Displacing Rydberg Electrons: The Mono-Cycle Nature of Half-Cycle Pulses

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A THz "half-cycle" pulse is a fast (<1 ps) unipolar pulse, followed by a slow unipolar pulse of opposite polarity. We found that the interaction of such THz pulses with very high Rydberg states results in a displacement of the electron within the atom, while the ionization is strongly suppressed. In classical terms: the first fast unipolar feature corresponds to a start kick of the Rydberg electron, while the second unipolar feature acts as a stop kick. A semiclassical model is presented which qualitatively reproduces the ionization suppression and redistribution.

The first experiments with half-cycle THz pulses (HCP's) [1] revealed that the interaction of Rydberg atoms with HCP's can be described as an impulsive kick, since the duration of the half-cycle pulse (<1 ps) is much shorter than the round-trip time of the Rydberg electron around the nucleus. The change in momentum ($\Delta \mathbf{p}$) is given by (atomic units are used, unless stated otherwise) $\Delta \mathbf{p} = -\int_{-\infty}^{\infty} \mathbf{F}(t)\, dt$, where $\mathbf{F}(t)$ is the (time varying) electric field. The change in momentum corresponds to an energy change of $\Delta E = \frac{1}{2}[2\mathbf{p}_0 \cdot \Delta \mathbf{p} + (\Delta \mathbf{p})^2]$ (where $\mathbf{p}_0$ is the initial momentum of the electron). Unless ΔE is greater than the binding energy, the ionization is strongly suppressed. At low THz amplitudes the ionization stems from electrons near the core and the ionization probability scales as $F \backsim \frac{1}{n^2}$, while large probabilities scale as $F \backsim \frac{1}{n}$ [1]. It has experimentally been demonstrated that an ionization probability of 10% of an ensemble of Rydberg atoms scales as $F_{10\%} \backsim \frac{1}{n^2}$, while a 50% ionization probability scales as $F_{50\%} \backsim \frac{1}{n^{3/2}}$ [1,2].

A HCP also appeared to be a good tool to design and manipulate Rydberg wave packets [3] and give a time dependent view of a wave packet [4]. Also a HCP can be used to study weakly bound Rydberg electrons [5]. Moreover, such HCP's have been used to recombine free electrons and ions [6]. The main difference with "conventional" laser illumination experiments, where photoionization or excitation can occur only when the electron density is peaked near the core, is that a HCP can excite or ionize the Rydberg electron throughout its orbit around the core.

For a freely propagating electromagnetic wave the term half-cycle pulse is not fully correct since Maxwell's equations demand that the time integral of the electric field in time is zero. In this case the "HCP" can best be described by an asymmetric mono-cycle (AMC) consisting of a very short half-cycle (duration: ~0.5 ps), followed by a much slower half-cycle of opposite polarity and a much lower amplitude (typical pulse amplitude asymmetry: 13:1 [7,8]). The duration of the AMC is then as long as ~70 ps (Fig. 1a). For Rydberg states with an orbiting time much shorter than the duration of this second unipolar pulse but

longer than the duration of the first unipolar pulse, ionization occurs during the first half-cycle of the AMC. The ionization can then be described by true half-cycle pulse ionization. Also, when the redistribution of Rydberg states by HCP's is studied in this regime, the second unipolar feature acts as a dc offset for the Rydberg electron, hardly affecting the redistribution process [9].

