Quantum-state measurement of ionic Rydberg wavepackets

X Zhang and R R Jones
Department of Physics, University of Virginia, Charlottesville, VA 22904-4714, USA
E-mail: bjones@virginia.edu

New Journal of Physics 11 (2009) 105050 (11pp)
Received 14 May 2009
Published 30 October 2009
Online at http://www.njp.org/
doi:10.1088/1367-2630/11/10/105050

Abstract. Nearly unipolar, sub-picosecond half-cycle pulses (HCPs) have been used to measure, via impulsive ionization, the aspects of the electronic quantum state relevant to the radial motion of Ba\(^+\) \(n\)g Rydberg wavepackets. Following laser excitation of the ionic wavepacket, the double ionization probability is measured as a function of the amplitude and delay of the HCP probe. Fourier transform of the time-dependent ionization signal facilitates the determination of the complex amplitudes of the constituent \(n\)g eigenstates in the wavepacket, enabling a full characterization of its time-dependent radial evolution. This is the first experimental demonstration of wavepacket recovery in ions, a capability that may prove to be invaluable for future measurements of quantum dynamics in multielectron systems.

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1 Author to whom any correspondence should be addressed.
1. Introduction

The creation and characterization of electron wavepackets with specific dynamical properties are a key means for controlling quantum interactions within and between atoms and molecules ([1, 2] and references therein). Ultrashort and intense optical pulses are the principal tools that enable such efforts. The production of ultrashort optical pulses with durations < 100 asec is now possible [3]. However, continued technological advances in this area will be critical for the generation of even shorter pulses to provide access to electron dynamics at the fastest timescales (\(\sim 1\) asec) which are relevant for chemical bonding and valence electron interactions. Despite the availability of ultrashort pulses, the development of experimental protocols for manipulating and viewing electron dynamics is an important prerequisite for achieving the desired level of control ([4]–[11]; [12] and references therein; [13]–[18]). This has become more important and difficult with the increase in the number of degrees of freedom available in few- and many-body systems.

Rydberg atoms provide a convenient alternative to ground-state atoms for developing and testing new methods for controlling and visualizing intra-atomic electron dynamics. The characteristic timescales within Rydberg atoms are amplified by orders of magnitude relative to those in the ground state, substantially relaxing the experimental pulse duration requirements. As a result, mature techniques for exquisite manipulation and characterization of femtosecond and picosecond pulses make it possible to implement sophisticated excitation and detection schemes using temporally shaped pulses and pulse sequences [1, 2, 12, 14], [19]–[24]. While methods for creation and observation of Rydberg wavepackets in one-electron atoms [1, 2, 4, 12] (or in two-electron atoms with only one Rydberg electron [25]–[40]) are well established, the extension of these techniques to studies of two or more Rydberg electrons has been difficult. Recently, we used a time-domain variant of isolated-core excitation to prepare double Rydberg wavepackets in which two electrons within the same atom are radially localized [41, 42]. Information about the time-dependent evolution of these atoms was inferred from time-independent measurements of the Rydberg ion population following autoionization. Future experiments will attempt to use time-domain probes to follow the dynamics directly. An important question that has to be addressed is what information about the state of the system at a specific instant in time can be extracted, since the electrons can continue to interact following the probe. Assuming these experiments proceed via a time-resolved isolated-core approach [27, 33], i.e. sequential excitation of Rydberg wavepackets on the neutral atom and singly charged ion, accurate characterization of the individual wavepackets will be important for interpreting the final results. Moreover, following autoionization, measurement of the final ionic wavepacket will likely be required to obtain complete information about the two-electron dynamics. To facilitate those experiments, we have demonstrated, and describe below, the first complete measurement of radial Rydberg wavepacket dynamics in an atomic ion.

