Coherent time evolution of highly excited Rydberg states in pulsed electric field: Opening a stringent way to selectively field-ionize the highly excited states

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Highly excited Rydberg states in the ramped electric field is one of the most interesting systems which provide ideal and versatile situations for investigating the coherence effects in the time evolution of a quantum system with many potential-energy curves crossing one another. In spite of this interesting feature and also of the potential applicability to the wide area of fundamental physics including cavity QED and quantum computation, the Rydberg states with high principal quantum number have not been investigated in detail, partly because of the difficulty in selectively detecting a particular state from many close-lying states; in such highly excited states, the field ionization process generally occurs through the non-adiabatic and adiabatic transitions, resulting multiple ionization thresholds and the selective detection of a particular state becomes increasingly more difficult.

The purpose of this Letter is to present the experimental results on the field ionization of the Rydberg states with n = 98 – 150. It was observed for the first time that in high slew rate regime in the applied pulsed electric field, the field ionization process has single threshold value: Specifically the ionization electric field takes discrete values successively with increasing slew rate, and this dependence varies with the position of the states relative to the adjacent manifold. This transitional behavior in the field ionization shows regular dependence on the principal quantum number n, thus indicating that this behavior is quite general and applicable to a wide range of higher-lying Rydberg states. Since the differences in the field ionization values were found to be large enough, i.e. 300 % for the 111p3/2 and 111s1/2 states, it is possible to stringently select a low-ℓ state from the close-lying states by field ionization. From these characteristic behaviors, it is strongly suggested that the coherence in the time evolution under the pulsed electric field plays deciseive role to the behavior of the field ionization.

The experimental setup is shown in Fig. 1. Thermal Rb atoms in the ground-state atomic beam are passed through a laser excitation region and then the field ionization region, which are about 40 mm apart each other. The whole volume of the excitation and the field ionization regions is surrounded with three pairs of planer copper electrodes to compensate the stray field in three axes and also to apply the pulsed electric field for the ionization. The selective field ionization (sfi) electrodes consist of two parallel plates of 120 mm length, in one of which a fine copper-mesh grid was incorporated into the area of 20 × 20 mm², thus allowing to pass and detect the field ionized electrons with a channel electron multiplier.

The sfi electrodes and the laser interaction region were attached to a cold finger in a cryostat, thus the temperature can be varied from room temperature down to lower temperature with liquid N₂ and He.

Two-step cw-laser excitation was adopted to excite the Rydberg n_j states from the 5S1/2 ground state of ⁸⁷Rb through the 5p3/2 second excited state. A diode laser (780 nm for the first step) and a dye laser of coumarin 102 excited by a Kr ion-laser (479 nm for the second step) were used. The main reason to use the cw lasers to excite the Rydberg states instead of the usually adopted pulsed lasers is that we want to use this field ionization scheme for the selective ionization of highly excited Rydberg states in a continuous mode as discussed later.

The pulse shape applied for the field ionization is shown also in Fig. 1. The pulse sequence was produced with a waveform generator N6411 and the field ionization signals were detected and analyzed with the LabVIEW data acquisition system on a pc computer. Repetition rate of the pulse was kept to 5 kHz so that the detection efficiency of the Rydberg states is optimum for the atoms with velocity of 350 m/s. The ionization mainly occurs at the steep rise of the pulse during the time t_f, but the peak field can be kept for a time t_h (holding time) to ionize also the states with longer life time than t_f under the electric field. This point will be discussed later.
During the course of the present experiment, stray field of \( \sim 80 \text{ mV/cm} \) was found to appear in the interaction region. By applying suitable compensation potentials in three axes, the stray field was reduced to less than 10 mV/cm. In this stray field, the \( p \) state with \( n \) less than 120 is well separated from the adjacent manifold levels. The effect of the mixing of the low-\( \ell \) states to the neighboring levels on the field ionization behavior is discussed later.

In Fig. 2 shown are field ionization spectra of 111s_{1/2} and 111p_{3/2} states as a function of the applied electric field \( F = \frac{v_f}{t_f} \) (\( t_f \) is the distance of the sfi electrode), which were measured by varying the slew rate \( S = \frac{F}{t_f} \). Here the slow component of the pulsed field \( v_s \) was set to zero.

In these spectra, only one prominent peak was found as the threshold electric field. Remarkably the sfi field value at the peak changes to a smaller value with increasing slew rate at a particular slew rate. Moreover the sfi fields for the \( s \) and \( p \) states are quite different from each other at the same slew rate. For example, the electric field necessary to ionize the 111p_{3/2} state at the slew rate of 11 V/(cm-\( \mu \)s) is 1.7 V/cm, while the value is 5.2 V/cm for the 111s_{1/2} state, more than 300 \% difference (see the spectrum \( d \) in Fig. 3).

