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# DECAY RATES OF LARGE-$l$ RYDBERG STATES OF MULTIPLY CHARGED IONS APPROACHING SOLID SURFACES

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**Abstract.** We investigate the ionization of large-$l$ multiply charged Rydberg ions approaching solid surfaces within the framework of decay model and applying the etalon equation method. The radial coordinate $\rho$ of the active electron is treated as a variational parameter and therefore the parabolic symmetry is preserved in this procedure. The complex eigenenergies are calculated from which the energy terms and the ionization rates are derived. We find that the large-$l$ Rydberg states decay at approximately the same ion-surface distances as the low-$l$ states oriented toward the vacuum and considerably closer to the surface comparing to the low-$l$ states oriented towards the surface.

## 1. INTRODUCTION

The neutralization and ionization processes of Rydberg ions during the ion-surface interaction represent the complex quantum events that require a detailed description of the intermediate stages of the processes.

Depending on the collision geometry and energy, the charge state of the ionic projectiles, and the quantum state of the active electron these two basic one-electron processes can be treated by rather different theoretical tools. Here we mention the coupled angular mode (CAM) method, see e.g. Borisov et al. (1996).

A specific description of the neutralization can be obtained within the framework of two-state vector model (TVM), see e.g. Nedeljković and Nedeljković (1998) and Nedeljković et al. (2003). The TVM has been applied both to the large-$l$ and low-$l$ cases, where $l$ is the angular momentum quantum number. It was demonstrated that the reionization of the previously populated Rydberg states could play an important role. However, only the low-$l$ ionization and reionization have been considered; the problems have been treated within the framework of etalon equation method (EEM) by Nedeljković and Nedeljković (2003, 2005) and by Nedeljković et al. (2006). The information about the (re)ionization of the large $l$ Rydberg states is still uncomplete.

In the present paper, we analyze the ionization of multiply charged ($Z \gg 1$) ions approaching solid surfaces at velocity $v$, being initially in the large-$l$ Rydberg states ($n \gg 1, l \approx n-1, m=0$). In contrast to the low-$l$ states, the large-$l$ states evolve, under plausible physical assumptions, into the low-eccentricity parabolic states in the

vicinity of the surface. We consider the ionization as a decay of the formed intermediate state $\bar{\Psi}$; the ionization rates follow directly from the complex eigenenergies corresponding to the eigenfunctions $\bar{\Psi}$. In solving the complex energy eigenvalue problem, we use the EEM adapted for the intermediate states of low-eccentricities.

## 2. DECAY MODEL

Here we extend the low-$l$ decay model of Nedeljković and Nedeljković (1998, 2003) to the large-$l$ case.

We consider the ionization process $\mathrm{A}^{(Z-1)+} + \mathrm{M} \rightarrow \mathrm{A}^{Z+} + \mathrm{M}(e)$ of the ionic projectile $\mathrm{A}^{(Z-1)+}$ in the large-$l$ Rydberg state (or more precisely, being initially in the large-$l$ state) approaching a solid surface (M) at velocity $v = v_{\perp} = -dR/dt$, along the $z$ axis orthogonal to the surface. At the time $t = t_{in}$ ($R = R_{in} \rightarrow \infty$) the active electron is in the state $\Psi_{\nu,in}$, where $\nu = (n, l, m)$ represents a given set of spherical quantum numbers.

The electron wave function in the absence of ionization evolves according to the law $\Psi(\vec{r}, t) = \hat{U}(t_{in}, t)\Psi_{\nu,in}$, where $\hat{U}(t_{in}, t) = \exp(-i\int_{t_{in}}^{t} \hat{H}dt)$ is the evolution operator and $\hat{H}$ is the corresponding Hamiltonian. Expanding $\Psi_{\nu,in}$ over the parabolic basis $\Psi_{\mu,in}$, characterized by the parabolic quantum numbers $\mu = (n_1, n_2, m)$, where $n_1 + n_2 + |m| + 1 = n$, we get

$$\Psi_{\nu,in} = \sum \langle \mu|\nu\rangle \Psi_{\mu,in}, \tag{1}$$

where $\langle \mu|\nu\rangle$ are the expansion coefficients. In the large-$l$ case ($l \approx n - 1$) and for $m = 0$, the main contribution to the expansion (1) give the terms with $n_1 \approx n_2$, i.e., $n_1 \approx (n-1)/2$. This can be proved if the coefficients $\langle \mu|\nu\rangle$ are expressed via Clebsh-Gordon coefficients; for example, for $l = n - 1$ and $m = 0$ we have

$$\langle \mu|\nu\rangle = \frac{\sqrt{2l+1}}{\sqrt{(2n-1)!}} \frac{[(n-1)!]^2}{n_1!(n-n_1-1)!}. \tag{2}$$