2. Experimental procedure

In the experiment, Rydberg ion radial wavepackets are produced by exposing Ba atoms to a sequence of laser pulses, which first singly ionize and then coherently excite the atoms to ionic Rydberg states. The excitation scheme is nearly identical to the one used previously to produce double Rydberg wavepackets [42] except that the first electron is promoted to the continuum rather than bound Rydberg states, with an energy just above rather than slightly below the lowest
Ba$^+$ ionization threshold. After a variable time delay, the coherently evolving Rydberg ions are exposed to a subpicosecond nearly unipolar, ‘half-cycle’, electric-field pulse (HCP) \[43]. The Ba$^{2+}$ yield is recorded as a function of the delay and field amplitude of the HCP probe. The delay-dependent double ionization signal reveals the initial wavepacket motion, its collapse due to dispersion and its subsequent revival \[1]. The complex amplitudes of the constituent Rydberg states in the ionic wavepacket are extracted from a Fourier transform analysis of the delay-dependent yield \[16].

The laser excitation method is similar to that described previously and is shown schematically in figure 1 \[41, 42, 44\]. Ground-state atoms in a thermal beam are exposed to a series of nanosecond and picosecond laser pulses that excite Ba$^+$ \(ng\), \(n \approx 38\) radial wavepackets via a sequential, three-step, four-photon process. First, a 350 nm pulse stimulates two-photon resonance-enhanced ionization, through the intermediate 5d6p\(^1P_1\) level, into the 6s$^+$ and 5d$^+_j$ continua. The 5 ns, 350 nm, \(\sim 50\ \mu J\) laser pulse is generated by frequency mixing, in a potassium dihydrogen phosphate (KDP) crystal, the output of a 522 nm pulsed dye laser and the 1064 nm fundamental of the Nd:YAG (YAG denotes Yttrium aluminum garnet) pump. A 300 fs, 233 nm, \(\sim 3\ \mu J\) pulse then excites the 5d$^+_j/2$ ions to the 4f$^{7/2}$ level. This second laser pulse is generated by frequency mixing the 200 fs, 582 nm output of an optical parametric amplifier (OPA) with the 150 fs, 390 nm OPA pump. The 390 nm pump is produced by frequency doubling, in a 0.1 cm thick beta barium borate (BBO) crystal, a portion of the 100 fs, 780 nm output from an amplified Ti:sapphire laser. Lastly, a 500 fs, 313 nm laser pulse launches the wavepacket by exciting a coherent superposition of Ba$^+$ \(ng\) Rydberg states \((n \sim 38)\) from the Ba$^+$ 4f\(^{7/2}\) state. The launch pulse is generated by frequency mixing, in a 0.1 cm thick BBO crystal, the 780 nm Ti:sapphire fundamental with the 200 fs, \(\sim 524\ \mu m\) output from a second OPA. The lasers fire at a 15 Hz repetition rate and the excitation pulses are linearly polarized in the vertical \((z)\) direction. A 350 mm fused silica lens focuses the lasers into the Ba beam in a vacuum chamber with a background pressure of \(\sim 10^{-6}\) Torr.

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**Figure 1.** Schematic laser excitation diagram for creating ionic \(ng^+\) Rydberg wave packets.
Our interest is in the characterization of the radial motion of the \( \text{Ba}^+ \) wavepacket. The radial evolution is determined by the complex amplitudes of the constituent \( n\ell \) eigenstates. The frequency spectrum of the launch laser pulse coupled with the \( n^{-3} \) scaling of the photoabsorption cross-section determines the \( n \)-state distribution. In principle, dipole selection rules allow Rydberg excitation to \( \ell = 2 \) or \( \ell = 4 \) orbital angular momentum states. However, the radial matrix elements between the 4f launch state and \( ng \) final states are substantially greater than those between the 4f and \( nd \) levels. Indeed, \( 4f \rightarrow n\ell \) photoexcitation measurements performed using tunable dye lasers show strong excitation of the \( ng \) states and no observable \( nd \) population [45]. Accordingly, the wavepackets created here are composed entirely of \( ng \) Rydberg states.

