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Ionization of oriented Rydberg states by subpicosecond half-cycle electromagnetic pulses

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The ionization of Rydberg states by subpicosecond, unipolar electromagnetic-field pulses shows a strong orientation dependence. A small static electric field permits us to study Rydberg-Stark states with oriented permanent electric dipole moments. The probability for ionizing a Stark state depends on its energy and the orientation of its electric dipole moment with respect to the direction of the pulsed electric field. This orientation dependence is most prominent for pulse durations comparable to the classical orbit period. Classical simulations reproduce many of the observed features.

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Recently developed sources of high-power, picosecond electromagnetic field pulses [1] have been used to study Rydberg atoms [2]. These pulses are nearly unipolar (i.e., "half" of an optical cycle), with peak powers up to 1 MW and coherent bandwidths of several THz, centered at 0.5 THz (~ 16.7 cm^{-1}) [1]. Because of the unique characteristics of this radiation, its interaction with atoms has interesting features that are not observed with conventional optical or microwave pulses. For example, Rydberg atoms ionized by half-cycle pulses (HCPs) display broad ionization thresholds and unconventional $1/n^{*2}$ threshold field scaling laws [2]. Classical analyses have been successful in describing the experimental observations [2,3].

HCP experiments introduce a new regime of strong-field, nonperturbative atom-radiation interactions. The time-dependent electric field in the HCP is also very similar to the electric field experienced by an atom undergoing a collision with a fast charged particle at a large, well-defined, impact parameter. Therefore, experimental [2] and theoretical [3,4] studies of the interactions of atoms with these pulses aid in the understanding of several different atomic physics problems.

This Rapid Communication describes a study of ionization of Na Rydberg states using HCPs in the presence of an additional, *static* electric field. The static electric field breaks the spherical symmetry of the atom, producing energy eigenstates that have permanent dipole moments, $d=-\langle z\rangle$, and are not eigenstates of angular momentum. These eigenstates experience a lowest-order energy shift

$$\Delta E = F\langle z\rangle \text{ a.u.} \tag{1}$$

due to the potential energy of the dipole in the field. The electric field F in Eq. (1) is assumed to point in the $+z$ direction. We will use this convention throughout the paper. States whose permanent dipole moments have different orientations with respect to the z axis may be selectively excited due to their different energies. The static electric fields used in the experiments are significantly smaller than those required for classical field ionization [5] and they induce energy shifts (i.e., Stark shifts) that are significantly smaller than the spacings between adjacent n states. Therefore, the major effect of the static field is to give the Rydberg states a particular orientation with respect to the z axis as well as provide small energy shifts that facilitate the excitation of states with a single orientation.

In the experiment, two tunable, 10-ns dye lasers excite a thermal beam of ground-state Na atoms through the $3P_{1/2}$ state to a Stark state with $|m_j|=\frac{1}{2}$. The dye lasers intersect the Na beam between two capacitor plates that provide the small static field in the interaction region. After a short delay (<300 ns) the atoms are irradiated by the HCP. Any ions that are produced by the HCP travel through a small hole (~ 1 mm diameter) in the negative capacitor plate to a charged-particle detector.

We use a biased, undoped, 0.5-mm-thick, $\langle 100\rangle$ GaAs wafer with a surface area of ~ 4 cm^2 to generate the HCPs [1,6]. The wafer is illuminated in vacuum with 100-fs pulses from a Ti:sapphire chirped-pulse amplifier operating near 780 nm. The freely propagating, radiated electric field pulse from the photoconductive switch has a dominant unipolar feature with a duration of ~ 450 fs [full width at half maximum (FWHM)] [1].

The HCP is collected with a paraboloidal mirror and directed toward the Rydberg beam ~ 10 cm away. Diffraction of the low-frequency components in the pulse occurs readily if the HCP is tightly focused or sent through a small aperture. This diffraction destroys the unipolar nature of the pulse, and great care is taken to avoid it. The diameter of the HCP beam is 0.5–1 cm at the interaction region. The oriented Rydberg atoms are subjected to HCPs whose electric field is in one of three possible directions—positive z, negative z, or perpendicular to z. The peak electric field in the HCP is varied by changing the bias voltage on the GaAs wafer. The amount of ionization is recorded as a function of the pulsed field amplitude.