At intermediate stages, the "large-$l$" wave function $\Psi(\vec{r}, t)$ behaves as a parabolic function corresponding to the intermediate parabolic quantum numbers $\mu$ with $n_1 \approx n_2$. On the other hand, the initially low-$l$ Rydberg states evolve into the intermediate parabolic states with $n_1 \approx 0$ (states of large eccentricities oriented toward the surface) and the states with $n_1 \approx n - 1$, which are highly eccentric, but which are oriented toward the vacuum.

The ionization process represents a decay of the function $\Psi(\vec{r}, t)$: $\Psi \rightarrow \bar{\Psi}$, where

$$\bar{\Psi}(\vec{r}, t) = \mathcal{E}_{\mu}(t)\Psi(\vec{r}, t)\Theta(z) + \chi(\vec{r}, t)\Theta(-z). \tag{3}$$

The regions inside and outside the solid ($z < 0$ and $z > 0$) are defined by the Heaviside function $\Theta(z)$. The function $\chi(\vec{r}, t)$ in Eq. (3) stands for the outgoing wave inside the solid. According to expression (3), the state $\bar{\Psi}$ represents a decaying state, with a "parabolic" decay factor of the following form:

$$\mathcal{E}_{\mu}(t) = \exp\left(-\frac{1}{2}\int_{t_{in}}^{t} \Gamma_{\mu}^{I}(t)dt\right), \tag{4}$$

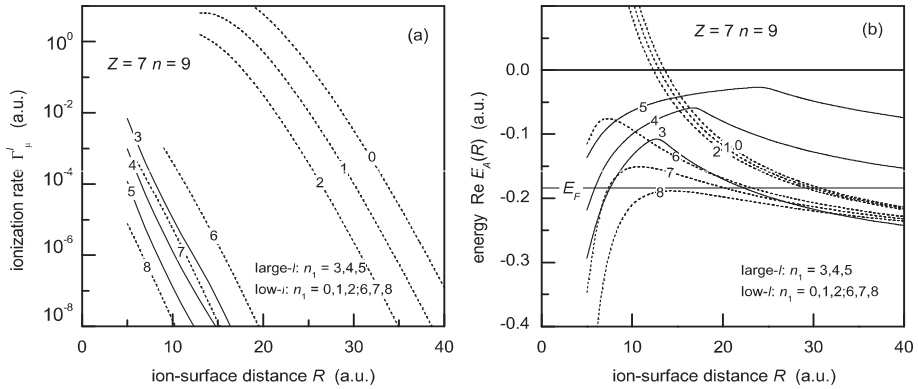


Figure 1: (a) Ionization rates $\Gamma^I_\mu(t)$ for the ion with core charge $Z = 7$ for the large-$l$ Rydberg states $n = 9$, $n_1 = 3, 4$ and 5 and $m = 0$ (full curves) and the low-$l$ states $n = 9$, $n_1 = 0, 1, 2; 6, 7, 8$ (dashed curves) and (b) the corresponding energy terms $\mathrm{Re}E_A(R)$.

where $\Gamma^I_\mu(t)$ is the ionization rate. The quantity $\Gamma^I_\mu(t)$ follows from the complex eigenenergies $E_A = \mathrm{Re}E_A - i\Gamma^I_\mu/2$.

The crucial difference of the large-$l$ case in comparison to the low-$l$ case is the fact that the radial coordinate $\rho$ in the surface potential can not be neglected. For that reason, the separation of variables in the parabolic coordinates is possible only if we consider the variable $\rho$ as parameter ($\rho = \bar{\rho}$). In adapting the EEM to the large-$l$ case, we define the appropriate functional $T[\Phi]$, which is stationary for $\Phi = \bar{\Psi}$. Accordingly, the parameter $\bar{\rho} = \bar{\rho}_0(\mu; R)$ can be derived from the variational requirement $\delta T = 0$. The proposed EEM enables us to evaluate the complex eigenenergies $E_A$, from which we get the ionization rates $\Gamma^I_\mu(t) = -2\mathrm{Im}E_A$ and the corresponding energies $\mathrm{Re}E_A$ in sufficiently accurate form. The low-$l$ EEM theoretical predictions for the ionization via tunneling mechanism follow from the large-$l$ expressions under the transformation $\bar{\rho}_0 \Rightarrow 0$.