Ionization by true half-cycle pulses on a capacitor for $n \geq 300$ has been studied by Reinhold *et al.* [5], with the aid of fast voltage pulse generators (pulse duration: ~2 ns). Here we study ionization of high Rydberg states ($n \gg 80$) by electromagnetic HCP's. The orbiting time of a Rydberg state scales as $\tau_n \backsim n^3$ ($\tau_n = 77$ ps, for $n = 80$), and thus for Rydberg states with sufficiently large n, this time becomes much longer than the duration of the AMC. Beyond this n, the duration of the AMC is short compared to the orbiting period of the Rydberg electron and the change in momentum is $\Delta \mathbf{p} = \int_{-\infty}^{\infty} \mathbf{F}(t)\, dt \approx 0$, resulting in very little energy change of the Rydberg electron.
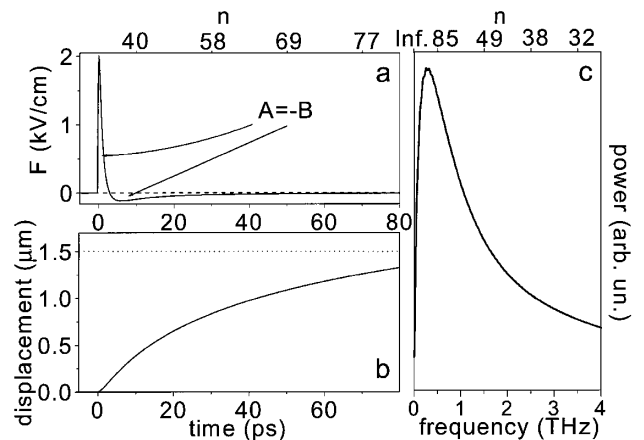

FIG. 1.   (a) A THz "half-cycle" pulse shape. Top axis: time is converted into n: $n = \left(\frac{t}{2\pi}\right)^{1/3}$. (b) A free electron is displaced by ≈ 1.5 μm by the "HCP" of (a) (amplitude: 2 kV/cm). (c) Fourier transform of the pulse shape depicted in (a). Top axis: frequency is converted into n: $h\nu = E = \frac{1}{2n^2}$.

Such an AMC does not leave the Rydberg state completely unaffected since the first unipolar feature kicks the electron, while the second delayed unipolar pulse stops the electron again. Therefore, a displacement of the electron is introduced. For a free electron this displacement is $\Delta x = \int_{-\infty}^{\infty} dt' \int_{-\infty}^{t'} \mathbf{F}(t) \; dt$. In Fig. 1b the displacement of an unaccelerated electron by an AMC with a peak amplitude of 2 kV/cm (as depicted in Fig. 1a) is plotted.

A Rydberg electron with an orbiting time much longer than the duration of the AMC will be significantly displaced by such a pulse since the change of velocity in an unperturbed orbit is negligible on the time scale of the duration of the AMC. A displacement of 1.5 μm of a free electron is calculated for the AMC of Fig. 1a, which means that a Rydberg electron can be displaced by as much as 1.5 μm.

In this Letter we experimentally investigate the effect of the second unipolar feature of "HCP's" in their interaction with Rydberg atoms, by studying ionization and bound state redistribution. The experiments were performed in a vacuum chamber (background pressure: 5.0×10^{-7} Torr). A thermal beam of gas-phase lithium atoms was directed in between two capacitor plates. A hole (outside diameter 10.0 mm) in the center of the anode plate was covered by a grid to minimize distortions on the electric field. This enabled the detection of electrons on a microsphere plate detector located 5.0 cm away from the anode plate. The cathode plate was connected to a pulse generator, which could ramp the voltage in a well defined time. The anode plate was connected to ground.

The unfocused beams (outside diameter 2 mm) of two narrow-band linearly polarized pulsed dye lasers were collinearly directed in between the parallel capacitor plates on to the lithium atoms. The first laser excited the lithium atoms from the ground state to the $2p$ level (670.97 nm), while the second laser (~700 nm, $\Delta\lambda \leq 0.01$ nm) was frequency doubled to excite lithium atoms from the $2p$ level to a well defined Rydberg state (mainly d character). The wavelength of the second exciting laser could be scanned in order to excite different Rydberg states.

The HCP's were produced 500 ns after the excitation by the second laser, by illuminating a biased GaAs wafer with a short (~150 fs) laser pulse (~800 nm), originated from a Ti:sapphire chirped pulse amplifier synchronized to the dye lasers. This radiation was directed collinearly with the optical laser beams, to the Rydberg atoms. The peak value of the electric field of the HCP's is proportional to the bias voltage over the wafer [1].