Other quantum numbers (e.g. \( J \) and \( M \), which correspond to total electronic angular momentum and its projection on the laser polarization axis, respectively) contribute to the full definition of the state of the electron, but have a negligible effect on the radial motion. The launch pulse excites both fine-structure components, \( J = 7/2, 9/2 \), of each \( ng \) state and the spin–orbit interaction results in angular beating with periods in the microsecond regime, orders of magnitude longer than the Kepler period for the radial motion. Moreover, Rydberg electrons with different \( M \) values are created and detected. Excitation of the 4f/7/2 state proceeds through the 5d5/2\( \ell = 0 \), \( M = 0 \) continuum, which is the only \( \text{Ba}^+ \) 5d5/2 channel that is significantly populated from the 5d6p\(^1\)P\( _1 \) intermediate level [46]. Projection of the two-electron continuum onto the single-electron states of the \( \text{Ba}^+ \) ion creates an incoherent distribution of all possible \( M \)-states (\( \pm 1/2, \pm 3/2, \pm 5/2 \)) in the 5d5/2 ion. This distribution of \( M \) levels is preserved by subsequent laser excitations. However, the radial motion of the \( ng \) Rydberg wavepackets is identical for each \( M \)-state in the Rydberg population. In the remainder of the paper, we explicitly consider only those aspects of the quantum state that are relevant to radial dynamics.

Following the launch pulse, the wavepacket evolves in the time-independent ionic potential. Given the frequency spectrum of the launch laser, we expect that the wavepacket consists of approximately eight \( ng \) Rydberg states centered near the principal quantum number, \( n_0 \simeq 38 \). Based on previous studies of radial wavepackets in neutral atoms, we would predict radial breathing at a characteristic Kepler period, \( \tau_K = \pi n_0^2/2 \approx 2.3 \) ps determined by the inverse of the mean energy difference between adjacent states in the wavepacket [1]. Unless otherwise indicated, atomic units are used throughout this paper. In general, the evolution of any wavepacket is completely determined by the complex coefficients of its constituent eigenstates. It is these coefficients that we seek to measure. To this end, the Ba atoms and ions in the laser–atom interaction region are exposed to a 0.6 ps HCP probe. Because the duration of the HCP is less than \( \tau_K \), it delivers an impulsive kick to the electron [9], [47]–[50]. The probability that the impulse ionizes the electron depends upon the strength of the kick and the wavepacket’s momentum distribution during the HCP. Accordingly, as shown previously for neutral wavepackets, the HCP can serve as an effective time-resolved probe of the ionic wavepacket momentum distribution [9]–[11].

The HCP is created by illuminating, in vacuum, a biased 1.5 cm \( \times \) 1.5 cm \( \times \) 0.05 cm GaAs wafer with a 100 fs, 780 nm Ti:sapphire laser pulse [43]. The HCP field is linearly polarized in the \( z \)-direction, parallel to the wafer bias field, and its magnitude is proportional to the bias voltage. The HCP beam is collected and directed into the laser–atom interaction region using a gold-coated off-axis parabolic mirror with a 10 cm focal length. The atoms interact with the lasers and HCPs between two, parallel, horizontal aluminum plates separated by 1.25 cm. Approximately 10 ns after the HCP, a small 60 V extraction pulse is applied to the lower field.
Figure 2. Double ionization signal versus HCP delay for different HCP field strengths. The HCP impulse increases from red (0.026 atomic units) to green (0.04 atomic units) to blue (0.048 atomic units). The HCP impulse was calibrated using the known ionization curves of the Ba 6s24d and 6s27d eigenstates [44]. The position of the blue curve has been vertically offset by $2 \times 10^{-7}$ in order to distinguish it from the green curve. For all three curves, the HCP impulse is less than that required to ionize the Ba$^+$ 38g state with 50% probability.

Plate. The extraction field pushes any positive ions in the interaction region through a 0.2 cm diameter hole in the upper field plate, toward a microchannel plate detector. Singly and doubly charged ions are distinguished by their time-of-flight to the detector. Doubly charged ions can be produced only via HCP ionization of the Ba$^+$ Rydberg wavepackets. The number of doubly charged ions created by impulsive ionization is recorded as a function of the time delay between the HCP and the excitation of the ionic wavepacket.

