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OPTICS AND SPECTROSCOPY. LASER PHYSICS

On the Effect of Interatomic Interactions on the Efficiency of the Rydberg Blockade

Yu. V. Dumin^{1,2*}

¹Astronomical Institute, Moscow State University, Moscow, 119234 Russia ²Space Research Institute, Russian Academy of Sciences, Moscow, 117997 Russia Received August 6, 2021; revised September 11, 2021; accepted September 13, 2021

Abstract—The process of the Rydberg blockade, which is currently regarded as one of the most promising approaches to quantum computation, can become significantly less effective due to interatomic interactions that shift the excitation frequency of initially blocked particles to the region of resonance with external laser radiation. In this article, we develop an analytical theory of this effect based on a self-consistent model of two Rydberg atoms acting on each other with their dipole electric fields and experiencing a Stark splitting of energy levels under the action of these fields. It follows from our calculations that the typical interatomic distances at which the unblocking effect due to "parasitic" resonances begins to occur are in rough agreement with earlier qualitative estimates; however, the number of such resonances can be much larger than in recent numerical simulations of some particular systems. Therefore, the possibility of breaking the Rydberg blockade at small interatomic distances should be carefully taken into account in any future applications to quantum computing.

Keywords: Rydberg atoms, Rydberg blockade, Stark effect

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INTRODUCTION

The Rydberg blockade is the impossibility of simultaneous excitation of several neighboring atoms by narrow-band laser radiation into the same Rydberg state, because after one atom is excited, it perturbs the energy levels of neighboring atoms with its electric field and thus takes them out of resonance with the external radiation. This phenomenon was theoretically proposed as a tool for quantum information processing in the very early 2000s [1]; a few years later its practical feasibility was confirmed experimentally [2, 3]. The Rydberg blockade is considered as one of the most promising approaches to creating a quantum computer [4, 5]. In particular, this effect underlies the recent numerous experiments on quantum simulation in ordered arrays of Rydberg atoms [6, 7].

Unfortunately, as we qualitatively showed in [8], there can be a serious problem of the undesirable ("parasitic") excitation of a neighboring atom (within the standard blockade radius) to highly perturbed energy levels with neighboring quantum numbers (see, e.g., Fig. 1 in [8] as well as Fig. 3 in this article). In other words, the Rydberg blockade is broken when the atoms are too densely arranged. Later, detailed calculations of this effect were made in [9]. In particular, both the positions and excitation rates of the main resonances corresponding to the quasimolecular states of two rubidium atoms that were asymptotically in 100*s* states were found, and the corresponding consequences for the reliability of quantum computation protocols were discussed.

However, as will be shown below, the total number of parasitic resonances leading to simultaneous excitation of two neighboring atoms can be much larger than that depicted in Fig. 1 from [9]. Apparently, this is due to the limited accuracy of the calculations in the standard quantum chemistry programs used in [9]. On the other hand, the analytical approach we present below allows us to identify all possible parasitic resonances (although it does not allow us to calculate their parameters in detail). In principle, all of them can break the Rydberg blockade, although with different efficiencies; therefore their detrimental effect on the reliability of quantum computation protocols should be taken into account.

We note that the possibility of breaking the Rydberg blockade due to the influence of neighboring quantum states was briefly discussed in [12]. How-

^{*}E-mail: dumin@sai.msu.ru;dumin@yahoo.com



Fig. 1. The scheme of the dipole electric field created by one of the atoms at the location of the second atom.

ever, it was entirely based on numerical calculations and the authors noted their poor convergence.

1. DESCRIPTION OF THE MODEL

Generally speaking, the Rydberg blockade effect can be calculated in various ways. From our point of view, one of the most effective methods is to consider the Stark splitting of the energy levels of each atom by the dipole electric field of its partner already excited into the Rydberg state. This approach was used in our previous works [8, 10] for the case of "sequential" excitation of two Rydberg atoms, i.e., on a time scale much larger than the inverse Rabi frequency. This situation is interesting primarily for experiments with ultracold Rydberg plasmas [11]. In this article, we aim at generalizing this approach to the case of "simultaneous" excitation, which is directly related to quantum computations. (Let us emphasize that exactly the same simultaneous excitation was numerically simulated in [9].)

