

Creating and Probing Electronic Wave Packets Using Half-Cycle Pulses

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Ultrashort, nearly unipolar electric field pulses are used to produce novel dynamic states in Rydberg atoms. A single 500 fsec field pulse redistributes essentially all of the population from a single stationary Rydberg state to hundreds of different final states. The dynamic evolution of the nonstationary wave packet is monitored using a second, time-delayed pulse. The time-dependent momentum-space probability distribution of the wave packet can be retrieved experimentally using a classical impulse approximation. [S0031-9007(96)00264-5]

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Recently, a significant amount of research interest has been aimed at studying the evolution of atomic and molecular systems during and subsequent to irradiation by non-perturbative electromagnetic pulses [1]. These studies are a necessary prerequisite to the creation of electronic and nuclear wave packets with arbitrary characteristics which are the key to coherent control of atoms and molecules [2]. This Letter describes an experiment which demonstrates that brief, unipolar “half-cycle” pulses (HCPs) can be used to create novel dynamic states in atoms and that HCPs with sufficiently short duration may be implemented to retrieve the momentum-space probability distribution of essentially any electronic wave packet.

Highly excited Rydberg eigenstates in alkali atoms have been exposed to subpicosecond HCPs [3–6]. The coherent bandwidth of these pulses is so large that many states of different angular momentum and principle quantum numbers are coupled in a single pulse [3,7–9]. Experimental and theoretical investigations have shown that even very small HCP fields are capable of completely depleting the population in the initial state [6,9]. The application of still higher fields initiates a diffusion of population through states of higher and lower energy and essentially all available angular momenta [6,9,10]. At extremely high fields, significantly larger than those required for “over the barrier” ionization, the diffusion proceeds into the continuum until the atom is ionized with 100% efficiency [3,10,11]. Considering the sheer number of intermediate states and enormous relative field strengths, the ionization of Rydberg atoms by HCPs is one of the most extreme examples of nonperturbative atom-field interactions. The goal of the experiment described in this Letter is to determine the temporal evolution of the complicated wave packet which is produced when a HCP interacts with a Rydberg atom.

An accurate measurement of the final state distribution of these wave packets is not only extremely difficult [6], it is also insufficient for studying their dynamic behavior. The complex amplitude of each eigenstate in the superposition must be known if the time-dependent probability distribution is to be determined. Furthermore, the precise

amplitude and phase in each of the vast number of constituent stationary states is extremely sensitive to small variations in the temporal structure of the HCP, making comparison with theory virtually impossible. However, the general shape and evolution of the electronic probability distribution can be less sensitive to experimental noise and uncertainties, making a direct measurement of the probability distribution more appealing and robust than a precise measurement of the eigenstate amplitudes.

It has been noted that HCPs might be used to study dynamic evolution in wave packets [12]. Since the probability for optical photon absorption and emission in Rydberg atoms is restricted to a small spatial volume near the nucleus, all previous studies of wave-packet evolution in atoms have only been sensitive to the small radius part of the electronic probability distribution [13]. However, the interaction of a bound electron with a HCP is significant over the entire volume of the Rydberg wave function. The results presented here show that a HCP, whose duration is much less than the evolutionary time scale of the wave packet, can be used in conjunction with a classical impulse approximation to retrieve the momentum probability distribution of the wave packet as a function of time.

Consider a classical electron with momentum \vec{p}_0 . If the duration of the HCP is negligible compared to the time scale for variations in the electron’s position and momentum, then the momentum of the electron after its interaction with a HCP is $\vec{p} = \vec{p}_0 + \vec{A}$ where \vec{A} is defined as

$$\vec{A} = - \int F_{\text{HCP}}(t) dt \quad (1)$$

and $F_{\text{HCP}}(t)$ is the electric field in the pulse. Atomic units are used throughout. The pulse induces a change in the electron’s total energy of

$$\Delta E = (\vec{p}^2 - \vec{p}_0^2)/2 = \vec{p}_0 \cdot \vec{A} + \vec{A}^2/2, \quad (2)$$

so that the energy gained (or lost) by the electron depends only on its initial momentum and the time integrated field.