Ionization and state redistribution of the Rydberg atoms by the HCP's were monitored with selective field ionization (SFI) [10]. A THz ionization curve for a Rydberg state with $n = 70$ is depicted in Fig. 2a. The recorded "S curve" is a typical signature of THz ionization. For these n, mainly depletion of the initially prepared state occurs, with hardly any redistribution (a fraction of $<1\%$ of the initial population is redistributed to higher n). To validate that the
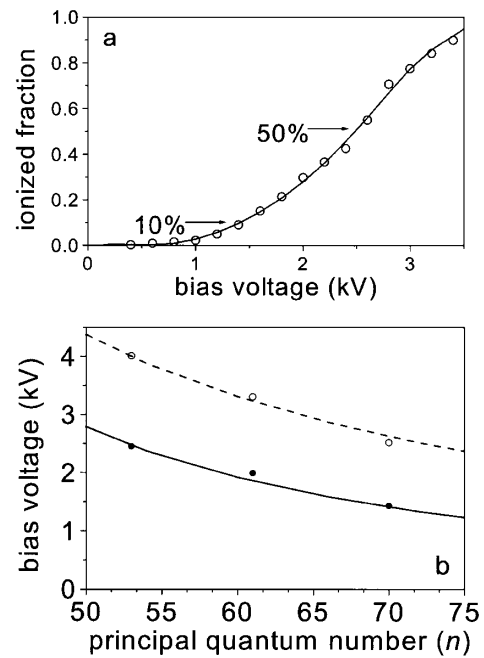

FIG. 2. Experimental THz ionization of Rydberg atoms for $\tau_n < \tau_{mc}$. (a) THz-ionization curve for a Rydberg state of $n = 70$ (solid line: 3 points smoothing of the experimental data). The two arrows denote the amplitude required to ionize 10% and 50% of the Rydberg atoms. (b) Experimentally determined ionization probabilities for Rydberg states of $n = 53$, 61, and 70. Open dots: 50% ionization probability; closed dots: 10% ionization probability. The data obey a $\frac{1}{n^2}$ (solid line) and $\frac{1}{n^{3/2}}$ (dotted line) scaling law.

ionization could be described by impulsive kicking of the Rydberg electron away from the nucleus, additional THz ionization curves were recorded for Rydberg states with $n < 70$. Figure 2b shows how the THz ionization probability of Rydberg atoms scales for the $n = 53$, 61, and 70 Rydberg states. Clearly the $F \sim \frac{1}{n^2}$ (10% ionization) and $F \sim \frac{1}{n^{3/2}}$ (50% ionization) scaling laws are retrieved.

For these Rydberg states, the round-trip time ($\tau_n = 2\pi n^3$) of the Rydberg electron ranges from 22 ps ($n = 53$) to 52 ps ($n = 70$), which is shorter than the duration of the AMC (τ_{mc}), but much longer than the duration of the first unipolar feature (τ_{hc}; see Fig. 1a), and therefore the ionization can be understood as a true half-cycle impulsive kick [1]. Now we focus on the regime where the duration of the second unipolar feature acts impulsively as well. This means that the duration of the AMC should be much shorter than the round-trip time of the Rydberg electron: $\tau_{mc} \ll \tau_n$. In this regime it is expected that the ionization mechanism of Rydberg states with these HCP's is altered by the second slower unipolar feature. This would correspond to Rydberg states of $n \gg 80$ [$\tau(n = 80) = 77$ ps], since the duration of the AMC is as long as ~70 ps (see Fig. 1a).

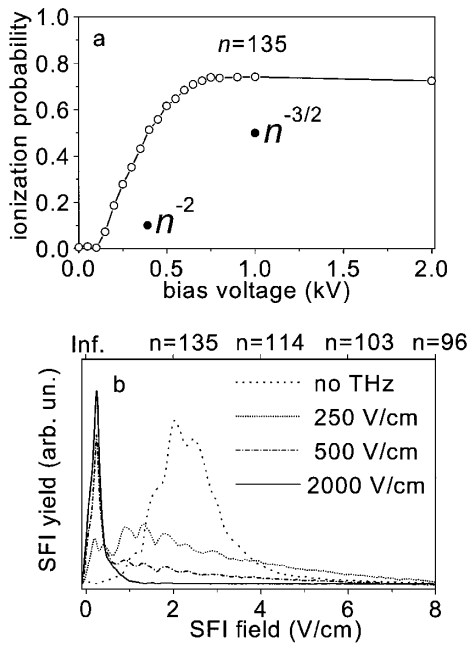In Fig. 3a the ionization probability is plotted versus the bias voltage over the wafer for a Rydberg state with

