

# One-Electron Rydberg Quasimolecules in a High-Frequency Linearly-Polarized Laser Field

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**ABSTRACT:** The problem of electron terms in the field of two stationary Coulomb centers of charges  $Z$  and  $Z'$  separated by a distance  $R$  is one of the most fundamental problems in quantum mechanics. When the charges  $Z$  and  $Z'$  approach each other and share the only one electron that they have, they form a quasimolecule. Such quasimolecules are encountered in various kinds of plasmas and play an important role in theoretical and experimental studies of charge exchange. When the electron is in a highly-excited state, it is a one-electron Rydberg quasimolecule (OERQ). There are extensive analytical studies of the OERQ by the methods of classical mechanics (which are appropriate for Rydberg states). In one of our previous papers we studied the OERQ subjected to a laser field in the situation where the laser frequency was much smaller than the highest frequency of the unperturbed system. In the present paper we consider the situation where the OERQ is subjected to a linearly-polarized laser field whose frequency is much greater than the highest frequency of the unperturbed system. For obtaining analytical results we use a generalization of the method of effective potentials. We show that as the amplitude of the laser field increases, the structure of the energy terms becomes more and more complicated. Moreover, the number of the energy terms increases. We also calculated analytically the shift of the radiation frequency of OERQ caused by the laser field. As the amplitude of the laser field increases, so does the shift. For a known amplitude of the laser field, by measuring the relative shift of the radiation frequency it should be possible to determine experimentally the distance of the orbital plane of the electron from the nucleus of the smaller nuclear charge.

**Key words:** two Coulomb centers; one-electron Rydberg quasimolecules; high-frequency laser field; method of effective potentials

## 1. INTRODUCTION

The problem of electron terms in the field of two stationary Coulomb centers (TCC) of charges  $Z$  and  $Z'$  separated by a distance  $R$  is one of the most fundamental problems in quantum mechanics. It presents fascinating atomic physics: the terms can have crossings and quasicrossings. On the one hand, the well-known Neumann-Wigner general theorem on the impossibility of crossing of terms of the same symmetry [1] is invalidated for the TCC problem of  $Z' \neq Z$  (see, e.g., paper [2]) – so, the terms can cross. On the other hand, when two potential wells (each corresponding to separated  $Z$ - and  $Z'$ -centers) have states  $\Psi$  and  $\Psi'$  of the same energies  $E = E'$ , of the same magnetic quantum numbers  $m = m'$ , and of the same radial elliptical quantum numbers  $k = k'$ , a quasicrossing of the terms occurs [3-5]. Then the electron has a much larger probability of tunneling from one well to the other (what constitutes charge exchange) compared to the absence of the quasicrossing.

In plasma spectroscopy, a quasicrossing of the TCC terms, by facilitating charge exchange, can result in local dips in the spectral line profile emitted by a  $Z$ -ion from a plasma consisting of both  $Z$ - and  $Z'$ -ions – see, e.g., theoretical and experimental papers [6-11]. In particular, this allows determining rates of charge exchange between multicharged ions – the reference data almost inaccessible by other experimental methods [11].

When the charges  $Z$  and  $Z'$  approach each other and share the only one electron that they have, they form a

quasimolecule. When the electron is in a highly-excited state, it is a one-electron Rydberg quasimolecule (OERQ). There are extensive analytical studies of the OERQ by the methods of classical mechanics (which are appropriate for Rydberg states) [12-20] – see also review [21] and book [22], Chapter 3. In particular, the following papers were devoted to studies of the OERQ in various external fields: namely, in a static magnetic field [15], in a static electric field [16, 17, 19], and in a laser field [20]. Specifically, in our previous paper [20] we analyzed the situation where the laser frequency was much smaller than the highest frequency of the unperturbed system.

In the present paper we consider the situation where the OERQ is subjected to a linearly-polarized laser field whose frequency is much greater than the highest frequency of the unperturbed system. For obtaining analytical results we use a generalization of the method of effective potentials [23] (see also book [24], Appendix A). We show that as the amplitude of the laser field increases, the structure of the energy terms becomes more and more complicated, and the number of the energy terms increases.