The energy exchange required to ionize a Rydberg electron is simply its binding energy, $1/2n^2$. Without

loss of generality, assume that \vec{A} lies in the $+z$ direction. Then from Eq. (2) it is clear that for each value of A , there exists a single value of $\vec{p}_0 \cdot \hat{z} = p_{0z}$ for which the atom ionizes at threshold

$$p_{0z} = (n^{-2} - A^2)/2A. \quad (3)$$

Hence, the probability that the Rydberg electron has a momentum component p_{0z} is equal to the probability that a HCP with a time integrated field $-A$ will ionize the atom at threshold. So by measuring the ionization probability as a function of field, the momentum distribution of the initial state along the field axis can be obtained using Eqs. (2) and (3).

Figure 1 shows an experimental determination of the probability distribution for p_{0z} for the Na $25d$ state. The distribution is obtained from the ionization vs peak field curve shown in the inset. The dashed curve shown with the experimental distribution is a theoretical momentum distribution for the $25d$ state in H. The theoretical distribution is obtained by taking the average z momentum of a classical electron during a time window, and weighting that momentum by the probability for finding the electron at each point in its orbit. The duration of the time window is equal to the 0.5 psec duration of the HCP. The windowed distribution is nearly identical to the true distribution for small momenta, but it drops off much more rapidly for large values. The window average is insensitive to the short time that the classical electron spends near the nucleus where its momentum is large. Both the theoretical and experimental distributions are normalized

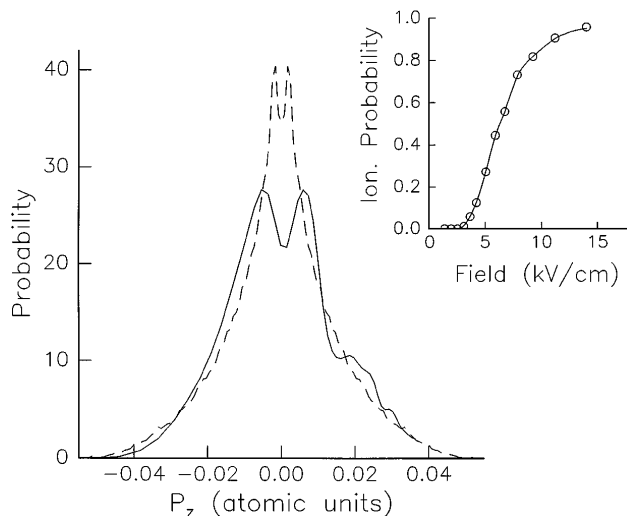


FIG. 1. Experimental probability distribution for p_{0z} for the Na $25d$ state obtained using the impulsive momentum retrieval (IMR) technique (solid curve). Shown with the data is the theoretical momentum distribution discussed in the text (dashed curve). Inset: The ionization probability vs HCP field data used to generate the experimental probability distribution. The continuous experimental probability distribution is obtained using the spline fit (inset solid line) to the data (O).

per unit momentum so that the integrated probability is equal to unity.

The model does a reasonable job of reproducing the general form of the expected distribution in Fig. 1. The impulse approximation rapidly breaks down for lower n values but becomes even more accurate for higher n states due to the variation of τ_K [8,10]. At the present time, the utility of a more formal treatment is questionable due to small uncertainties in the precise temporal shape of the pulse [5,7]. However, the respectable agreement between the impulse model and experiment in Fig. 1 suggests that it may be possible to retrieve information on the dynamic evolution of wave packets using this simple approach. Since wave packets have no well defined energy, the value of n which is used in Eq. (3) must be an average value for the distribution. In practice, for bound wave packets, the value of n is adjusted so that the time-averaged momentum of the distribution is zero as is required for a bound state.

The impulsive momentum retrieval (IMR) technique discussed above has been used to study the evolution of the wave packet created by a strong, 0.5 psec HCP. In the experiment, ground state Na atoms in a thermal beam are excited to the $25d$ Rydberg state through an intermediate $3p_{1/2}$ level using two, tunable nanosecond dye lasers [3–6]. The Rydberg atoms are then irradiated by two time-delayed HCPs. The HCPs are produced by illuminating two, separate biased GaAs photoconductive switches with 150 fsec, 780 nm pulses from an amplified Ti:sapphire oscillator [7]. The first pulse has insufficient amplitude to ionize the atoms, but instead redistributes the Rydberg population, creating a complicated, dynamically evolving wave packet. The total ionization yield produced by the second pulse is monitored as a function of its peak field and delay relative to the first pulse. The HCPs counterpropagate through the laser-atom interaction region at right angles to the atomic and laser beams. Approximately 50 nsec after the HCPs interact with the atoms, a small voltage ~ 50 V is applied to the lower of two capacitor plates which straddle the interaction volume. Any ionized atoms are pushed through a $50 \mu\text{m}$ slit in the upper plate toward a microchannel plate detector. The narrow extraction slit makes it possible to achieve a temporal delay resolution of < 200 fsec in the counterpropagating pulse geometry [6].