An important feature of the Stark splitting, which should be taken into account in the following analysis, is the significant nonuniformity of the dipole electric field created by the Rydberg atoms at characteristic distances comparable to their sizes. As far as we know, the only special consideration of the Stark effect in a strongly nonuniform field was made by Bekenstein and Krieger in 1970 [13] based on a very specific version of the quasi-classical approximation. In [10], we made similar calculations without using the quasi-classical approximation and found some corrections to the Bekenstein–Krieger formula. (However, these are quite insignificant for the states with small orbital momenta, which are usually used in the Rydberg blockade experiments.) Therefore, the general expression for the Stark energy level splitting of the hydrogen-like atom can be written in the following form (here and below, the atomic system of units is used) [10]:

$$\delta E_n \equiv E_n + \frac{1}{2n^2} \approx g_1 \mathcal{E}_z - g_2 \mathcal{E}_z^2 + g_3 \frac{d\mathcal{E}_z}{dz}, \quad (1)$$

where the coefficients g_i are defined as follows

$$g_1 = \frac{3}{2}n\Delta, \qquad (2a)$$

$$g_2 = \frac{n^4}{16} \left[17n^2 - 3\Delta^2 - 9m^2 + 19 \right], \qquad (2b)$$

$$g_3 = \frac{n^2}{4} [5\Delta^2 + 2n_1 n_2 + (n-m)(m+1) + 1].$$
(2c)

Here, \mathcal{E} is the electric field strength, n is the principal quantum number, n_1 and n_2 are the parabolic quantum numbers traditionally used in solving the Schrödinger equation in parabolic coordinates ($n_{1,2} \ge 0$), $\Delta = n_1 - n_2$ is the so-called electric quantum number, and m is the *absolute value* of the magnetic quantum number (i.e., we follow the Bethe–Salpeter notation system [14]).

It is known that the above quantum numbers are related to each other by the following formula:

$$n = n_1 + n_2 + m + 1,$$

so that the following inequalities are satisfied:

$$\begin{split} m &\ge 0, \\ n &\ge m+1, \\ 0 &\le n_1, n_2 &\le n-m-1. \end{split}$$

In addition, it is easy to see that $g_{2,3} \ge 0$.

The first two terms in the right-hand side of formula (1) represent well-known expressions for the Stark effect of the first and second orders in a uniform field [14–16], while the third term describes the contribution of the electric field nonuniformity (see its derivation in [10]; as mentioned above, it differs somewhat from the quasi-classical result [13]). In principle, one could also include here higher-order corrections with respect to the electric field amplitude (e.g., explicit expressions for terms up to the fourth order were given in [17], and the general algorithm for calculating the terms of arbitrary order was described in [18]). However, as can be shown by a closer analysis, such corrections are quite insignificant for our purposes.¹⁾ We note, however, that the gradient term included in formula (1) is actually important.

¹⁾This fact is not surprising; we will be interested in perturbations of the order of the energy level difference of states with neighboring values of the principal quantum number, while higher order corrections become significant only on the scale of the ionization energy of the atom.

Let us now consider the possibility of the *simul-taneous* excitation of two closely spaced atoms into Rydberg states under the action of narrow-band laser radiation. Each of these atoms has a dipole electric field²⁾ and thereby perturbs its partner, producing a corresponding Stark splitting of its energy levels. Therefore, we will further analyze the system of two atoms mutually perturbing each other.

Let the first atom be located at the origin of the coordinate system (X, Y, Z) and the position of the second atom be given by the radius vector \mathbf{r}_0 directed along the axis X, as shown in Fig. 1. The electric dipole moment $\mathbf{d}_e^{(1)}$ of the first atom is tilted at an angle θ to this axis. Next, we place the origin of the coordinate system (x, y, z) at the center of the second atom and orient the axis z along the direction of the electric field at that point $\mathcal{E}(\mathbf{r}_0)$; so that, by definition, the only nonzero component is \mathcal{E}_z .

The electric potential of the dipole is obviously given by the formula

$$\Phi = \frac{\mathbf{d}_e \cdot \mathbf{r}_0}{r_0^3},$$

and the corresponding electric field strength and its gradient can be written as follows:³⁾

$$\mathcal{E}_z = \frac{d_e}{r_0^3} (1 + 3\cos^2\theta)^{1/2},$$
$$\frac{d\mathcal{E}_z}{dz} = -\frac{3d_e}{r_0^4} \frac{3 + 5\cos^2\theta}{1 + 3\cos^2\theta} \cos\theta.$$

For simplicity, we will carry out the following detailed analysis only for the cases of parallel and antiparallel orientation of the dipoles. Then the above formulas will be reduced to

$$\mathcal{E}_z = \frac{2d_e}{r_0^3}, \quad \frac{d\mathcal{E}_z}{dz} = -\epsilon_\theta \frac{6d_e}{r_0^4},\tag{3}$$

where $\epsilon_{\theta} = 1$ for $\theta = 0$ and -1 for $\theta = \pi$.

