

Effects of imperfect unipolarity on the ionization of Rydberg atoms by subpicosecond half-cycle pulses

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Rydberg atoms are ionized by nearly unipolar, subpicosecond electromagnetic pulses. Deviations from a perfectly unidirectional pulse are found to alter substantially the ionization probability as a function of peak field. Quantitative agreement between classical theory and experiment is achieved if the pulse imperfections are significantly attenuated.

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In recent experiments [1,2], Rydberg atoms have been exposed to ultrashort, electric-field pulses. The electric field in these pulses is nearly unipolar, and has a temporal shape which resembles one-half of a cycle of THz-band radiation [3]. In the experiments [1,2], the 0.5-psec duration of these "half-cycle pulses" (HCPs), τ_{HCP} , is shorter than or comparable to the classical Kepler period of the Rydberg states, $\tau_K = 2\pi n^3$. The dynamics of the ionization process in these experiments is distinctly different from that in either a long field pulse ($\tau_{\text{pulse}} > \tau_K$) [4] or a short laser pulse ($\tau_{\text{laser}} < \tau_K$) [5].

In a long electric-field pulse, essentially no energy is transferred to the Rydberg electron. Instead, ionization occurs due to the modification of the electronic binding potential [4]. In a HCP, the electron is unable to respond to the rapid changes in the binding potential, and the electron must gain energy from the field in order to escape the Coulomb attraction of the nucleus. Classically speaking, the electron receives an energy "kick" or impulse from the rapidly changing field [1],

$$\Delta E = - \int_{-\infty}^{\infty} \vec{F}(t) \cdot \vec{v}(t) dt \quad (\text{a.u.}), \quad (1)$$

where $\vec{F}(t)$ is the HCP field and $\vec{v}(t)$ is the velocity of the electron. Therefore the ionization probability is strongly dependent on the velocity or momentum distribution of the initial-state wave function as well as the temporal shape of the electric-field pulse.

The time dependence of the electric field is extremely important in the ionization of Rydberg atoms where $\tau_{\text{pulse}} \ll \tau_K$. In a Rydberg atom, the electron probability distribution is peaked far from the ion core, and the probability for finding the electron near the nucleus during the pulse is $\tau_{\text{pulse}}/\tau_K$. Therefore, in order to have an ionization probability greater than $\tau_{\text{pulse}}/\tau_K$, energy transfer between the electron and the time-dependent field must occur far from the ion core where the electron is essentially free. Equation (1) clearly shows that a free electron can gain energy from a unipolar field pulse. However, this electron cannot gain energy from a pulse whose time-integrated electric field is zero. Indeed, Rydberg atoms may be ionized with 100% efficiency by a HCP, but not by a short laser pulse [1,5].

The results of the ionization experiments [1] are in qualitative agreement with classical simulations [1,6].

The agreement is quantitative if the experimental field values are rescaled by a multiplicative factor of 2.5. A more recent field calibration suggests that the field discrepancy is actually only a factor of 1.6. A description of the differences between the two calibration techniques is given later in the paper. A possible source for the persistent disagreement between experiment and theory is the presence of small, nonunipolar field components in the HCPs used in the experiments.

This report describes an experiment which studies the effects of HCP imperfections on the ionization of Rydberg atoms. Specifically, an optical gating technique is used to substantially attenuate unwanted components in the pulse which appear after the main half-cycle [3,7]. In the following sections we describe the experimental techniques and results. Using these results, quantitative agreement between classical theory and experiment is achieved.

With the exception of the gating device, the experimental apparatus is identical to one described previously [1,2]. Rydberg atoms are produced from a thermal beam of ground-state Na atoms using two, nanosecond pulsed-dye lasers. The Rydberg atoms interact with the HCP between two parallel capacitor plates (see Fig. 1). A ~ 50 -V pulse is applied to the lower plate, pushing any ions which are formed by the HCP through a small hole in the upper field plate toward a microchannel plate detector.