We also calculate analytically the shift of the radiation frequency of OERQ caused by the laser field. The results show that for a known amplitude of the laser field, by measuring the relative shift of the radiation frequency it should be possible to determine experimentally the distance of the orbital plane of the electron from the nucleus of the smaller nuclear charge.

## 2. ANALYTICAL RESULTS

We consider a TCC system with the charge  $Z$  placed at the origin, and the  $Oz$  axis is directed at the charge  $Z'$ , which is at  $z = R$ . Atomic units ( $\hbar = e = m_e = 1$ ) are used throughout this article. The system is subjected to a high-frequency linearly-polarized laser field of amplitude  $F$  and frequency  $\omega$ , the laser field being directed along the internuclear axis. The Hamiltonian for the electron in this configuration is

$$H = H_0 + zF\cos\omega t, H_0 = \frac{1}{2}(p_z^2 + p_\rho^2 + \frac{p_\phi^2}{\rho^2}) - \frac{Z}{r} - \frac{Z'}{r'} \quad (1)$$

where  $r = (\rho^2 + z^2)^{1/2}$  is the distance from the electron to the nucleus  $Z$ ,  $r' = (\rho^2 + (R - z)^2)^{1/2}$  is the distance from the electron to the nucleus  $Z'$ , and  $(\rho, \phi, z)$  are the cylindrical coordinates positioned in such a way that the nuclei  $Z$  and  $Z'$  are on the  $z$ -axis at  $z = 0$  and  $z = R$  accordingly. Due to  $\phi$ -symmetry,  $\phi$  is a cyclic coordinate and its corresponding momentum is conserved:

$$p_\phi = \rho^2 \frac{d\phi}{dt} = L \quad (2)$$

For the systems in a high-frequency field, whose frequency is much greater than the highest frequency of the unperturbed system, it is appropriate to use the formalism of effective potentials [25, 26, 23]. As a result, the Hamiltonian acquires a time-independent term. The zeroth-order effective potential,

$$U_0 = \frac{1}{4\omega^2} [V, [V, H_0]] = \frac{F^2}{4\omega^2} \quad (3)$$

where  $V = zF$  and  $[P, Q]$  are the Poisson brackets, is a coordinate-independent energy shift that does not affect the dynamics of the system. The first non-vanishing effect on the dynamics of the system originates from the first-order effective potential

$$U_1 = \frac{1}{4\omega^4} [[V, H_0], [[V, H_0], H_0]] = \frac{F^2}{4\omega^4} (Z \frac{\rho^2 - 2z^2}{(\rho^2 + z^2)^{5/2}} + Z' \frac{\rho^2 - 2(R - z)^2}{(\rho^2 + (R - z)^2)^{5/2}}) \quad (4)$$

and the Hamiltonian of the electron in the high-frequency field is

$$H = \frac{1}{2}(p_z^2 + p_p^2) + \frac{L^2}{2\rho^2} - \frac{Z}{\sqrt{\rho^2 + z^2}} - \frac{Z'}{\sqrt{\rho^2 + (R-z)^2}} + U_1 \quad (5)$$

where  $U_1$  is given by (4). The electron is considered to be in a circular state\*. Therefore  $p_z = p_p = 0$ , and thus, its energy can be represented in the form

$$E = \frac{L^2}{2\rho^2} - \frac{Z}{\sqrt{\rho^2 + z^2}} - \frac{Z'}{\sqrt{\rho^2 + (R-z)^2}} + \frac{F^2}{4\omega^4} \left( Z \frac{\rho^2 - 2z^2}{(\rho^2 + z^2)^{5/2}} + Z' \frac{\rho^2 - 2(R-z)^2}{(\rho^2 + (R-z)^2)^{5/2}} \right) \quad (6)$$

Using the scaled quantities

$$w = \frac{z}{R}, v = \frac{\rho}{R}, \varepsilon = -\frac{R}{Z}E, b = \frac{Z'}{Z}, \ell = \frac{L}{\sqrt{ZR}}, r = \frac{Z}{L^2}R, \theta = \frac{F}{\omega^2 R} \quad (7)$$

we obtain the scaled energy of the electron

$$\varepsilon = \frac{1}{\sqrt{w^2 + v^2}} + \frac{b}{\sqrt{(1-w)^2 + v^2}} - \frac{\ell^2}{2v^2} + \frac{2w^2 - v^2}{(w^2 + v^2)^{5/2}} \frac{\theta^2}{4} + b \frac{2(1-w)^2 - v^2}{((1-w)^2 + v^2)^{5/2}} \frac{\theta^2}{4} \quad (8)$$

