On the fidelity of quantum information processing with Rydberg atoms: Role of the non-molecular resonances

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Abstract. Rydberg blockade of ultracold atoms is considered now as one of the most promising tools for the implementation of quantum computing, but its fidelity can be substantially compromised by detrimental excitation of the neighbouring atoms. This phenomenon has been investigated recently in detail for the particular case of molecular resonances (i.e., resulting in the formation of quasi-bound states). However, as will be shown in the present paper, an even greater effect can come from the non-molecular resonances, which therefore should be taken into account very carefully.

PACS numbers: 32.60.+i, 32.80.Ee

Submitted to: \textit{J. Phys. B: At. Mol. Opt. Phys.}
1. Introduction

The phenomenon of Rydberg blockade – impossibility of simultaneous excitation of the nearby Rydberg atoms due to the shifts of their energy levels – was suggested as a tool for quantum information processing by Lukin, et al. in the early 2000’s [1]. A few years later, the practical feasibility of such a protocol was confirmed experimentally [2, 3], and now it is widely discussed in the context of quantum computing [4].

Unfortunately, as was qualitatively discussed in our paper [5], an important problem that can arise in this way is the possibility of detrimental excitation of a nearby atom (within the standard blockade radius) caused by the strongly-perturbed energy levels with neighbouring values of the quantum numbers (see, for example, figure 1 in paper [5] as well as figure 2 below). Later, this effect was studied in detail in the work [6]. In particular, the calculation was performed both for the positions and excitation rates of the major resonances forming the quasi-molecular states of two Rb atoms that are asymptotically in 100s states, and the corresponding consequences for the fidelity of the quantum computing were discussed.

However, as will be shown below, the total number of resonances breaking the Rydberg blockade (i.e., resulting in the simultaneous excitation of two nearby Rydberg atoms) should be much greater than in figure 1 of the above-cited paper. The most of these resonances are non-molecular, i.e., do not necessarily result in the formation of binding potentials. Anyway, they can break the Rydberg blockade and, therefore, their detrimental effect on the fidelity of quantum processing protocols must be kept in mind.

2. Basic formulae

From our point of view, an efficient method for treating the Rydberg blockade can be based on the idea of Stark splitting of the atomic energy levels by the external electric field by a neighbouring already-excited atom. This approach was pursued in our previous papers [5, 7] for the case of “sequential” excitation of two Rydberg atoms (i.e., at the time scales much greater than the inverse Rabi frequency), which is actually most interesting for the experiments with ultracold Rydberg plasmas [8]. Here we are going to generalize this approach to the case of “simultaneous” excitation, which is immediately relevant to the quantum-computing applications. (Let us emphasize that the same simultaneous excitation was considered in the work [6].)

An important aspect of the Stark splitting that must be taken into account in such analysis is the substantially nonuniform character of the dipolar electric field produced by one Rydberg atom at the characteristic scale of another atom. As far as we know, the only special treatment of Stark effect in the nonuniform field was done in 1970 by Bekenstein and Krieger [9], who used a specific kind of the quasi-classical approximation. We performed such calculation beyond this approximation in paper [7] and found some corrections to the Bekenstein–Krieger formula. (However, these corrections are quite small for the low-angular-momentum states, which are commonly employed in the
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So, the general expression for Stark splitting of hydrogen-like energy levels can be written in the atomic units as:

\[ \delta E_n = E_n + \frac{1}{2n^2} = g_1 \mathcal{E}_z - g_2 \mathcal{E}_z^2 + g_3 \frac{d\mathcal{E}_z}{dz}, \tag{1} \]

where the coefficients \( g_i \) are

\[ g_1 = \frac{3}{2} n\Delta, \tag{2a} \]

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

\[ g_3 = \frac{n^2}{4} \left[ 5\Delta^2 + 2n_1n_2 + (n - m)(m + 1) + 1 \right]. \tag{2c} \]

Here, \( \mathcal{E} \) is the electric field intensity, \( n \) is the principal quantum number, \( n_1 \) and \( n_2 \) are the parabolic quantum numbers (\( n_{1,2} \geq 0 \)), \( \Delta = n_1 - n_2 \) is the so-called electric quantum number, and \( m \) is the absolute value of the magnetic quantum number (we accept here the Bethe–Salpeter designations \[10\]).