Figure 2 shows the probability for ionizing the wave packet produced by the first HCP as a function of relative delay and field in the second, analyzing pulse. The $25d$ initial state and ~ 2.5 kV/cm peak amplitude of the first pulse are identical in all the scans. The modulations in the ionization signal as a function of time are due to temporal variations in the wave-packet momentum distribution. The peak field calibration is obtained by ionization of the $25d$ state directly with a single pulse [3,5,11]. Because the HCPs contain small nonunipolar features up to 15 psec after the main pulse [5,7], a

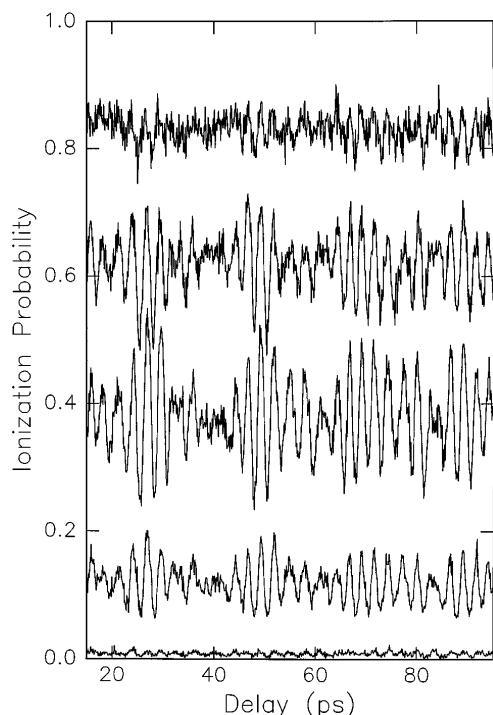


FIG. 2. Probability for ionizing a wave packet as a function of time for several analyzing field amplitudes. The wave packet is created by exposing the $25d$ state to a strong HCP whose amplitude is only slightly below threshold for ionization (approximately 2.5 kV/cm). From bottom to top, the peak analyzing HCP field amplitudes are 2.0, 3.7, 5.2, 6.9, and 9.5 kV/cm, respectively.

minimum delay of 15 psec is shown in the scans. Hence, none of the observed features are due to temporal overlap between the two fields.

Using the ionization probability vs delay scans for 13 different analyzing pulse amplitudes, the ionization probability as a function of peak field can be extracted for any time delay. With the aid of Eq. (3), this ionization probability vs peak field data can be used to retrieve the time-dependent momentum distribution of the wave packet. The results are shown in Fig. 3, where clear oscillations in the momentum distribution can be seen as the wave packet evolves in time. The oscillations in p_{0z} can be understood qualitatively by considering the initial eigenstate of the atom as a stationary charge distribution which is symmetric about the x - y plane. The HCP produces an impulsive force on the charge distribution, giving it a kick along the z axis. The kinetic energy gained by the distribution is converted into potential energy as it pulls away from the nucleus in analogy to a mass on a spring. The distribution then accelerates in the direction opposite to the impulse, overshoots its initial equilibrium position, and executes quasiperiodic motion. Because of the anharmonicity of the Coulomb potential, the wave packet alternately disperses and revives as it oscillates [13]. Since the IMR technique measures

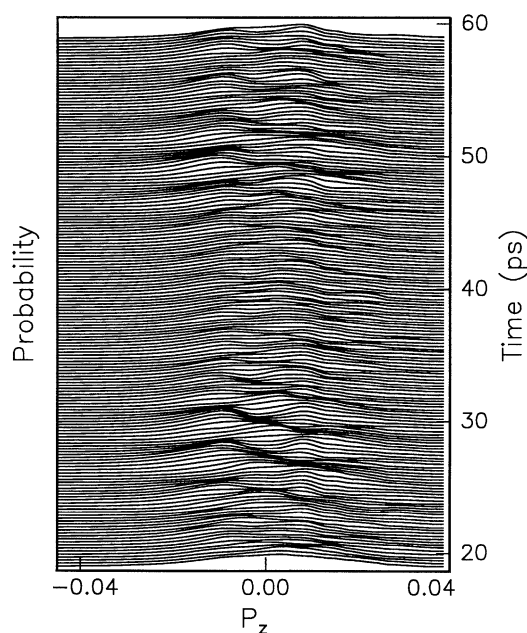


FIG. 3. Temporal evolution of the momentum distribution for the same wave packet as in Fig. 2. Oscillations in the distribution are clearly visible. The time interval between adjacent traces is ~ 230 fsec. Each curve is obtained using a smoothed spline fit to ionization vs field data similar to that shown in Fig. 1.