We can seek the equilibrium points in the  $(w, v)$ -plane by finding the two partial derivatives of  $\varepsilon$  with respect to  $w$  and  $v$  and setting them to zero. The second equation gives the equilibrium value of the scaled angular momentum

$$\ell = v^2 \sqrt{\frac{(w^2 + v^2)^2 + 3(w^2 - \frac{v^2}{4})\theta^2}{(w^2 + v^2)^{7/2}} + b \frac{((1-w)^2 + v^2)^2 + 3((1-w)^2 - \frac{v^2}{4})\theta^2}{((1-w)^2 + v^2)^{7/2}}} \quad (9)$$

and the first equation gives the equilibrium value of  $v$

$$w \frac{(w^2 + v^2)^2 + \frac{3}{2}(w^2 - \frac{3}{2}v^2)\theta^2}{(w^2 + v^2)^{7/2}} = b(1-w) \frac{((1-w)^2 + v^2)^2 + \frac{3}{2}((1-w)^2 - \frac{3}{2}v^2)\theta^2}{((1-w)^2 + v^2)^{7/2}} \quad (10)$$

Fig. 1 shows the equilibrium plot in the  $(w, v)$ -plane for  $b = 3$  and  $\theta = 0.1$ . We see that, in addition to the properties described in [13], there is a multivalued range in the neighborhood of  $w = 0$  and  $w = 1$ , which increases as  $\theta$  increases.

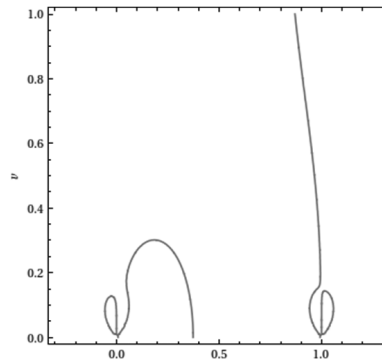


Fig. 1. Equilibrium plot in the  $(w, v)$ -plane for  $b = 3$  and  $\theta = 0.1$ .

If we scale the internuclear distance  $R$  as  $r = (Z/L^2)R$ , and given  $\varepsilon = -(R/Z)E$  from (7), then  $E = -(Z/L)^2 \varepsilon_1$ , where  $\varepsilon_1 = \varepsilon/r$  is the scaled energy whose scaling includes only  $Z$  and  $L$ . From (7),  $\ell^2 = L^2/(ZR)$ , so this yields  $r = 1/\ell^2$ , with  $\ell$

\* Circular states of atomic and molecular systems are an important subject. They have been extensively studied both theoretically and experimentally for several reasons (see, e.g., [12-15, 17, 27-40] and references therein): (a) they have long radiative lifetimes and highly anisotropic collision cross sections, thereby enabling experiments on inhibited spontaneous emission and cold Rydberg gases, (b) these classical states correspond to quantal coherent states, objects of fundamental importance, (c) a classical description of these states is the primary term in the quantal method based on the  $1/n$ -expansion, and (d) they can be used in developing atom chips.

taken from (9), giving us the expression for  $r(w, v, b, \theta)$ . Then we substitute the value of  $\ell$  from (9) into (8) and obtain  $\varepsilon(w, v, b, \theta)$ , which we divide by  $r = 1/\ell^2$ , with  $\ell$  again taken from (9), obtaining  $\varepsilon_1(w, v, b, \theta)$ , whose explicit form is

$$\begin{aligned} \varepsilon_1 = & v^4 \left( \frac{(w^2 + v^2)^2 (w^2 + \frac{v^2}{2}) + (w^4 - \frac{5}{2} w^2 v^2 + \frac{v^4}{4}) \frac{\theta^2}{2}}{(w^2 + v^2)^{7/2}} + \right. \\ & + b \frac{((1-w)^2 + v^2)^2 ((1-w)^2 + \frac{v^2}{2}) + ((1-w)^4 - \frac{5}{2} (1-w)^2 v^2 + \frac{v^4}{4}) \frac{\theta^2}{2}}{((1-w)^2 + v^2)^{7/2}} \Big) \times \\ & \times \left( \frac{(w^2 + v^2)^2 + 3(w^2 - \frac{v^2}{4}) \theta^2}{(w^2 + v^2)^{7/2}} + b \frac{((1-w)^2 + v^2)^2 + 3((1-w)^2 - \frac{v^2}{4}) \theta^2}{((1-w)^2 + v^2)^{7/2}} \right) \end{aligned} \tag{11}$$