FIG. 3.   Experimental THz ionization of Rydberg atoms for $\tau_n \gg \tau_{mc}$. (a) THz ionization curve for a Rydberg state of $n = 135$. The two dots (•) denote what amplitude is required, predicted by the classical ionization laws, to obtain a 10% and 50% ionization probability. (b) Four SFI traces after exposing Rydberg atoms ($n = 135$) to a THz "half-cycle" pulse for different amplitudes.

$n = 135$ ($\tau_n = 371$ ps). Two striking results are apparent. First, it is not possible to achieve a 100% ionization probability (the maximum recorded ionization probability was 75%). Second, there is a strong deviation of the classical ionization scaling law, since for low ionization probabilities less peak amplitude is required for ionization than predicted by the classical scaling laws (denoted by the dots in Fig. 3a). A Rydberg state with $n > 60$ (binding energy: $E = -\frac{1}{2n^2} = -1.4 \times 10^{-4}$ a.u.) has already some probability to be ionized by a single, high-frequency photon of the HCP (1 THz $\cong 30\,cm^{-1} \cong 1.37 \times 10^{-4}$ a.u., see also Fig. 1c). Therefore, the observation that lower amplitudes are required of the HCP than predicted by the classical scaling laws can be explained by the fact that we are studying ionization in the one-photon ionization regime for these high Rydberg states ($n > 70$) as opposed to the multiphoton regime for Rydberg states with $n < 70$, for which the classical scaling laws were deduced. It does, however, not explain the suppression of the maximum ionization yield.

In Fig. 3b three SFI traces are depicted for $n = 135$ Rydberg atoms exposed to HCP's of different amplitudes (bias voltage: 250, 500, and 2000 V/cm). The SFI trace obtained with no THz shows the initial state distribution. Apparently, for these high n states, redistribution by the THz pulse to mainly higher n states occurs, instead of ionization. This significant redistribution is not observed

when we start out with Rydberg states with $n < 70$, indicating that the redistribution occurs due to the impulsive character of the second slower unipolar feature in HCP's. The shape of the SFI trace obtained by exposing Rydberg atoms to a HCP of 500 V/cm was not significantly altered when the amplitude was increased up to 2000 V/cm.

THz ionization curves for Rydberg states of different n ($n > 70$) showed that ionization suppression is stronger for Rydberg states with higher n. Moreover, a clear disagreement is observed between the experimentally determined ionization probability and the ionization probability predicted by the classical scaling laws, accounting for the first unipolar feature only.

Classical trajectory calculations were performed to qualitatively explain our observations. The ionization probability was calculated by determining the energy of a Rydberg electron after interaction with a HCP, for various starting times of the HCP during one Kepler orbit period. Calculations were performed for several Rydberg states ($l = 2$) with $n \leq 50$ by a HCP (pulse shape: Fig. 1a). Identical ionization curves were calculated when calculations were performed with a true HCP (using only the first fast half-cycle as the time varying field in the calculations). This shows that for Rydberg states with a sufficiently low n ($\tau_n \ll \tau_{mc}$), the second slow unipolar feature does not significantly contribute in the ionization process. From the calculated THz ionization the classical scaling laws were reproduced.

If the classical model is applied to a Rydberg state with $n = 135$ ($l = 2$), a different ionization behavior is obtained. Because of the second unipolar feature, ionization is strongly suppressed. This can be seen in Fig. 4a where the calculated ionization curve by the AMC (as depicted in Fig. 1a) is compared to the calculated ionization curve by a true HCP (FWHM 0.5 ps). For a free electron the energy exchange with an AMC would be zero, and thus it is understood that ionization is suppressed. The small residual ionization probability stems from the time dependent velocity of the electron in its orbit around the nucleus and thus $\Delta E = \int_{-\infty}^{\infty} \mathbf{v} \cdot \mathbf{F}(t)\, dt$ is not exactly zero.