As is known, the above-mentioned quantum numbers are related to each other as:

\[ n = n_1 + n_2 + m + 1, \tag{3} \]

so that the following inequalities are satisfied:

\[ m \geq 0, \tag{4a} \]

\[ n \geq m + 1, \tag{4b} \]

\[ 0 \leq n_1, n_2 \leq n - m - 1. \tag{4c} \]

Besides, it is easy to see that \( g_{2,3} \geq 0 \).

The first two terms in the right-hand side of formula (1) represent the well-known expressions for the first- and second-order Stark effect in the uniform field \[10, 11, 12\], and the third term takes into account the electric-field nonuniformity (it was derived in our paper \[7\] and, as have been already mentioned above, slightly differs from the quasi-classical result by Bekenstein and Krieger \[9\]). In principle, it might be possible to include here also the higher-order corrections with respect to the electric field amplitude: for example, the explicit expression with terms up to the fourth order was obtained by Alliluev and Malkin \[13\], and a general algorithm for deriving the terms of arbitrary order was described by Silverstone \[14\]. However, as follows from the subsequent analysis, such higher-order corrections are completely negligible for our purposes. However, the gradient term will be really important.

Next, let us consider two simultaneously-excited Rydberg atoms. Each of them forms a dipolar electric field and, thereby, disturbs its partner, producing the corresponding Stark splitting of the energy levels. So, we shall analyze further the mutually-perturbed diatomic system, not assuming any specific kind of the interatomic interaction (such as formation of the binding potential).

\[ \dagger \text{This fact is not surprising: we are interested in the perturbation of energy levels on the order of difference between the states with neighboring values of the principal quantum number; while the higher-order corrections become significant on the scale of the binding energy.} \]
Let the first atom is located in the origin of the coordinate system $(\xi, \eta, \zeta)$, and the position of the second atom is given by the radius vector $r_0$ directed along $\xi$-axis (figure 1). The electric dipole moment $d_e$ of the first atom is tilted at the angle $\theta$ with respect to this axis. Next, the origin of the coordinate system $(x, y, z)$ is placed in the location of the second atom, and its $z$-axis is oriented along the direction of the electric field in this point $E(r_0)$; so, by definition, the only nonzero component is $E_z$.

The electric field potential is evidently given by formula:

$$\Phi = \frac{d_e \cdot r_0}{r_0^3}; \quad (5)$$

while the corresponding field intensity and its gradient can be written as

$$E_z = \frac{d_e}{r_0^3} (1 + 3 \cos^2 \theta)^{1/2}, \quad (6a)$$

$$\frac{dE_z}{dz} = -\frac{3d_e}{r_0^4} \frac{3 + 5 \cos^2 \theta}{1 + 3 \cos^2 \theta} \cos \theta. \quad (6b)$$

For the sake of simplicity, we shall consider below in detail only the cases of parallel and anti-parallel alignment of two dipoles. Then, the above formulae are reduced to

$$E_z = \frac{2d_e}{r_0^3}, \quad \frac{dE_z}{dz} = -\epsilon_\theta \frac{6d_e}{r_0^4}, \quad (7)$$

where $\epsilon_\theta = 1$ at $\theta = 0$, and $-1$ at $\theta = \pi$.

Since the electric field $E$ is considered here in the classical approximation, then its source should be the expectation value of the electric dipole operator $\hat{d}_e = -\hat{r}_e$, where $\hat{r}_e$ is the electron radius vector inside the atom. The corresponding matrix element is well known, e.g., from calculations of the first-order Stark effect \[ 10, 11, 12\]. So, for the first atom it will be equal to

$$d_e = -\langle \xi_e \rangle = -\frac{3}{2} n \Delta. \quad (8)$$
As can be easily seen,

$$\epsilon_\theta = -\text{sign}(\Delta).$$  \hfill (9)

For subsequent applications it will be convenient to measure the Stark energy shifts with respect to the energy of the unperturbed state $\bar{n}$ whose blockade is studied and which will be denoted by a bar:

$$\delta E_{\bar{n}} = \frac{1}{2\bar{n}^2} - \frac{1}{2n^2} + \delta E_n.$$  \hfill (10)