We note that the extraction field can produce Ba$^{2+}$ through field ionization of very highly excited ($n \geq 100$) Rydberg ions ([51] and references therein). These can be created during the interaction of the wavepackets with the HCP. Because we were unable to distinguish these very highly excited Rydberg ions from Ba$^{2+}$, in our analysis we assume that the energy $E_i = -2n^{-2}$ for $n = 100$ serves as the effective ionization limit for the ionic Rydberg states.

3. Results and discussion

Figure 2 shows the double ionization signal, $I(t)$, as a function of HCP delay for three different HCP field amplitudes. The delay-dependent variations in the double ionization yield reflect the changes in the wavepacket’s momentum distribution as it evolves. The connection between the ionization probability and the wavepacket’s momentum distribution can be understood by considering the energy exchange between an electron and an instantaneous field kick [9].
[48]–[50]. In classical terms, the HCP delivers an impulse \( \vec{A} = -\int \vec{F}_{\text{HCP}}(t)dt \), which ionizes the Rydberg electron if, at the instant preceding the pulse, the component of the electron’s momentum in the kick direction, \( p \), exceeds a critical value [9, 20],

\[
p_c = \frac{1}{A} \left( E_0 - \frac{1}{2} A^2 \right).
\]

Here, \( E_0 \) is the effective binding energy of the electron, and \( \vec{F}_{\text{HCP}}(t) \) is the time-dependent HCP field [9, 49, 50]. Quantum mechanically, the electron is represented by a wavepacket \( \Psi(\vec{p}, t) \), and the probability that the Rydberg atom is ionized by an impulse, \( A \), is equal to the probability that \( p > p_c \) at the instant of the pulse,

\[
I(t) = \int_{p_0}^{\infty} |\Psi(p, t)|^2 dp.
\]

We anticipate that immediately after the laser excitation, the \( \text{Ba}^+ \) 38g wavepacket will expand radially, reflect from the outer turning point in the \( \text{Ba}^+ \) potential barrier, and return to the nucleus in a time, \( \tau_K \) [1]. This process then repeats itself. According to equation (1), for a weak HCP probe, \( p_c \) is large and positive. HCP ionization can be efficient only at those times when the electron is near the ion core where it can have a large velocity component in the direction of the kick. Thus, we predict maxima in the \( \text{Ba}^{2+} \) yield when the wavepacket is localized near the nucleus and minima when the packet is moving slowly away from the ion [10, 52]. Conversely, for a strong HCP probe, \( p_c \) is large and negative. Only those electrons that are near the core and moving rapidly opposite to the kick direction will not be ionized. Accordingly, maxima in the double ionization signal are observed when the wavepacket is near its outer turning point [10, 52]. Signatures of the characteristic radial breathing collapse due to the anharmonicity of the potential, and subsequent fractional and integer revivals of the anharmonic wavepacket can be observed directly in the double ionization yields shown in figure 2.

A more complete view of the wavepacket evolution can be obtained from the time-dependent double ionization probabilities by extracting the complex amplitudes of the constituent eigenstates [16]. In general, the wavefunction for a Rydberg wavepacket can be expressed as

\[
\Psi(\vec{p}, t) = \sum_j C_j \psi_j(\vec{p}) e^{-i(E_j t + \phi_j)},
\]

where the index \( j \) represents all electronic quantum numbers and the \( \psi_j(\vec{p}) \) are real momentum–space eigenfunctions of the ionic potential with corresponding energies \( E_j \). For the radial motion considered here, the summation need only extend over the principal quantum number, \( n \). The real, time-independent amplitudes \( C_j \) and constant phases \( \phi_j \) are determined by the launch laser field and the transition moments between the initial \( \text{Ba}^+ 4\ell_{7/2} \) state and the Rydberg states. The time-dependent probability distribution is then

\[
P(\vec{p}, t) = |\Psi(\vec{p}, t)|^2 = \sum_{j,k} C_j C_k \psi_j(\vec{p}) \psi_k(\vec{p}) e^{-i[(E_k - E_j) t + (\phi_k - \phi_j)]}.
\]