Figure 1 shows the ionization probability for two $n=17$ Stark states as a function of HCP field amplitude for pulsed field directions in the $+z$ and $-z$ directions. The initial state in Fig. 1(a) exhibits the most *positive* dc Stark energy shift (i.e., the "uphill" state) and Fig. 1(b) displays data from an initial level with the most *negative* dc Stark shift (i.e., the

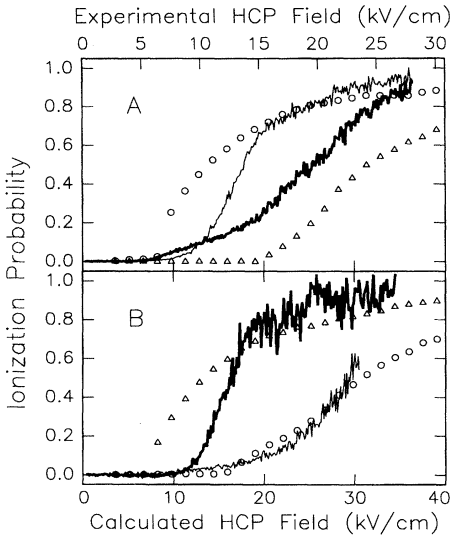

FIG. 1. Ionization probability as a function of HCP field for two different $n=17$ oriented states in a static field of 700 V/cm. (A) State with the most positive Stark shift (i.e., the "uphill" state); (B) state with the most negative Stark shift (i.e., the "downhill" state). Bold solid lines denote HCP field aligned with the static field and light solid lines indicate that the pulsed field opposes the static field. △ and ○ are the results of the classical calculation with the two fields aligned and opposed, respectively.

"downhill") state [7]. Recalling that the static field is assumed to be directed in the $+z$ direction, Fig. 1 shows that the "uphill" state is more easily ionized by a HCP whose direction opposes that of the static field, while "downhill" states are most easily ionized by fields that are aligned. Stated in a more consistent fashion, *oriented Rydberg states are more easily ionized by HCPs whose electric field is aligned with the permanent dipole moment of the atom.*

The difference between the ionization probability for an aligned state with positive and negative HCP field directions also depends on the projection of the permanent dipole moment on the HCP field axis. We define an asymmetry parameter β as the difference between the ionization probabilities for pulsed fields aligned with and opposed to the static field. In Fig. 2 the asymmetry parameter is plotted for five different states in the $n=17$ Stark manifold. The orientation and size of the permanent dipole moment for each state are shown pictorially by the arrows at the right of the figure. The asymmetry parameter is large and positive for the "downhill" state and large and negative for the uphill level. As expected, ionization of the state that has zero dipole moment (and therefore, no particular orientation about the xy plane) is independent of the sign of the pulsed field. In addition, if the HCP is polarized perpendicular to the static field, ionization of all states is found to be independent of the sign of the field pulse. This feature is also expected since the Stark states have no particular orientation in the xy plane.

The ionization thresholds are not significantly altered if the magnitude of the static field is changed. This observation indicates that the static field can be considered as an alignment tool that does not substantially affect the ionization dynamics in any other way. However, the characteristics of

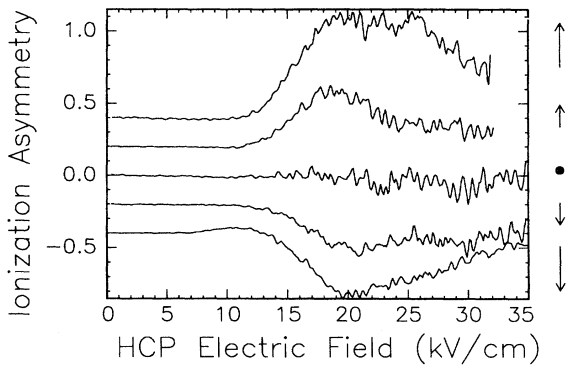

FIG. 2. Ionization asymmetry parameter β vs HCP field for the ionization of five different $n=17$ Stark states in a static field of 700 V/cm. The direction and approximate relative magnitude of the permanent dipole moment for each state are shown at the right. All of the curves have a value of zero at zero field amplitude and are offset along the y scale to prevent them from overlapping.

the ionization thresholds change significantly with increasing principal quantum number. The asymmetry between ionization with positive and negative HCPs is strongest for the lowest energy states ($n=14$) and is essentially zero for states with $n>25$. In Fig. 3, the ionization probability for several downhill states is plotted as a function of electric field for the two different HCP field directions. The decrease in the ionization asymmetry with increasing n is clear even over the small range of states shown.