It should also be noted here that the transitional behavior in the 109d state (not shown in Fig. 2 to avoid complexity) is the same as in the s_{1/2} state. This means that the transitional behavior depends on the position of the states relative to the adjacent manifold: As seen from the relevant Stark energy-field diagram in Fig. 2, the upper – positioned \( p \) state shows different transitional behavior from the lower – positioned \( s \) and \( d \) states.

In Fig. 3 shown is the effective principal-quantum-number \( n^* \) dependence of the critical slew rate \( S_c \) and the ionization electric fields \( F_c \) for the \( s \) and \( p \) states. Here the critical slew rate \( S_c \) is defined as the value at which the transition of the sfi field just starts to a new value. Also the sfi field corresponds to the peak position of the ionization signal. These values vary quite regularly with \( n^* \) ranging from 95 to 147; approximately \( F_c \propto (n^*)^{-4.0} \) for both the \( p \) and \( s \) states, while \( S_c \propto (n^*)^{-4.0} \) for the \( p \) state and \( S_c \propto (n^*)^{-2.8} \) for the \( s \) state.

These results indicate that this transitional behavior is quite general for a wide range of \( n \) and thus can be applicable to selectively ionize the \( s \) and \( p \) states for a wide range of higher excited states. The difference in the ionization field is \( \sim 300 \% \) for the \( s \) and \( p \) states, quite large compared to the case of adiabatic transition in which the difference would be only 5 \% for the states at \( n \sim 110 \).

Related to the selectivity in the Rydberg states, it should be noted that there observed some sfi signals at the lower field region (1.0 \( \sim 2.4 \text{ V/cm} \)) in the sfi spectrum of \( s \) state: The signal peak at this portion changed discretely with slew rate as in the same manner as of the \( p \) state. This part of signal counts is due to the effect of blackbody radiations which induce the transition from the initial \( s \) state to the \( p \) state. In fact the counts at this portion measured by varying the temperature of the excitation-detection region from 120 K to 40 K was found to depend linearly on the temperature as expected. The observed transition rate to the \( p \) state is in rough agreement in its absolute values and in good agreement in its temperature dependence with the theoretical predictions. This agreement indicates also that the selectivity of the excited states with the field ionization method in the pulsed electric field regime is quite good even at such
highly excited region. The detailed discussion on the s to p transitions and the effect of blackbody radiations will be reported elsewhere.

The above results were all obtained without the slow component \( v_s \) of the pulsed field. Switching on this value, the transitional behavior of the s state remains the same, while that of the p state changes drastically as in the following: When the applied slow component field with its slew rate less than 1 mV/(cm-µs) exceeds the first anti-crossing field \( (\sim 75 \text{ mV/cm}) \) for the \( 111p_{3/2} \) state, see Fig. 3, the transitional behavior changes abruptly, becoming the same as of the s state. This means that once the first anti-crossing is traversed adiabatically and the state is mixed with the adjacent manifold levels, then the following sfi behavior for the p state under the high slew rate regime changes completely, resulting no difference in their behavior between the states of opposite positions to the adjacent manifold.

These experimental results, especially the transitional behavior, are in general not in agreement with simple predictions from the incoherent contributions of adiabatic and non-adiabatic transition processes: The experimental sfi spectra show only one prominent peak and this peak field does neither correspond exactly to the expected position from the purely adiabatic (paths 2 or 3 in Fig. 3); nor diabatic transitions (paths 1 or 4) leading to the reddest or bluest trajectory in the adjacent manifold; the expected fields for these paths (1 to 4) in \( n = 108 \) (corresponding to the adjacent manifold of \( 111s \) and \( 111p \) states from the quantum defect values in Rb) are 1.8, 2.4, 2.4, and 4.2 V/cm respectively, which should be compared to the observed field values of 1.7 and 5.2 V/cm for the p and s states, respectively. Moreover importantly, these spectra show very clear slew rate dependence as described above which can not be explained from a simple incoherent process.

Recently Harmin [2] examined coherent time evolution on a grid of Landau-Zener anti-crossing under a linear ramped electric field in which each manifold levels are treated as linear in time, parallel, and equally spaced and infinite number. The time development of an initially populated state is then governed by two level Landau-Zener (LZ) transitions at avoided crossings and adiabatic evolution between them. The key parameters in this model study are 1) the LZ transition probabilities \( D \) for making a non-adiabatic transition process at the anti-crossing traversals and 2) the dynamical phase unit \( \varphi \). The phase unit \( \varphi \) is the area covered by the pair of the adjacent up- and down-going levels. The overall difference in phase advance \( \Delta \Phi \) for two paths from zero to the ionization field is equal to the sum of the phase units \( \varphi \) in the whole area covered by the two paths. In the actual Stark energy-field grid system, the phase units \( \varphi_i \) are not a constant but vary with respective anti-crossings. These parameters \( D \) and \( \varphi \) are estimated approximately by \( D = \exp(-\pi \hat{\mu}^2 \varphi) \), \( \varphi \sim (3Fn^{10})^{-1} \), where \( \hat{\mu} \) is an average low-\( \ell \) quantum defect and \( F \) is the slew rate of the applied electric field.

