Systematic observation of tunneling field-ionization in highly excited Rb Rydberg atoms

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Abstract

Pulsed field ionization of high-\(n\) (90 \(\leq n \leq 150\)) manifold states in Rb Rydberg atoms has been investigated in high slew-rate regime. Two peaks in the field ionization spectra were systematically observed for the investigated \(n\) region, where the field values at the lower peak do not almost depend on the excitation energy in the manifold, while those at the higher peak increase with increasing excitation energy. The fraction of the higher peak component to the total ionization signals increases with increasing \(n\), exceeding 80\% at \(n = 147\). Characteristic behavior of the peak component and the comparison with theoretical predictions indicate that the higher peak component is due to the tunneling process. The obtained results show for the first time that the tunneling process plays increasingly the dominant role at such highly excited nonhydrogenic Rydberg atoms.

Key words: Rydberg atom, pulsed field ionization, tunneling process, \(^{85}\text{Rb}\)
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Rydberg atoms have been utilized in fundamental physics research, such as in the field of cavity quantum electrodynamics\[1\]. Area of their application fields has recently been more enhanced with the development of the quantum computing and its related quantum measurements\[2\]. In these experimental studies, the field ionization method has been widely used for the detection of highly excited atoms. Most applications of Rydberg states up to now have been performed by using alkali atoms with the principal quantum number \(n\) less than 60 or so and it is known in this region that the field ionization occurs mostly around the classical saddle-point field-value given by \(F_c = W^2/4\) (in a.u.), where \(W\) is the binding energy of the Rydberg states. There often observed are two peaks in the field ionization spectra due to the adiabatic and non-adiabatic processes in the time evolution of the Rydberg atoms in the electric field\[4,5\]. This ionization is due to the level mixing in the ionic core of the bound blue state with the continuum red states coming down from the higher \(n\) states (autoionization-like process) \[3–5\]. This field ionization due to the level mixing effect occurs at the field region between the classical saddle point value and the value expected from the tunneling process\[4,5\]. At the low \(n\) (less than \(\sim 60\)) region, the ionization due to the tunneling process has not been observed usually in nonhydrogenic atoms because the ionization due to the autoionization-like process has much shorter ionization-lifetime than that from the tunneling process (see, however, Neijzen and Dönszelmann\[6\] and Rolfes, Smith and MacAdam\[7\]).

Since the level mixing in the ionic core interior is weaker with increasing \(n\) by a factor of \(n^2\), it is naturally expected that the competition between the autoionization-like and the tunneling processes occurs at some \(n\) region and thereafter the tunneling process will dominate the ionization process. To our best knowledge, no detailed investigations have been reported for the field ionization behavior in highly excited Rydberg atoms with \(n\) higher than 70. In addition to the above intrinsic interests in the physics of pulsed field ionization mechanism in highly excited Rydberg atoms, such studies are inevitably important to extend the applications of the Rydberg atoms to still higher excited region where the coupling to the external electromagnetic field is increasingly stronger and thus open new areas of application fields.

We report here the experimental results of the detailed systematic investigation of field ionization behavior of the highly excited Rb Rydberg states with \(n\) ranging from 90 to 150. Specifically we focussed on the high-\(n\) manifold states which are degenerate in energy at zero electric field except for some low angular-momentum (\(\ell\)) states, but are split under the electric field. After exciting these splitting manifold states from a low-lying 5p\(_{3/2}\) state with lasers under a small electric field, pulsed electric field was applied to ionize the states in high slew rate regime. It was found for the first time that the tunneling process is increasingly the dominant process in the pulsed field ionization at such highly excited Rydberg atoms. Here the systematic occurrence of the
tunneling process and comparison of the results with theoretical predictions are presented and discussed with applications of such highly excited Rydberg states.

Schematic diagram of the present experimental setup is shown in Fig. 1. A thermal atomic beam of Rb was introduced at the center of the field ionization electrodes, where the $^{85}$Rb atoms in the ground state $5s_{1/2}$ were excited to the Rydberg manifold states through the second excited $5p_{3/2}$ state by the two-step laser excitation. A diode laser (780 nm) and a dye laser (479 nm) with a dye of coumarin 102 pumped by a Kr ion laser were used for the first and second step excitations, respectively. The polarizations of the laser lights adopted in the present experiment are parallel and perpendicular to the electric field applied for the first and the second lasers, respectively.