In principle, if one has access to \( P(\vec{p}, t) \), then its Fourier transform can be used to extract values \( C_j \) and \( \phi_j \) for each eigenstate, \( \psi_j \) [16]. In practice, this complex amplitude retrieval method can be implemented by (i) measuring \( I(t) \) to obtain the integral of \( P(\vec{p}, t) \) over a momentum volume in which the eigenfunctions \( \psi_n(\vec{p}) \) are known (see equation (2)) and (ii) recording \( I(t) \) over a delay range that is sufficiently large so that peaks associated with each
energy difference, $\Delta E_{kj} = E_k - E_j$, can be resolved in its Fourier transform. These conditions are satisfied for the ionic radial wavepackets studied here.

To extract the radial eigenstate amplitudes and phases, it is advantageous to consider weak HCP kicks for which the double ionization probability $I < 50\%$. In this case, the delay-dependent variations in the ionization signal, $I(t)$, reflect the changes in the highest momentum portion of the wavepacket probability density. For these large momenta, the wavefunctions of adjacent eigenstates, $\psi_n$ and $\psi_{n+1}$, are nearly identical up to normalization factors $n^{-3/2}$ and $(n + 1)^{-3/2}$, respectively.

However, very weak kicks are not optimum because the relationship between the ionization signal and the wavepacket probability distribution in equation (2) is only approximate [53]. Firstly, by definition, the wavepacket has an energy uncertainty, $\delta E_0$, so, according to equation (1), $p_c$ is not well defined. Secondly, because of the non-zero HCP duration, the ionization signal is determined by the probability that the electron’s average, not instantaneous, momentum during the pulse exceeds the critical momentum, i.e. $\bar{p} > p_c$ [53]. Interestingly, the errors associated with both these effects can be reduced by using larger impulses. According to equation (1), the uncertainty in the critical momentum, $\delta p_c = \delta E_0/A$, decreases with increasing impulse. In addition, the probability that $\bar{p} > p_c$ becomes equal to the probability that $p > p_c$ as $p_c \to 0$, defining the 50% ionization point [15, 20]. This is because as $p_c$ decreases, smaller momentum values, for which the impulse approximation is more accurate, make up a larger fraction of the portion of the probability distribution that is ionized. As described in more detail below, we utilize the time-dependent ionization probabilities acquired with different impulses and, as a consistency check, compare the quantum-state amplitudes and phases obtained from each.

Figure 3 shows the magnitude of the Fourier transform, $S(\Delta E)$, of $I(t)$ for eight different impulse values. For all but the two weakest kicks, clear peaks appear at energies $10\text{ cm}^{-1} < \Delta E < 20\text{ cm}^{-1}$. These primary spectral features have magnitudes $S_{n+1,n}$ and phases $\Phi_{n+1,n}$ and are centered at energies corresponding to the splitting between adjacent Rydberg states with principal quantum numbers $n$ and $n+1$ [16]. In figures 3(a) and (b), no distinct primary peaks are observed, but a broad local maxima in $S(\Delta E)$ can be seen in the same energy region as the primary energy splittings. However, the maxima is clearly shifted to lower frequencies as compared to the distributions in the curves acquired at higher impulses. The shift at low impulses is a result of the variation in $p_c$ over the constituent eigenstates noted above. Specifically, the more weakly bound states in the wavepacket are more easily ionized by the weak HCP, skewing the apparent frequency distribution. Conversely, the intensity distributions for the primary peaks are approximately identical for figures 3(c)–(h). Apparently, the uncertainty in $p_c$ plays a negligible role at these kick strengths, making them more appropriate for characterizing the wavepacket. Secondary features, associated with energy differences between states $n$ and $n+2$, are also visible for $25\text{ cm}^{-1} < \Delta E < 35\text{ cm}^{-1}$ [16].