The HCP is generated by illuminating a large-aperture GaAs photoconductive switch with a 120-fsec, 780-nm laser pulse [3,8]. The radiation which is transmitted through the GaAs wafer is collected by a parabolic mirror and directed into the interaction region. The parabolic mirror is used to collimate the radiation, not focus it. The diameter of the HCP beam is approximately 1 cm in the interaction region. The peak field in the HCP is varied by changing the bias voltage across the switch.

Before entering the interaction region, the HCP passes through a second GaAs wafer. We will refer to this wafer as the attenuator. As shown in Fig. 1, a second 120-fsec laser pulse travels through the interaction region antiparallel to the HCP and is absorbed in the attenuator. The frequency of the laser pulse is above the direct band gap in GaAs; therefore on absorption electrons in the crystal are promoted to the conduction band. The pres-

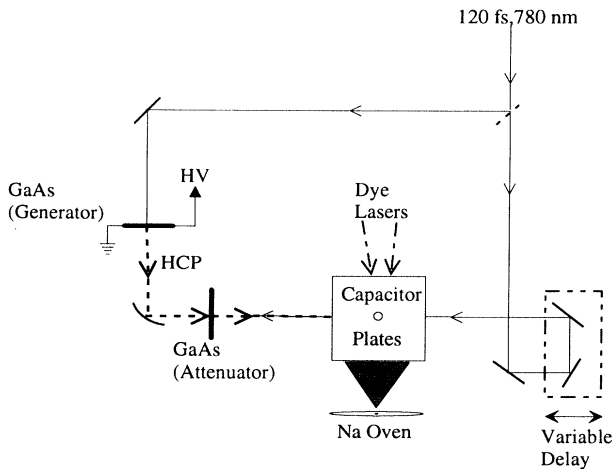


FIG. 1. Schematic of the apparatus showing the GaAs wafer which generates the HCP, the attenuating GaAs wafer, the atomic oven, and relative orientations of the various laser and HCP beam paths. The Rydberg atoms are unaffected by the unfocused 120-fsec laser pulse which travels through the interaction region to the attenuator.

ence of the essentially free electrons on the wafer surface reduces the transmission of the HCP through the wafer by a factor of ~ 10 . The attenuation persists for hundreds of picoseconds. Therefore, to a good approximation, the attenuator “slices off” any parts of the HCP which reach the attenuator surface after the laser pulse [3]. The duration of the HCP which is allowed to reach the Rydberg atoms is selected by varying the time at which the laser pulse strikes the attenuator.

Figure 2 shows the ionization signal for the $25d$ state as a function of the delay of the 120-fsec laser pulse

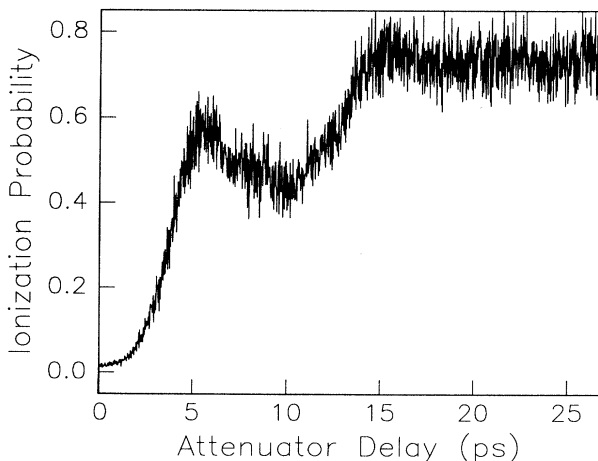


FIG. 2. Ionization signal as a function of the relative delay between the HCP generation and the laser which triggers the transient attenuator. The initial state is $25d$ and the peak HCP field is 12 kV/cm. The ionization signal rises sharply as the attenuator is delayed through the main half-cycle component in the field pulse. The decrease and then increase in the signal at later times are due to the “negative tail” and “reflection” pulse, respectively.

which is used to initiate the transient attenuation. Note that for sufficiently small delays, the entire HCP is attenuated and no ionization is observed. As the delay is increased, the ionization signal rises steadily. At still longer delays, additional signal increases and decreases can be observed. The structure in the ionization signal after the initial large increase is due to nonunipolar components in the pulse (see Fig. 3).