A linearly ramped electric field was applied for the field ionization in the present experiment. For this purpose a sequence of voltage pulse was generated by a waveform generator AWG420 (Sony Tektronix) and amplified by fast amplifiers before applying to the electrodes. The electrodes consist of two parallel plates of 52 mm length and 40 mm width, the distance between them being 24 mm.

The electrons liberated by the field ionization were guided to a channel electron multiplier through two fine-mesh grids placed in one of the electrode plates. Ionization signals of electrons from the channel electron multiplier were amplified by a preamplifier and a main amplifier and then fed to a multichannel scaler P7886 (FAST ComTek) after the pulse height discrimination. The ionization events were counted as a function of the elapsed time from the starting time of the ramp field with the dwell time of 500ps. From the correspondence

3 No appreciable change in the experimental results was observed, however, with other configurations such as both parallel and perpendicular cases.
Fig. 2. Relevant Stark energy diagram and field ionization spectra for the $112$ manifold states. Only every one of three states in the split $n=112$ Stark manifold states are shown for clarity. Dotted and solid lines are the theoretical predictions of the ionization threshold values expected from the classical saddle point ionization ($F_c$) and from the tunneling process ($F_t$), respectively. The manifold states were initially populated under the static electric field of 75 mV/cm, and subsequently ionized with a pulsed electric field at the slew rate of $13.4 \text{ V/(cm-µs)}$. Here $W_r$ is a parameter representing the excitation position of the manifold states in which the values $+1.0$ and $-1.0$ correspond to the uppermost (highest energy) and the lowermost (lowest energy) states, respectively.

of the applied electric field and the time bin, the observed timing spectra were converted into the field ionization spectra as a function of the applied electric field.

The manifold states were excited under a small electric field applied. The applied field was set to $\sim 0.7F_c$, where $F_c(=1/3n^3)$ is the field at which $n$ and $n \pm 1$ manifolds cross first. This field is larger than the first avoided-crossing field between the $p$ state and the adjacent manifold in $^{85}\text{Rb}$, and yet smaller than $F_c$. It is noted here that the manifold states can be excited from the low-lying $p$ state only under the electric field because they are intrinsically high $\ell$ states in zero field. Even under the electric field, however, the extreme states near the bluest and the reddest states are hard to be excited appre-
ciably, thus being unable to be used as a reference to the excitation position of the manifold states. Precise position of the excited states in the manifold was therefore determined from the wavelength difference of the second laser relative to the excitation of $s$ and $p$ states at the applied electric field. For this purpose, exact excitation energies of the $s$ and $p$ states, which already enter into the manifold and thus not being well separated from the manifold states, were evaluated from the calculated excitation energies at which the $s$ and $d$ components in their wave functions have maxima among the close-lying manifold states. The Stark energy structure calculated with the method of Hamiltonian diagonalization[8] is shown to be precise enough for this purpose even for such high-lying states[9].

Typical field ionization spectra measured at the $n = 112$ manifold with slew rate $13.4V/(cm\cdot\mu s)$ are shown in Fig. 2. There observed are two prominent peaks, the higher one of which moves appreciably in its field ionization value with the excitation position in the manifold, while the lower one of which shows almost no dependence on the excitation position in the manifold.

Observed values of the ionization field corresponding to the higher peak are plotted in Fig. 3 for the two cases of $n$ as a function of the excitation position $W_r$ in the manifold. Here the excitation position $W_r$ in the manifold with its binding energy $W$ is expressed by

$$W_r = 2\frac{W-W_{\min}}{W_{\max}-W_{\min}} - 1,$$

where $W_{\max}$ and $W_{\min}$ are the binding energies of the uppermost (bluest state) and the lowermost (reddest state) energy states in the manifold, respectively.