Since we consider the electric field \mathcal{E} in the classical approximation, its source is the average value of the operator of the electric dipole moment $\hat{\mathbf{d}}_e = -\hat{\mathbf{r}}_e$, where \mathbf{r}_e is the radius-vector of the electron inside the atom. The corresponding matrix element is well known, e.g., from the calculations of the first-order Stark effect [14–16]. Therefore, for the first atom (i.e.,



Fig. 2. The possible mutual orientations of electric dipoles and corresponding physical parameters as well as directions of coordinate axes.

the field source in Fig. 1), the projection of the dipole moment on the axis *X* will be as follows

$$d_{eX}^{(1)} = -\langle \hat{X}_e \rangle = -\frac{3}{2}n\Delta^{(1)},$$

and the absolute value, respectively,

$$d_e^{(1)} = \frac{3}{2}n|\Delta^{(1)}| \tag{4}$$

and similarly for the second atom.⁴⁾ It is obvious that

$$\epsilon_{\theta} = -\operatorname{sgn}(\Delta).$$

Four possible orientations of the dipoles $\mathbf{d}_{e}^{(1)}$ and $\mathbf{d}_{e}^{(2)}$ together with their corresponding physical parameters are shown in Fig. 2.

In the following consideration, it will be convenient to measure the Stark shifts of energy not with respect to the same unperturbed level n but with respect to the unperturbed state with the principal quantum number \bar{n} whose blockade is studied (and which will be denoted by a dash above). In other words, the energy of the perturbed level can be written, on the one hand, as $-1/(2n^2) + \delta E_n$, and, on the other hand, as $-1/(2\bar{n}^2) + \delta E_{\bar{n}}$. By combining these two expressions, we obtain:

$$\delta E_{\bar{n}} \equiv rac{1}{2 \bar{n}^2} - rac{1}{2 n^2} + \delta E_n.$$

In addition, we will normalize all lengths and energies by the characteristic size and energy of this state \bar{n} and the corresponding quantities will be denoted by tildes:

$$r_0 = \bar{n}^2 \tilde{r}, \quad E = \tilde{E}/(2\bar{n}^2).$$
 (5)

²⁾For simplicity, we consider the case of strongly degenerate (hydrogen-like) energy levels when a constant dipole moment can exist.

³⁾The fact that these formulas involve only one angle θ should not be surprising; we consider the situation only in the electric field symmetry plane given by the direction of the first dipole $\mathbf{d}_{e}^{(1)}$ and the direction to the second atom \mathbf{r}_{0} .

⁴⁾The parabolic quantum numbers used here will generally have different signs if the axes X and z are oriented opposite to each other. However, since only the absolute values of the dipole moments will be used hereafter, we can assume that they are defined in the same coordinate system.

(For brevity, the normalized radius-vector of the atom is written without the superscript '0'.)

Finally, by combining formulas (1), (2a), (3), (4), and (5), we obtain the energy level shifts of the second atom caused by the first atom:

$$\begin{split} \delta \tilde{E}_{\bar{n}}^{(2)} &= 1 - \frac{\bar{n}^2}{n^{(2)2}} + 9 \Bigg[\frac{1}{\tilde{r}^3} \frac{n^{(1)} n^{(2)} |\Delta^{(1)}| \Delta^{(2)}}{\bar{n}^4} \\ &- \frac{2}{\tilde{r}^6} \frac{g_2^{(2)} n^{(1)2} \Delta^{(1)2}}{\bar{n}^{10}} + \frac{2}{\tilde{r}^4} \frac{g_3^{(2)} n^{(1)} \Delta^{(1)}}{\bar{n}^6} \Bigg], \end{split}$$

where the superscripts in parentheses denote the number of the atom (parentheses are used to avoid confusion with power exponents). A similar formula with interchanged superscripts will obviously give the energy shifts in the first atom.

In order to avoid cumbersome calculations, we further consider in detail only two "symmetric" types of excitations in such a two-atom system:⁵⁾

(a)
$$|n_1, n_2, m\rangle^{(1)} |n_1, n_2, m\rangle^{(2)}$$
,
i.e., $n \equiv n^{(1)} = n^{(2)}$, $\Delta \equiv \Delta^{(1)} = \Delta^{(2)}$,
(b) $|n_1, n_2, m\rangle^{(1)} |n_2, n_1, m\rangle^{(2)}$,
i.e., $n \equiv n^{(1)} = n^{(2)}$, $\Delta \equiv \Delta^{(1)} = -\Delta^{(2)}$.