Besides, we shall normalize all lengths and energies to the characteristic size and energy of the above-mentioned state $\bar{n}$, and the resulting quantities will be marked by tildes:

$$r_0 = \bar{n}^2 \tilde{r}, \quad E = \tilde{E}/(2\bar{n}^2).$$  \hfill (11)

(For brevity, the scaled radius vector of the atom is written without subscript ‘0’.) Finally, combining the formulae (1) and (7)–(11), we get the shifts of energy levels in the second atom produced by the first atom:

$$\delta \tilde{E}_{\bar{n}}^{(2)} = 1 - \frac{\bar{n}^2}{n_{(2)2}} + 9 \left[ \frac{1}{\tilde{r}^3} \frac{n_{(1)2}^{(2)} |\Delta^{(1)}| \Delta^{(2)}}{\bar{n}^4} - \frac{2}{\tilde{r}^6} \frac{g_2 \bar{n}^2 \Delta^{(2)}}{n_{(2)2}^{(1)}} + \frac{2}{\tilde{r}^4} \frac{g_3 n_{(1)} \Delta^{(1)}}{\bar{n}^6} \right],$$  \hfill (12)

where superscripts in parentheses denote the number of the atom (to avoid its confusion with exponents). The same expression with interchanged superscripts will evidently give the energy shifts in the first atom.

To avoid cumbersome computations, we shall consider in detail only two “symmetric” types of excitation in this diatomic system:

(a) $|n_1, n_2, m_{(1)}^{(1)} \rangle |n_1, n_2, m_{(2)}^{(2)} \rangle$, i.e.,

$$n \equiv n_{(1)} = n_{(2)}, \quad \Delta \equiv \Delta^{(1)} = \Delta^{(2)},$$

(b) $|n_1, n_2, m_{(1)}^{(1)} \rangle |n_2, n_1, m_{(2)}^{(2)} \rangle$, i.e.,

$$n \equiv n_{(1)} = n_{(2)}, \quad \Delta \equiv \Delta^{(1)} = -\Delta^{(2)}.$$  

In other words, the atoms are excited either exactly to the same states or to the states with interchanged parabolic quantum numbers. As can be easily shown, case (a) corresponds to parallel orientation of the dipoles, while case (b) represents the anti-parallel orientation (either towards or away from each other).

So, the energy shifts in both atoms under the above assumptions will be the same and equal to

$$\delta \tilde{E}_{\bar{n}} \equiv \delta \tilde{E}_{\bar{n}}^{(1)} = \delta \tilde{E}_{\bar{n}}^{(2)} = 1 - \frac{\bar{n}^2}{n^2} + 9 \left[ \frac{1}{\tilde{r}^3} \frac{n^2 \Delta^2}{\bar{n}^4} \epsilon \text{sign}(\Delta) - \frac{2}{\tilde{r}^6} \frac{g_2 n^2 \Delta^2}{\bar{n}_{(2)2}^{(1)}} + \frac{2}{\tilde{r}^4} \frac{g_3 n \Delta}{\bar{n}^6} \right],$$  \hfill (13)

where $\epsilon = 1$ and $-1$ for parallel and anti-parallel orientation of the dipoles, respectively. Note that the first and second terms in the square brackets result from the first- and
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Figure 2. The Stark-split energy curves \( \delta \tilde{E}_n \) in each of the interacting atoms as function of distance \( \tilde{r} \) between them for parallel (\( \epsilon = 1 \)) and anti-parallel (\( \epsilon = -1 \)) orientation of the dipoles: \( n = \bar{n} = 100, n_1 = 0, 25, 50, 75, 99 \) (green curves), \( n = 99, n_1 = 0, 25, 49, 73, 98 \) (blue), \( n = 98, n_1 = 0, 25, 49, 73, 97 \) (cyan), \( n = 101, n_1 = 0, 25, 50, 75, 100 \) (red), and \( n = 102, n_1 = 0, 25, 50, 75, 101 \) (violet). The horizontal shaded (yellow) strip denotes the energy excitation band of laser irradiation, \( \Delta \tilde{E} = 5 \cdot 10^{-3} \).

second-order Stark effect in the uniform field, while the third term comes from the first-order perturbation by the electric-field gradient.