In Fig. 4b the calculated redistribution of an $n = 135$ Rydberg state is depicted. An average redistribution to higher n is observed. The SFI spectrum was simulated (Fig. 4c) starting from the calculated state (re)distribution. This was done by converting the n of a Rydberg state into a field strength for which the specific Rydberg state would field ionize ($F = \alpha \frac{1}{n^4}$). These simulations reproduce the experimental observations accurately (compare Fig. 4c with Fig. 3b). From the experimental SFI traces it is deduced that with a bias voltage of 2000 V redistribution occurs to Rydberg states with $\langle n \rangle = 200$. The expectation value of the radial coordinate of the initial $n = 135$ Rydberg state is $\langle r \rangle = 2n^2 = 2.1\ \mu$m, and for a Rydberg state of $n = 200$ this expectation value is $\langle r \rangle = 4.2\ \mu$m. Therefore, a Rydberg state of $n = 135$ is displaced by a HCP with a bias voltage of 2000 V/cm by $\sim 2\ \mu$m.
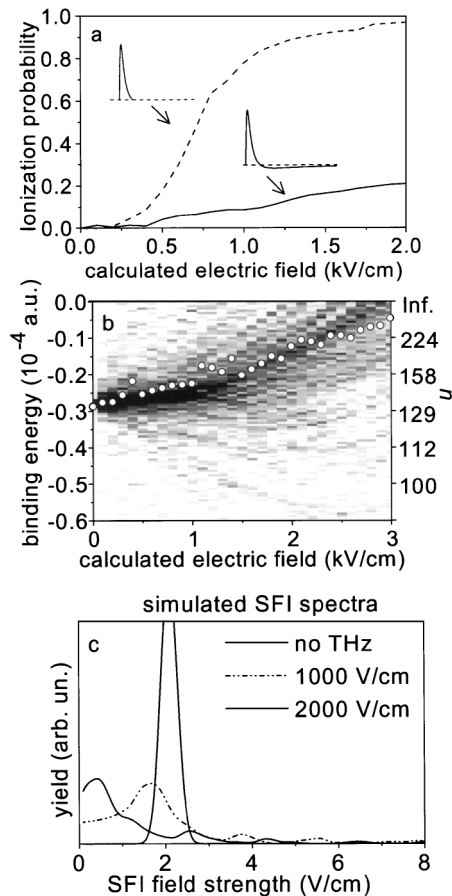
FIG. 4.   (a) Calculated THz ionization curves for a Rydberg state with $n = 135$ ($l = 2$). Dotted line: ionization by a true HCP. Solid line: ionization with a pulse shape as depicted in Fig. 1a. (b) Calculated redistribution of a $n = 135$, $l = 2$ Rydberg state as a function of the amplitude of the AMC. Open dots: average binding energy of redistributed states as a function of amplitude; intensity graph: the actual state distribution. (c) Simulated SFI spectra from the state (re)distribution obtained by the calculations.

In this Letter it is demonstrated that Rydberg atoms with high n ($n \gg 80$) are robust against ionization by HCP's. Instead of ionization, redistribution to high Rydberg states is observed. An intuitive mechanism for the ionization

suppression is presented in which the HCP can best be described by a fast start kick and a slow stop kick of opposite polarity and lower amplitude. Instead of kicking the Rydberg electron away from the nucleus, the HCP induces a new Rydberg orbit in which the Rydberg electron is displaced. This holds only in the regime where the time duration of the AMC is shorter than the round-trip time of the Rydberg electron. In this regime population transfer mainly to high Rydberg states is predicted by classical calculations in agreement with experimental observations. Note that the observed onset of ionization at much lower fields for higher n is not recovered by these classical trajectory calculations and remains an open question that deserves further theoretical attention.

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