## 3. RESULTS

According to Eqs. (3) and (4), the problem of ionization in the large-$l$ case is reduced to the analysis of the "parabolic" rate $\Gamma^I_\mu(t)$. The calculation of the quantity $\Gamma^I_\mu(t)$ can be performed in two steps. In the first step we calculate the rates $\Gamma^I_\mu(\bar{\rho}, R)$ with $\bar{\rho}$ as a parameter. In the next step, we calculate the quantity $\bar{\rho} = \bar{\rho}_0(\mu; R)$; in that way we obtain the rates $\Gamma^I_\mu(\bar{\rho}_0(\mu; R), R) \equiv \Gamma^I_\mu(t)$. The EEM yields directly the quantities $\mathrm{Re}E_A(\bar{\rho}, R)$ and $\Gamma^I_\mu(\bar{\rho}, R)$, without explicit calculation of the eigenfunction $\bar{\Psi}$.

In Fig. 1 we present the characteristic behavior of the large-$l$ rates and energies (full curves) via ion-surface distance $R$, for the Rydberg states $n = 9$, $n_1 = 3, 4$ and 5, $m = 0$ and ion with core charge $Z = 7$. The dashed curves in Fig. 1 represent the rates and energies for the low-$l$ parabolic Rydberg states with $n = 9$, $n_1 = 0, 1, 2, 6, 7$ and 8. By $E_F$ we denote the Fermi level of the Al-surface with work function $\phi = 5$ eV.

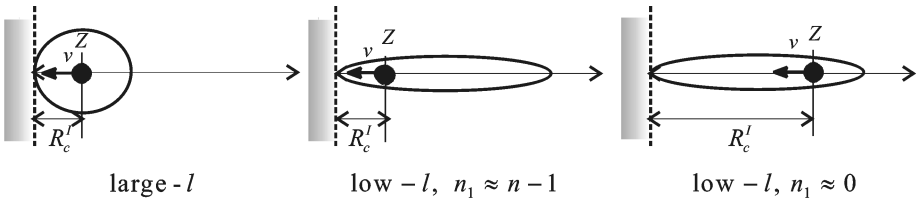


Figure 2: Behavior of the Rydberg states in the decay region $R \approx R_c^I$.

The rates presented in Fig. 1(a) are localized in two separate regions of the ion-surface distances: the large-$l$ rates and the low-$l$ rates with $n_1 \approx n-1$ (I group of states) are localized closer to the surface comparing to the rates corresponding to the low-$l$ case with $n_1 \approx 0$ (II group of states). The discussed positions of the rates indicate that the two group of states (I and II) will decay at separate ion-surface distances $R_c^I$: the almost spherical large-$l$ states and the eccentric low-$l$ states with $n_1 = 6$ and 7, oriented from the surface toward the vacuum, will decay at smaller ion-surface distances in comparison to the low-$l$ states ($n_1 = 0, 1, 2$) - oriented toward the surface. The CAM-rates of Borisov et al. (1996) are positioned at ion-surface distances just between our large-$l$ and low-$l$ EEM rates.

From Fig. 1(b) we see that the energy terms of the considered two group of states have also different behaviors: the energy terms corresponding to the first group: the large-$l$ energy terms (full curves) and the low-$l$ energy terms for $n_1 \approx n-1$ (dashed curves) have the characteristic increasing-decreasing behavior with decreasing $R$. On the other hand, the low-$l$ energy terms with $n_1 \approx 0$ increase with decreasing $R$. The energy terms of CAM method behaves as the low-$l$ terms from the second group of states.

In Fig. 2 we illustrate the ionization of these two group of states within the framework of classical "bubble" picture in which the ionization occurs when the ion touches the surface.

We recall that in the presented decay model the large-$l$ and low-$l$ states are related with the $l$-values of the initial states; these states evolve into the low- and large-eccentricity intermediate parabolic states, respectively. The quantum number $l$ can be used to characterize the intermediate stages of the electron-exchange process only within the TVM in which the two functions are used to define the quantum state of a single electron.

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