the momentum-space probability distribution, not the momentum-space wave function, no direct information on the spatial characteristics of the wave packet can be obtained. Fortunately, the response of the wave packet to subsequent interactions can be formulated in momentum space as well as in configuration space.

Although the huge number of stationary states involved makes plotting the quantum mechanical distribution very difficult, it is straightforward to calculate the time-dependent expectation value $\langle p_z \rangle$. To this end, the time-dependent Schrödinger equation is integrated using approximately 1000 basis states, including all n and l values from $n = 17$ to 47 [6]. The calculation assumes that the HCP which creates the wave packet has a Gaussian temporal profile with a 0.5 psec full width at half maximum (FWHM) and a peak amplitude of 2.5 kV/cm. The radial matrix elements for Na are calculated using a standard Numerov integration algorithm [14]. The results of the simulation are shown in Fig. 4 along with the average experimental values of p_z obtained from curves similar to those shown in Fig. 3. The agreement between theory and experiment is quite respectable, especially when one considers that the calculation predicts that hundreds of states are actually excited by the first pulse, and that the largest population in any of the states is less than 8%. Note that experimental results are shown for a second pulse polarized parallel (measuring p_z) or antiparallel (measuring $-p_z$) to the initial pulse. The features in the two data sets are 180° out of phase as expected.

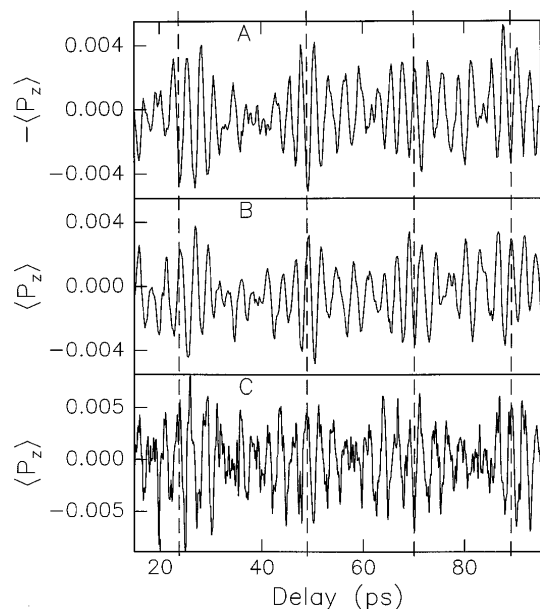


FIG. 4. Average momentum $\langle p_{0z} \rangle$ for the wave packet. (A),(B) The distributions obtained when the field of the analyzing pulse is directed in the $-z$ and $+z$ directions, measuring $\langle -p_{0z} \rangle$ and $\langle p_{0z} \rangle$, respectively. (C) The time-dependent expectation value $\langle p_{0z} \rangle$ obtained from a quantum calculation. The dashed lines demonstrate the good overall agreement in the positions of the oscillations.

In summary, this work demonstrates a new IMR technique which can be used to directly measure the momentum distribution of extremely complicated electronic wave packets created by highly nonperturbative fields. Measurement of the full three-dimensional momentum distribution should be possible by alternately rotating the polarization of the analyzing pulse along three orthogonal axes. The method is applicable to any wave packet whose temporal evolution is slow compared to that of the HCP so that the impulse approximation is valid. The IMR method was used to study the evolution of novel wave packets produced by exposing Rydberg atoms to strong, subpicosecond HCPs. The experimental results are in good agreement with quantum calculations in spite of uncertainties in the precise shape of the pulse [5,7]. This observation is further evidence that the specific probability distribution of wave packets created by nonperturbative fields may be far less sensitive to experimental noise and

uncertainties than the individual amplitudes and phases of the constituent stationary states.

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