In other words, both atoms are excited either to the same states or to states with interchanged parabolic quantum numbers. It is easy to show that case (a) corresponds to a parallel orientation of the dipoles, while case (b) corresponds to an antiparallel orientation (toward or opposite to each other).

Therefore, in any case the energy shifts in both atoms will be the same:

$$\delta \tilde{E}_{\bar{n}} \equiv \delta \tilde{E}_{\bar{n}}^{(1)} = \delta \tilde{E}_{\bar{n}}^{(2)}$$

$$= 1 - \frac{\bar{n}^2}{n^2} + 9 \left[\frac{1}{\tilde{r}^3} \frac{n^2 \Delta^2}{\bar{n}^4} \epsilon \text{sgn}(\Delta) - \frac{2}{\tilde{r}^6} \frac{g_2 n^2 \Delta^2}{\bar{n}^{10}} + \frac{2}{\tilde{r}^4} \frac{g_3 n \Delta}{\bar{n}^6} \right], \quad (6)$$

where $\epsilon = 1$ and -1 for parallel and antiparallel orientations of the dipoles, respectively. We note that the first and second terms in the square brackets are due to the Stark effect of first and second order in a uniform field, while the third term corresponds to the first order perturbation from the electric field gradient.

2. CALCULATION RESULTS

The formulas presented in the previous section can be applied to calculate the Rydberg blockade of any chosen state. As an example, let us consider the blockade of the 100s state of hydrogen-like atoms $(\bar{n} = 100, m = 0)$. The behavior of several Starkshifted energy levels in the vicinity of this state as a function of the interatomic distance calculated using formula (6) is shown in Fig. 3. For clarity, the width of the laser emission band depicted by the dashed line along the horizontal axis was taken as sufficiently large, $\Delta \tilde{E} = 5 \times 10^{-3}$ (in real experiments, it is one or two orders of magnitude less). In addition, in order not to clutter the figure, we do not depict here the entire set of split levels; only the curves corresponding to the five parabolic quantum numbers are shown for each of the values of the principal quantum number.

Let us comment briefly on the relative role of the different terms of formula (6) in the behavior of the resulting curves. As follows from a more detailed analysis, the main contribution stems from the well-known first-order Stark effect in a uniform field. The first order gradient term is also quite important; moreover, it qualitatively changes the behavior of the energy curves at small \tilde{r} in the case of antiparallel orientation of the dipoles. The second-order Stark effect in a uniform field is usually insignificant. It becomes more or less noticeable only at small distances in the case of the antiparallel orientation of the dipoles, when first-order perturbations by the field amplitude and its gradient largely compensate each other.

Figure 3 clearly shows the physical mechanism of the Rydberg blockade: when two atoms approach each other, the energy levels of the excited state \bar{n} (which at large distances were degenerate and were in resonance with the laser radiation) experience larger and larger perturbations and finally leave the excitation band. This is the reason why the blockage effect develops.

However, when the interatomic distance is further reduced, strongly perturbed energy levels from the states with neighboring values of the principal quantum number $(n \neq \bar{n})$ begin to enter the laser emission band, and thus the possibility of excitation of two perturbed atoms is restored. Therefore, the Rydberg blockade is broken at some set of "resonance" radii.⁶⁾

It is difficult to determine the exact total number of levels with neighboring values of the principal quantum number n involved in breaking the Rydberg

⁵⁾In order to avoid confusion, we emphasize that the situation when the principal quantum numbers of the two atoms are equal is not "preferable" from the physical point of view. In this case, it is only an assumption to simplify the calculations.

⁶⁾We did not show the effect of "anticrossing" the energy curves in Fig. 3, because the key question of our consideration is the point of their intersection with a narrow excitation band along the horizontal axis; the behavior of the curves outside this region is of no interest.