The temporal electric-field profile shown in Fig. 3 is inferred from a measurement of the ionization of a Rydberg atom as a function of delay between two, temporally separated (ungated) pulses. The ionization signal vs delay is similar to a cross correlation of the two pulses. For these measurements, the atom acts as a field detector. Consequently, information on the field that the atoms actually experience is obtained. The pulse shape derived from these two-pulse measurements is consistent with those obtained using conventional pulse-energy detection combined with optical gating or field autocorrelations [3].

The initial sharp increase in the ionization signal in Fig. 2 is due to the main unidirectional peak in the HCP temporal profile. The dominant feature is followed by a *negative* component to the ionization probability. This ionization inhibition is due to the “negative tail” of the field pulse which can be seen in Fig. 3. During this part of the pulse, electrons which had been promoted to the continuum by the half-cycle component of the pulse are recombined with the ion. Approximately 11 psec after the main pulse, a second increase in the ionization signal occurs. This increase is caused by a “reflection” of the main pulse due to imperfect transmission of the pulse through the radiating wafer. Figure 3 clearly shows that the field direction in this part of the pulse is opposite to that of the main pulse. However, unlike the “negative tail,” the ionization probability increases due to this reflection.

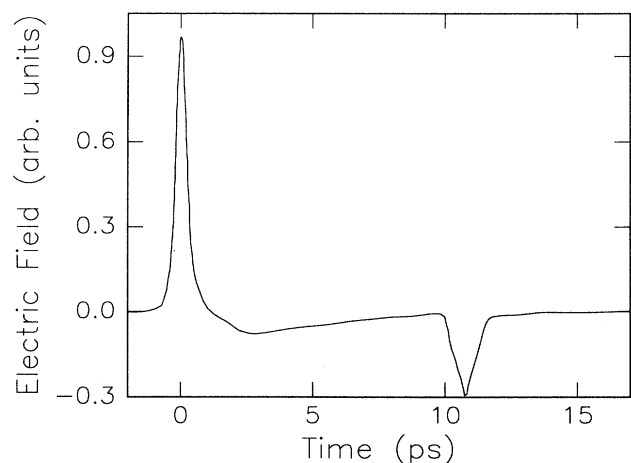


FIG. 3. Temporal shape of the HCP field inferred from measurements of ionization as a function of delay between two identical pulses. The uncertainty in the field at any time is approximately 5% of the peak field. This pulse shape is consistent with previous measurements including electric-field autocorrelations and cross correlations utilizing a transient attenuator as discussed in Ref. [3].

Evidently, the response of the Rydberg atom to the various electric-field components depends on the history of the pulse. This observation can be qualitatively understood through the use of classical mechanics. During the dominant unipolar component of the pulse, the bound electrons get a momentum "kick." This kick is in the same direction for all electrons, regardless of their initial position or momentum. If the energy transfer is large enough, some electrons will be promoted to the continuum. A fraction of these continuum electrons leave the atom directly without passing by the nucleus, while others scatter from the ion core one last time before departing. If the applied field changes direction before any of the ionized electrons scatter from the nucleus, then each of these electrons receives an additional momentum kick which decreases its energy. Therefore the ionization probability drops. However, if the field changes sign long after the initial pulse, the ionized electrons will be far from the atom. Due to ion-core scattering, field inhomogeneities, and stray fields, this field reversal is unlikely to recombine the free electrons with the atom. On the other hand, any electrons which remain bound to the nucleus have a second chance to ionize. Therefore field reversals after long delays will cause ionization enhancement. The degree to which this enhancement occurs depends on the delay, the amplitude of the additional field component, the energy of the initial Rydberg state, and other less well-characterized factors such as stray electric and magnetic fields.

