller, Phys. Rev. A **34**, 1058 (1986); J. Parker and C. R. Stroud, Jr., Phys. Rev. A **41**, 1602 (1990); K. Burnett, P. L. Knight, B. R. M. Piraux, and V. C. Reed, Phys. Rev. Lett. **66**, 301 (1991); R. R. Jones and P. H. Bucksbaum, Phys. Rev. Lett. **67**, 3215 (1991); L. D. Noordam, H. Stapelfeldt, D. I. Duncan, and T. F. Gallagher, Phys. Rev. Lett. **68**, 1496 (1992).

[8] B. I. Greene, J. F. Federici, D. R. Dykaar, R. R. Jones, and P. H. Bucksbaum, Appl. Phys. Lett. **59**, 893 (1991).

[9] D. You, R. R. Jones, D. R. Dykaar, and P. H. Bucksbaum, Opt. Lett. (to be published).

[10] G. Morou, C. V. Stancampiano, and D. Blumenthal, Appl. Phys. Lett. **38**, 470 (1981); D. H. Auston, K. P. Cheung, and P. R. Smith, Appl. Phys. Lett. **45**, 284 (1984); A. P. De Fonzo, M. Jarwala, and C. Lutz, Appl. Phys. Lett. **50**, 1155 (1987); Ch. Fattinger and D. Grischkowsky, Appl. Phys. Lett. **53**, 1480 (1988); P. R. Smith, D. H. Auston, and M. C. Nuss, IEEE J. Quantum Electron. **24**, 255 (1988); B. B. Hu, J. T. Darrow, X.-C. Zhang, and D. H. Auston, Appl. Phys. Lett. **56**, 886 (1990); B. I. Greene, D. R. Dykaar, and J. D. Wynn, IEEE J. Quantum Electron. (to be published).