3. Results of calculations

Now, the mathematical formalism outlined in the previous section can be applied to the problem of Rydberg blockade. Let us consider, for example, the blockade of 100s state of the hydrogen-like atoms (i.e., \( \bar{n} = 100 \) and \( m = 0 \)). A sample of the Stark-split energy curves in the vicinity of this state as function of the interatomic distance, calculated by formula (13), are presented in figure 2. For better visibility, the excitation band of laser irradiation, shown by a shaded strip along the horizontal axis, was taken here quite large \( \Delta \tilde{E} = 5 \cdot 10^{-3} \) (it is usually one or two orders of magnitude less in the real experiments).

It is interesting to discuss briefly a significance of the various terms in formula (13). As follow from the detailed analysis, the dominant contribution comes from the first-
order uniform-field Stark effect. The gradient term is always important and, moreover, qualitatively changes the behavior of energy curves at small \( \tilde{r} \) in the case of anti-parallel dipoles. The second-order uniform-field Stark effect is usually quite insignificant. It becomes noticeable only at small distances in the case of anti-parallel dipoles, when the first-order perturbations by the uniform field and its gradient compensate each other to a large extent.

As can be seen in figure 2 when two atoms approach each other, the energy levels of the basic manifold \( \tilde{n} \), which have been asymptotically degenerate and resonant with laser radiation at large separations, experience an increasing perturbation and eventually leave the bandwidth of the exciting irradiation. As a result, the Rydberg blockade develops. Nevertheless, when the interatomic separation decreases further, the strongly-perturbed energy levels from the neighboring Stark manifolds (with \( n \neq \tilde{n} \)) begin to enter the excitation band, thereby restoring the possibility of excitation. Hence, the Rydberg blockade should be broken at a set of the “resonant” radii, where the energy curves intersect the horizontal axis. (The effect of avoided crossings of the energy levels was not plotted in this figure just because we are interested only in the points of intersection of the energetic curves with the horizontal axis, while accurate identification of the corresponding states is of no importance in this context.)

The total number of the Stark-split manifolds participating in breaking the Rydberg blockade cannot be specified exactly and is actually limited by the scope of applicability of the perturbation theory. This is roughly given by the condition that the energy shift should be less than the absolute value of the unperturbed energy:

\[
|E_n - E_{\tilde{n}}| \lesssim |E_n|,
\]

implying that \( n \lesssim \sqrt{2} \tilde{n} \) for the upper-lying energy levels. On the other hand, it can be easily seen that the perturbation theory is applicable to all the low-lying levels, because the criterion (14) is always satisfied at \( n < \tilde{n} \).

The most important fact following from our analysis is that the total number of
resonant radii turns out to be much greater than in the “quasi-molecular” approach [6]. This is illustrated for the particular case of 100s states in figure 3: the number of resonances $N_{\text{res}}$ at the distance $r_0 \geq 2 \mu\text{m}$ is an order of magnitude greater than in the quasi-molecular approximation for the case of anti-parallel orientation of the dipoles, and by two orders of magnitude greater for the case of parallel orientation.

4. Conclusion

Unfortunately, it is impossible to compare the particular positions of the resonances found in our calculations and in paper [6] because of the considerable quantum defects in Rb, which was analyzed in the last-cited work. However, the striking disagreement apparent in figure 3 can be hardly explained just by the quantum defects. So, we should conclude that the quasi-molecular approach takes into account only a small fraction of all the resonances breaking the Rydberg blockade.

In other words, formation of the bound (quasi-molecular) states is not a necessary prerequisite for violating the Rydberg blockade. The blockade will be broken each time when two nearby atoms are excited simultaneously, irrelevant of the particular type of interaction between them. So, we believe that the detrimental influence of the non-molecular resonances – which was overlooked before – should be taken into account very carefully in any future applications of Rydberg blockade in the quantum computing and other quantum-optics experiments.

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§ Let us emphasize that the patterns of the detrimental resonances are almost insensitive to the bandwidth $\Delta E$ of the exciting irradiation. Really, as can be seen in figure 2 onset of the Rydberg blockade strongly depends on $\Delta E$: the narrower is the width, the greater is the radius of the blockade. However, the radii of the blockade breaking are approximately insensitive to $\Delta E$ because the corresponding energy curves intersect the horizontal axis almost vertically.
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