As alluded to above, spectral features associated with energy differences, $\Delta E_{kj}$, in figures 3(c)–(h), provide the necessary information to make an approximate reconstruction of the components of the quantum state relevant to the radial motion of the wavepacket [16]. In particular, we can set

$$
\frac{C_j}{C_k} = \frac{n_k^{3/2}S_{ji}}{n_j^{3/2}S_{ik}},
$$

(5)
Energy (cm$^{-1}$)

Figure 3. Discrete Fourier transform of measured delay-dependent double ionization signals for different HCP kick strengths. The HCP impulse increases from top to bottom with $A = 0.02$, 0.026, 0.03, 0.033, 0.037, 0.041, 0.044 and 0.048 arbitrary units, for curves a–h, respectively. For all impulses, the double ionization probability is less than 50% for the Ba$^+$ 38g eigenstate.

where $n_j$ and $n_k$ are the principal quantum numbers of states $\psi_k$ and $\psi_j$, respectively, and

$$\Delta \phi_{ji} = \Phi_{ji}. \quad (6)$$

Without loss of generality, we define the phase of the Ba$^+$ 38g state, $\phi_{38g} = 0$. Using this definition, the normalization condition, $\sum_j |C_j|^2 = 1$, and the relations (5) and (6), we recursively determine $C_j$ and $\phi_j$ for each state in the wavepacket.

Figure 4 shows two different determinations of the radial eigenstate amplitudes and phases for the Ba$^+$ ng wavepacket. To the best of our knowledge, this is both the first characterization of an ionic Rydberg wavepacket and the first demonstration of the Fourier transform method, rather than state-selective field ionization, to determine the eigenstate amplitudes in a Rydberg wavepacket. The two parameter sets are extracted from ionization measurements made with relatively small and large HCP impulses, respectively. We consider these separately as a check of the independence of the recovered amplitudes and phases on the HCP amplitude. The blue points show the average values computed using curves in figures 3(c) and (d). The red points are the averages computed from data acquired with higher impulses shown in figures 3(g) and (h). The difference in kick strength for the data sets in each group is small compared to the average impulse for those data. The same constituent states are observed in both cases, and the recovered state amplitudes and phases are almost identical within measurement error. That said, the errors in the state amplitudes extracted from thousands of laser shots in our experiment is greater than

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Figure 4. Amplitudes and phases of constituent radial eigenstates in a $n g^+$ ionic wavepacket in Ba retrieved using small (blue crosses) and large HCP impulses (red circles) via the method described in the text. The red points are average values extracted from the curves in figure 3(c) and (d), whereas the blue points were obtained from figures 3(g) and (h). The error bars represent the standard error of the mean for two different data sets in each group.

that typically attained from state-selective field ionization measurements averaged over only a handful of shots. Therefore, the field-ionization method, when feasible, is preferable.

We have performed wavepacket simulations utilizing the results from figure 4 as input for the eigenstate compositions. The wavepacket evolution predicted by these simulations, i.e. radial breathing, collapse, fractional and integer revivals, is in excellent agreement with that suggested by the ionization data shown in figure 2. We note that the measured phase variation across the constituent states is important for obtaining the correct dynamics. Specifically, due to the wavepacket chirp, the full revival occurs sooner than predicted for a wavepacket with uniform phase at $t = 0$. The primarily quadratic frequency chirp is likely to be the result of group velocity dispersion in the launch laser pulse due to its passage through various transmissive optical elements prior to entering the interaction region.

4. Conclusion

In summary, we have demonstrated for the first time that the time-dependent evolution of ionic wavepackets can be measured using HCP ionization. In particular, the collapse and revival characteristics of an ionic Ba$^+$ $n g$ radial wavepacket have been observed. Moreover, we have shown that the quantum-state components relevant to radial motion can be determined using an extension of a complex phase-retrieval method that was previously demonstrated for neutral wavepackets [16]. Accurate characterization of such ionic wavepackets is critical for controlling and probing double Rydberg wavepackets in the future.
Acknowledgments
This work was supported by the NSF.

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