The interaction of the HCP with the Rydberg electron can be described classically [2]. The Kepler period, $\tau_K=2\pi n^3$, for the motion of a classical electron is longer than (or comparable to) the duration of the HCP, τ_{HCP}, for all of the states

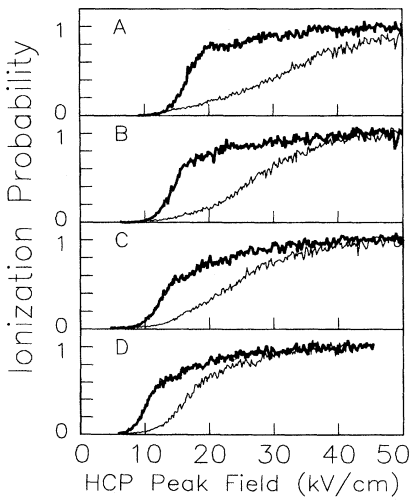

FIG. 3. Ionization probability for several different "downhill" states as a function of the peak HCP field for two different field directions. Bold solid lines indicate aligned static and HCP fields and light solid lines denote opposing fields for (A) $n=16$, $F_{\mathrm{dc}}=1000$ V/cm; (B) $n=17$, $F_{\mathrm{dc}}=800$ V/cm; (C) $n=18$, $F_{\mathrm{dc}}=640$ V/cm; (D) $n=19$, $F_{\mathrm{dc}}=580$ V/cm.

studied. Ionization occurs when the pulse transfers energy to the electron in excess of its binding energy. The electron receives an energy "kick" or impulse from the HCP which can be written as

$$\Delta E = -\int_{-\infty}^{\infty} \vec{F}(t)\cdot\vec{v}(t)dt \text{ a.u.}, \quad (2)$$

where $\vec{F}(t)$ is the HCP field and $\vec{v}(t)$ is the velocity of the electron. The initial position and velocity of the electron are critical in evaluating the integral in Eq. (2). This process is completely different from ionization in longer pulsed fields ($\tau_{\text{pulse}} > \tau_K$), where ionization is made possible due to a modification of the binding potential and not by energy exchange with the field [5].

Our observations can be qualitatively understood by inspection of Eq. (2) if we recast our primary observation in classical terms. *Oriented Rydberg states are more easily ionized by a HCP that gives the electron an energy kick away from the nucleus.* Consider an ensemble of classical electrons in a particular oriented orbit, the uphill state, as an example. The classical motion of an electron in this state is essentially one dimensional and occurs along the $+z$ axis [8]. Just prior to the pulse arrival, half of the electrons in the ensemble are moving away from the nucleus (i.e., in the $+z$ direction) and the rest are moving toward the nucleus. If the HCP field is in the $+z$ direction, electrons only gain energy if their velocity is directed toward the nucleus. For $n<20$, all electrons that gain energy by traveling toward the nucleus during the beginning of the HCP will pass the nucleus and reverse direction before the end of the pulse. Hence, according to Eq. (2), energy gain during the pulse is always partially frustrated, and the minimum field required for ionization is necessarily high. Conversely, if the HCP field is in the $-z$ direction, electrons that travel away from the nucleus during the pulse gain energy. The velocity of these electrons is not rapidly reversed by the nucleus, and they are allowed to gain energy over the entire pulse. Therefore, large energy transfer is possible at lower peak fields. Using these arguments, we expect that uphill states are more easily ionized by HCP fields polarized in the $-z$ direction in good agreement with our observations. The argument holds for reversed directions of the HCP field for the downhill states. Of course for nonoriented states with symmetric charge distributions, electrons on one side of the atom will be pushed away from the nucleus while those on the opposite side will be pushed toward the nucleus for either HCP field direction, and no asymmetry is expected.

This illustration also explains the lack of asymmetry for levels where $\tau_K \gg \tau_{\text{HCP}}$. Since the probability for finding the Rydberg electron near the nucleus is proportional to $1/\tau_K$, the probability that the electron will scatter from the nucleus during the pulse is just $\tau_{\text{HCP}}/\tau_K = \tau_{\text{HCP}}/2\pi n^3$. Therefore, the probability that an electron will reverse its velocity via core scattering during the pulse drops sharply with increasing n. In the absence of core scattering, the electrons are able to gain energy over the entire pulse for either HCP field direction, and no ionization asymmetry is observed [9].