Then, solving (10) numerically for  $v$  and substituting it into (11) and into  $r(w, v, b, \theta)$ , we obtain, for the given value of  $b$  and  $\theta$ , the parametric dependence  $\varepsilon_1(r)$  representing the scaled energy terms, with the parameter  $w$  running over the allowed range determined by (10). The asymptote  $w_3$ , corresponding to  $v \rightarrow \infty$ , is the same as in the case of  $\theta = 0$ , and is equal to  $b/(b + 1)$ , and other limits on  $w$  can be determined numerically.

Figures 2 and 3 show the scaled energy terms for the values of the scaled amplitude of the laser field for  $\theta = 0.01$  and  $\theta = 0.1$ , respectively, in comparison to the unperturbed energy terms for  $\theta = 0$ . It is seen that for small values of  $\theta$ , the lower term is the first affected, and the terms take on a more complicated form as  $\theta$  further increases. We plot  $-\varepsilon_1$  on the vertical axis for it to have the same sign as  $E$ .

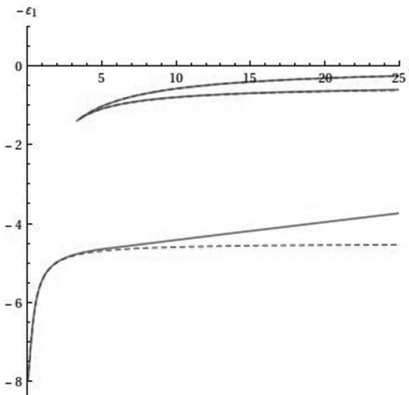


Fig. 2. The plot of the scaled energy terms  $-\varepsilon_1(r)$  (with  $r$  on the horizontal axis and  $-\varepsilon_1$  on the vertical) for the scaled amplitude of the laser field  $\theta = 0.01$ , with  $b = 3$ , shown in blue, solid curves, against the terms for  $\theta = 0$ , with  $b = 3$ , shown in red, dashed curves.

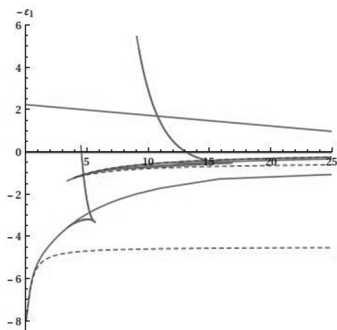


Fig. 3. The plot of the scaled energy terms  $-\varepsilon_1(r)$  (with  $r$  on the horizontal axis and  $-\varepsilon_1$  on the vertical) for the scaled amplitude of the laser field  $\theta = 0.1$ , with  $b = 3$ , shown in blue, solid curves, against the terms for  $\theta = 0$ , with  $b = 3$ , shown in red, dashed curves.

It is seen that as the scaled amplitude  $\theta$  of the laser field increases, the scaled energy terms  $-\varepsilon_1(r)$  become more and more complicated. In particular, at some ranges of  $\theta$ , the number of the scaled energy terms increases from 3 (which was the case for  $\theta = 0$ ) to 4 or even 5.

At this point it might be useful to clarify the relation between the classical energy terms  $-\varepsilon_1(r)$  and the energy  $E$ . The former is a scaled quantity related to the energy as specified above in the 1<sup>st</sup> line after Eq. (10):  $E = -(Z/L)^2 \varepsilon_1$ . The projection  $L$  of the angular momentum on the internuclear axis is a *continuous* variable. The energy  $E$  depends on both  $\varepsilon_1$  and  $L$ . Therefore, while the scaled quantity  $\varepsilon_1$  takes a *discrete* set of values, the energy  $E$  takes a *continuous* set of values (as it should be in classical physics).