Fig. 3. The Stark shifts of energy levels $\delta \tilde{E}_{\bar{n}}$ in each of the interacting atoms as a function of the distance \tilde{r} between them for parallel ($\epsilon = 1$) and antiparallel ($\epsilon = -1$) orientations of the dipoles. For $n = \bar{n} = 100$, the curves are shown for the parabolic quantum numbers $n_1 = 0, 25, 50, 75, 99$; for n = 99, for the parabolic quantum numbers $n_1 = 0, 25, 49, 73, 98$; for n = 98, for $n_1 = 0, 25, 49, 73, 97$; for n = 101, for $n_1 = 0, 25, 50, 75, 100$; and for n = 102, for $n_1 = 0, 25, 50, 75, 101$. The shaded horizontal regions near the horizontal axis show the laser excitation band, $\Delta \tilde{E} = 5 \times 10^{-3}$.

blockade, because it is limited by the scope of applicability of the perturbation theory. Roughly speaking, it is given by the condition that the energy shift should be less than its absolute value in the unperturbed state:

$$|E_n - E_{\bar{n}}| \lesssim |E_n|; \tag{7}$$

it follows that the condition is $n \leq \sqrt{2n}$. On the other hand, it is easy to see that for all lower states, $n < \bar{n}$, the condition of applicability of perturbation theory (7) is automatically satisfied.

The most interesting and important result arising from our analysis is that the total number of resonance radii turns out to be much greater than the number of "quasimolecular" resonances obtained by computer simulations using quantum chemistry methods [9]. For the particular case of 100*s*-states, this fact is illustrated in Fig. 4: in our calculations, the number of resonances $N_{\rm res}$ in the range of interatomic distances $r_0 \ge 2 \ \mu {\rm m}$ turns out to be an order of magnitude greater than in the quasi-molecular approximation in the case of antiparallel orientation of the dipoles and two orders of magnitude greater in the case of parallel orientation.⁷

Unfortunately, it is almost impossible to compare the specific positions of the resonances obtained in our calculations with those found in [9], because the rubidium atoms simulated in it have significant quantum defects. (This probably explains the appearance

⁷⁾Note that the presented pattern of resonances is little sensitive to the width of the excitation band $\Delta \tilde{E}$. Indeed, as can be seen in Fig. 3, the development of the Rydberg blockade depends significantly on $\Delta \tilde{E}$, i.e., the narrower this band, the larger the size of the blocked region. However, the radii in which the blockade is broken are almost independent of $\Delta \tilde{E}$, because the corresponding energy curves cross the horizontal axis almost vertically.



Fig. 4. The scheme of quasi-molecular resonances for the 100*s* state of rubidium [9] (middle band) in comparison with all possible resonances of the same state of hydrogen-like atoms (upper band is for the parallel orientation of dipoles and lower band is for the antiparallel orientation). Two quasi-molecular resonances presented in Fig. 1 from [9] but not listed in Table 1 of the same article are dotted.

of two resonances in rubidium at very large distances, approximately 6 μ m.) On the other hand, the huge difference in *total number* of resonances shown in Fig. 4 clearly cannot be related only to quantum defects. This discrepancy is most likely due to the limited accuracy of numerical calculations performed using standard quantum chemistry computer programs. It is also possible that a significant part of the resonances found in our calculations has a very low excitation efficiency, as a result of which they were not identified in the numerical simulations. (This is also indicated by the results of [12]; see, e.g., Fig. 2 there.)

CONCLUSIONS

1. We qualitatively predicted the possibility of breaking the Rydberg blockade at small interatomic distances due to the effect of strongly perturbed neighboring energy levels in [8, 10]. This was done for the case of "sequential excitation" of two atoms (i.e., on a time scale much larger than the inverse Rabi frequency), which is of interest mainly for the experiments with Rydberg plasmas [11]. In this work, a similar analytical approach based on the Stark shift of energy levels was developed for the case of "simultaneous" excitation, which is of main interest in quantum information problems [4], as well as in experiments on the detailed spatial study of the Rydberg blockade [19].

2. Numerical simulations of breaking the Rydberg blockade [9] carried out for the same purpose by other authors were based on the use of standard computational packages of quantum chemistry. They allowed only a few basic resonances to be calculated; most of them were not identified at all. In this sense, our analytical approach provides us with a more adequate picture of the complete set of parasitic resonances, although it does not allow us to obtain their exact parameters, because it is limited only to the hydrogenlike approximation and to the dipole nature of interaction.

3. The high density of resonances at small interatomic distances obtained in our calculations indicates that the effect of breaking the Rydberg blockade can play a large role and should be taken into account very carefully when creating any experimental setup designed to implement quantum calculations. Another, fundamentally different mechanism of breaking the Rydberg blockade was indicated earlier [20]. This is a specific superposition of excitations in a system of three (or more) atoms. However, such a mechanism seems less dangerous than that I am considering, since it requires the simultaneous approach of particles.

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CONFLICT OF INTEREST

The author declares that he has no conflicts of interest.

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