Figure 2 clearly shows that the imperfectly characterized features in the HCP substantially affect the ionization probability for a given peak field. However, the primary concern of this experiment is not the determination of the exact pulse shape. Instead, the purpose of this experiment is to establish quantitatively the effects of the nonunipolar components of the HCP on the measured ionization thresholds. To this end, the ionization probability as a function of peak HCP field has been measured for several nd Rydberg states using (1) as full "unclean" (i.e., unattenuated) HCP and (2) a "clean" HCP with all but the main half-cycle component attenuated.

In Figs. 4(a) and 4(b) we plot the peak electric fields required to ionize 10% and 50% of the Rydberg population with "clean" and "unclean" HCP's. The general shapes of the ionization vs field curves are not affected by the pulse cleaning. However, the threshold fields observed with a "clean" HCP are larger by a factor of 1.3 for both 10% and 50% ionization. As mentioned above, using a different calibration, the measured fields required for ionization using an "unclean" HCP are a factor of 1.6 below those predicted using a Gaussian pulse (Ref. [1] reported a discrepancy of a factor of 2.5).

The field calibration used in Ref. [1] was based on combined measurements using bolometric (Infrared Laboratories) and pyroelectric (Molelectron P1-42cc) detectors [3]. The systematic errors acquired using that technique

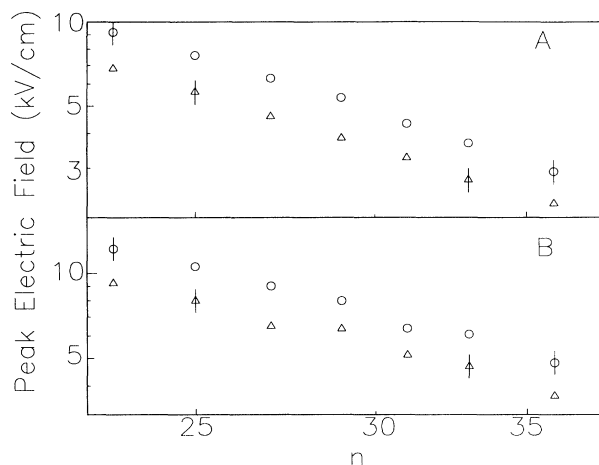


FIG. 4. Peak HCP field required to ionize (A) 10% and (B) 50% of the Rydberg population as a function of n for a (O) "clean" pulse and (Δ) "unclean" pulse. Significantly attenuating the imperfections in the pulse increases the requisite fields by a factor of 1.3.

[9] include (1) the combined deviations of the spectral response of the two detectors from their accepted values over the bandwidth of the pulse (the manufacturers suggest that this could be as much as a factor of 2 for both devices [10]); (2) geometric factors due to the different angular acceptance of the two devices (the bolometric detector includes a paraboloidal collection cone); (3) errors in the temporal pulse shape used to infer the peak electric-field amplitude from the pulse-energy measurements.

The present calibration involves direct energy measurement with the pyroelectric detector. Using this technique removes any errors due to geometric considerations as well as the response of the bolometric detector. Therefore we believe it to be more reliable. However, the manufacturer's uncertainties in the energy response of the pyroelectric detector result in a systematic error as high as a factor of 1.4. Using the present calibration, the results using the "clean" pulse agree with the classical theory to within 25%. Therefore this error is within the systematic uncertainty of the field calibration.

In conclusion, the dynamics of the ionization of Rydberg atoms by subpicosecond half-cycle pulses are significantly altered by imperfections in these pulses. An optical gating technique has been used to "clean" HCPs so that they more closely resemble pure, unipolar field pulses. Using these "clean" HCP's, the ionization probability vs peak electric field is in quantitative agreement with classical theory.

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