We also studied the shift of the radiation frequency caused by a high-frequency linearly-polarized laser field. The angular momentum of the electron can be expressed as

$$L = \rho^2 \frac{d\phi}{dt} = \Omega \rho^2 \quad (12)$$

where  $\Omega$  is the frequency of the motion of the electron. Using the scaled quantities from (7), we have

$$\Omega = \sqrt{\frac{Z}{R^3}} \tilde{\Omega}, \tilde{\Omega} = \frac{\ell}{v^2} \quad (13)$$

where  $\tilde{\Omega}$  with the wave above denotes the scaled frequency. The relative shift of the frequency is determined by

$$\delta = \frac{\Omega - \Omega_0}{\Omega_0} = \frac{\Omega}{\Omega_0} - 1 = \frac{\tilde{\Omega}}{\tilde{\Omega}_0} - 1 = \frac{\ell}{\ell_0} \frac{v_0^2}{v^2} - 1 \quad (14)$$

where the subscript index “0” refers to the default case ( $\theta = 0$ ) and the value of  $v$  is taken to be the equilibrium value (determined by (10)).

Figures 4 and 5 show the plot of the relative shift of the frequency for the ratio of the nuclear charges  $b = 3$  and the values of  $\theta = 0.01$  and  $\theta = 0.1$ , respectively. As we can see, the shift increases when  $\theta$  increases, and it is the smallest around the point  $w = w_3 = b/(b + 1)$ .

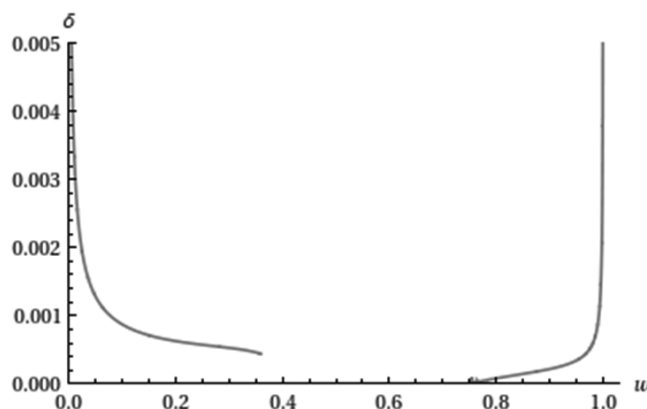
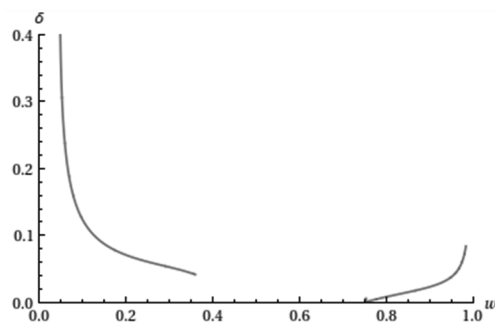


Fig. 4. The relative blue shift of the radiation frequency of the electron versus its scaled axial coordinate for  $b = 3$  and  $\theta = 0.01$ .



**Fig. 5.** The relative blue shift of the radiation frequency of the electron versus its scaled axial coordinate for  $b = 3$  and  $\theta = 0.1$ .

Thus, for a known amplitude of the laser field, by measuring the relative shift of the radiation frequency it should be possible to determine experimentally the distance of the orbital plane of the electron from the nucleus of the smaller nuclear charge.

### 3. CONCLUSIONS

We considered the situation where one-electron Rydberg quasimolecules (OERQ) are subjected to a linearly-polarized laser field whose frequency is much greater than the highest frequency of the unperturbed system. For obtaining analytical results we used a generalization of the method of effective potentials. We found out that as the amplitude of the laser field increases, the structure of the energy terms becomes more and more complicated. Moreover, the number of the energy terms increases.

We also calculated analytically the shift of the radiation frequency of OERQ caused by the laser field. As the amplitude of the laser field increases, so does the shift. For a known amplitude of the laser field, by measuring the relative shift of the radiation frequency it should be possible to determine experimentally the distance of the orbital plane of the electron from the nucleus of the smaller nuclear charge.

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