Also shown in this figure with dotted and dashed lines are the theoretical predictions of the ionization field from the tunneling process; the expected ionization field from this process was calculated with a semi-empirical formula of ionization rate in the electric field by Damburg and Kolosov[10]. Although this formula was originally developed for Hydrogen and less accurate in its prediction of ionization lifetime compared to the density-of-state method by Luc-Koenig and Bachelier[11], Damburg and Kolosov prediction for the ionization field is considered to be accurate enough for the present purpose.

The theoretical predictions are in good agreement with the experimental results if the ionization paths under the applied pulsed electric field from the respective excitation positions are along the diabatic trajectories in the Stark energy diagram as shown by thick solid lines with arrows in Fig. 2. This agreement was found to be well satisfied for all the investigated Rydberg states with $90 \leq n \leq 150$.

The fraction of the higher peak component to the total ionization signals is shown in Fig.4 as a function of $n$. The fraction increases with increasing
Fig. 3. Observed values of the ionization field corresponding to the higher peak component as a function of the excitation position in the manifold for \( n = 112 \) and 127. Lines are the theoretical predictions of the field values expected from the tunneling process.

Another important experimental result is that this fraction of tunneling process increases with higher slew rate as shown in Fig. 5. This feature is also consistent with the theoretical prediction that the field ionization due to the autoionization-like process has finite lifetime which depends on the electric field applied\(^9\); steeper the applied electric field pulse, shorter time is allowed for the atoms to be ionized with this autoionization-like process, thus inducing relative increase of the fraction of the ionization component due to the tunneling process.

These observations are the first clear indications of the systematic occurrence of the tunneling process in these highly excited nonhydrogenic Rydberg atoms with \( n \geq 90 \). Previously two groups\(^6,7\) observed a field ionization peak due to the tunneling process for the \( 66^p \) \((^2P)\) state in In and \( 34d \) state in Na.

Finally we comment on the lower peak behavior. Contrary to the clear behavior of the higher peak component from the tunneling process, the lower peak can not be explained simply from the expected behavior of the autoionization-like process; if the ionization path follows the same diabatic line as in the case of higher peak component (tunneling process), the ionization field would depend on the excitation position of the manifold states, approximately given by the crossing point between the line \( F_c \) and each straight line of diabatic trajectory in Stark energy diagram as shown in Fig. 2. This is not the case in the actual situation as shown in the present experiment. The behavior is rather consistent with that expected from the assumption that the initial manifold states take adiabatic paths to ionization at the classical saddle point values \( F_c \), although the slew rate of the electric field pulse applied actually is too high for allowing the adiabatic paths in the individual avoided crossings. More detailed
Fig. 4. Fraction of the higher peak component to the total ionization signals as a function of $n$. Excitation position in the manifold is at the center of the manifold, $W_r = 0$, where the external electric field $F \sim 114 \cdot (100/n)^5 \text{mV/cm}$ was applied at its initial excitation. Slew rate of the applied pulse is $13.4 \text{V/(cm·µs)}$.

Fig. 5. Slew rate dependence of the fraction of the higher peak component to the total ionization signals for the $n=98$ manifold.

experimental results together with discussions on the characteristic feature of the lower peak component will be presented elsewhere.

In conclusion we have studied the pulsed field ionization behavior of highly excited Rb Rydberg atoms in high slew rate regime. Specifically we focussed on the field ionization behavior of the manifold states in high $n$ region. It was found for the first time that the tunneling process plays increasingly the dominant role for the field ionization at such highly excited nonhydrogenic Rydberg atoms with $n$ ranging from 90 to 150. The fraction of the ionization component from the tunneling process increases with increasing slew rate. This behavior brings us an important application of high-lying Rydberg atoms to serve as a sensitive microwave single-photon detector over the 10-cm wavelength region[12]; in this new method, two low-$\ell$ states which lie below and above the adjacent manifold, respectively, are selected as the lower and the upper states for the Rydberg-atom microwave detector. The lower state initially pre-
pared (the upper state populated by absorbing photons) is then transferred to the lowermost (uppermost) state in the adjacent manifold adiabatically. The resulting extreme states in the manifold are then selectively ionized with the following pulsed electric field in high slew rate regime. The method was demonstrated to be quite stringent in selectivity for the highly excited Rb Rydberg atoms[13].

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