An orientation dependence is also seen in *hydrogenic* atoms, even if $\tau_K \ll \tau_{\text{HCP}}$ [10]. In that case, the integrand in Eq. (2) undergoes rapid oscillations as the electron moves between its inner and outer turning points during the relatively long HCP. The Rydberg electron can only gain energy from the field if it passes over (or tunnels through) the potential "saddle point" created by the HCP. After moving past the potential barrier, the electron's motion is no longer oscillatory, and therefore it can gain energy from the pulsed field and ionize. Of course, only those atoms whose permanent dipole moment is parallel to the HCP field direction can sample the saddle point. Therefore, an ionization asymmetry is observed with the same orientation dependence as that caused by the ion core scattering discussed above. The transient modification of the electronic potential is not important in the short pulse limit, $\tau_{\text{HCP}} \ll \tau_K$, because the electron has insufficient time to sample the saddle point during the pulse.

The qualitative arguments used above can be made quantitative using a classical numerical simulation [2]. An ensemble of points on classical Stark trajectories [11] describes the initial position of the electron at the beginning of the HCP, which is modeled as an inverted parabolic (or Gaussian) pulse of 450 fs duration (FWHM). The equation of motion is then numerically integrated and the energy and position of the electron are recorded at the end of the HCP. The electron is considered to be ionized (i) if its final energy is above the static potential barrier or (ii) if it is located beyond the static saddle point. The ionization probability is calculated by counting the number of ionized electrons weighted by the probabilities of finding these electrons at their respective initial positions.

The results of our calculation for the "uphill" and "downhill" states are plotted with the experimental data in Fig. 1. They show that the oriented atoms are more easily ionized when the HCP field is aligned with the atomic dipole moment. There are no adjustable parameters in the numerical simulation. The results of the simulation are qualitatively the same if the initial-state trajectories are assumed to be oriented low angular momentum orbits without any static field present, justifying our qualitative description of these Stark states as oriented Rydberg states.

Figure 1 clearly shows that the asymmetry predicted by the classical simulation is somewhat larger than that observed in the experiment. The simulation assumes hydrogenic Stark states that have a well-defined permanent dipole moment. However, in Na, the non-Coulombic potential of the electron-ion core causes the dipole moments of the various Stark states to precess. Although the time average dipole moment is very similar to that in hydrogen, the instantaneous dipole moment of a Na Stark state can be in the opposite direction of the time-averaged moment. Therefore, the precession should at least partially smear out the differences between ionization with opposite polarity pulses.

In addition, due to the strong diffraction of the near-zero-frequency components in the pulse as it propagates, the temporal shape of the HCP may not be purely unipolar in the interaction region. We have observed vastly different ionization behavior if the HCP is either tightly focused or apertured, presumably due to pulse shape effects. In addition, a reflection pulse of $\sim 25\%$ field amplitude appears approximately 11 ps after the main pulse, due to imperfect transmission of the HCP through the GaAs wafer. This reflection pulse has the opposite polarity of the main pulse, causing the entire wave form to differ from perfect unipolarity. The discrepancy of a factor of 1.3 between the experimental and

theoretical HCP field values in Fig. 1 is essentially removed if these small non-half-cycle components of the pulse are attenuated [12].

Close inspection of Figs. 1(a) and 2 reveals another effect that is observed predominantly in the ionization spectra for the uphill states. For low ionization probabilities, these states consistently show that ionization occurs more easily if the electric field opposes the dipole moment of the atom. For higher ionization fractions, the ionization asymmetry reverses and agrees qualitatively with the classical predictions. The peak electric field at which this reversal occurs is definitely pulse shape dependent. However, the effect is not completely removed if the negative components of the HCP are significantly attenuated [12]. Furthermore, the fact that the asymmetry reversal is only observed for the blue states suggests that it is a nonhydrogenic effect.

In conclusion, we have explored the ionization of oriented Rydberg states using high-field nearly unipolar subpicosecond pulses. The ionization process is extremely sensitive to the relative orientation of the permanent dipole moment of the atom and the direction of the pulsed electric field. A straightforward classical model qualitatively describes the major features in the data. More sophisticated calculations, including effects such as the non-Coulombic ion core potential, may be needed to reproduce the finer details of the experimental results.

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[7] Due to the large quantum defect of the Na s states ($\delta_s = 0.35$ modulo 1) these states have essentially zero dc Stark shift in the static fields used in the experiment. Therefore, the "downhill" state is taken to be the lowest energy state corresponding to a given n value not including the s state.

[8] The motion is not truly one dimensional, but this approximation is not unreasonable for a qualitative description of the ionization process. The "downhill" state orbits in a similar fashion, but along the $-z$ axis. States with intermediate Stark shifts have more complicated classical orbits that contain high angular momentum components and cannot be described in one dimension.

[9] If we neglect scattering with the nucleus, we are assuming that the electrons in these high n states are essentially free. Obviously, the energy gained by a free electron is independent of the sign of the HCP.

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