The probability \( D \) increases monotonically with increasing slew rate, while the overall difference in phase advance \( \Delta \Phi \) of the state wave function through the grid is effective in modulo \( 2\pi \) and strongly affects the coherent nature of the process through the interference between many states populated along the way of traversals in the applied electric field.

In the present experimental setup with slew rate \( \sim 20 \text{ V/(cm-µs)} \) at \( n \sim 100 \), the probability of non-adiabatic transition \( D \) is quite high, reaching \( \sim 99.8\% \) from the above estimation so that our case corresponds to the non-adiabatic limit in Harmin’s treatment in a good approximation.

![Fig. 3. Stark energy diagram near the 108 manifold in Rb together with the classical field-ionization threshold-line \( (E = -6.12\sqrt{F}) \), where all the avoided crossings are not explicitly shown for simplicity. The arrows 1 to 4 indicate the possible trajectories for the adiabatic (2 and 3) and the extreme non-adiabatic (1 and 4) transitions.](image)

![Fig. 4. Dependence of the selective field ionization (sfi) value and the critical slew rate on the effective principal-quantum-number \( n^* \) in Rb Rydberg states. Discrete two values of the sfi field observed and their corresponding slew rates are plotted together with the fitted lines of \( n \)-dependence. The number to each plot is the coefficient \( \alpha \) in the fitting of \( (n^*)^{-\alpha} \) dependence with the estimated error of \( \pm 0.2 \).](image)
low $\ell$ states, estimated phase unit $\varphi$ varies from $10^{-2}$ to $10^{-3}$ with increasing electric field. The number of avoided crossings traversed between zero field and the field ionization region in Rb is estimated to be $N_{\text{blue}} \sim 900 \left( \frac{n}{100} \right)^2$, $N_{\text{red}} \sim 1150 \left( \frac{n}{100} \right)^2$ for the up- (bluest) and down- (reddest) going trajectories, respectively. At the region of $n \sim 110$, this number of anti-crossing traversals suggests that the phase advance is over $2\pi$ at some field value. When this resonance condition is fulfilled, the constructive interference between the many number of Stark states along the advance of the pulsed electric field may result in one prominent peak in the sfi field. In Harmin’s analysis [2], the population of the states under the electric field was found to make a series of resonances in a form of lanes along the up- or down-going directions of the initially excited state at zero field.

This general feature in the non-adiabatic limit seems to be satisfied experimentally, since a discrete sequence of sfi threshold field was found, depending on the slew rate. However there is a significant difference in the experimental observations compared to the model calculation in that the observed sfi field values for both of the $s$ and $p$ states decrease with increasing slew rate. Contrary to this observation, the model calculation predicts that with increasing slew rate, the most populated state approaches to the limiting trajectory in the manifold, i.e. to the bluest state for the up-going initial state or the reddest state for the down-going initial state, depending on the direction (up- or down-going) of the initial state. This suggests thus that the sfi threshold field for the $s$ and $d$ states in Rb should increase with increasing slew rate, in disagreement with the experimental results.

Finally we note that the sfi spectra observed with the holding time $t_h$ extended up to 500 $\mu$s showed no distinguishable difference from those taken with the short holding time of 1$\mu$s, indicating all the states ionized have decay lifetime shorter than 1 $\mu$s in the electric field. Also we observed no significant sfi-signal over 10 $V$/cm in the spectra measured. The decay rate of the blue states in the electric field by the tunneling process, estimated from the hydrogenic approximation, is much higher than $2 \times 10^9 s^{-1}$. Therefore the ionization process for the states along the blue lines is not due to the tunneling process but of autoionization-like one due to their mixing to the red continuum, even though such coupling becoming weaker as $n$ increases [1].

In conclusion, we observed for the first time a discrete transition of the threshold sfi field with slew rate in the highly excited Rydberg states in Rb, the behavior of which depends also on the position of the low $\ell$ states relative to the adjacent manifold. The experimental results strongly suggest that the coherent interference effect in the time evolution on the grid of anti-crossings under the pulsed electric field plays decisive role for the occurrence of such transitional behavior.

The transitional behavior observed here brings us a new powerful method to selectively field-ionize the low $\ell$ states from the many close-lying states, thus opening a new way to apply the highly excited Rydberg states to fundamental physics research. One of the example of such applications is to search for dark matter axions with a Rydberg-atom cavity detector [7]. In this kind of search experiment, it is essential to do the experiment in a continuous way so as to keep the detection efficiency as high as possible. It is thus inevitable to use cw lasers to excite the Rydberg atoms continuously which is the main reason for developing the present experimental setup.

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[4] Taking into account the velocity distribution of atoms and the detection efficiency in the pulsed field ionization, the overall detection efficiency is estimated to be 75 %.
[5] Note that the sfi detector used is of integral type in a sense that the sfi signals are integrated with increasing sfi field $v_f$ and thus the derivative of the spectra with respect to $v_f$ is the true counts at each bin which is shown in Fig. 2.
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