Antiblockade in Rydberg excitation of an ultracold lattice gas

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It is shown that the two-step excitation scheme typically used to create an ultracold Rydberg gas can be described with an effective two-level rate equation, greatly reducing the complexity of the optical Bloch equations. This allows us to solve the many-body problem of interacting cold atoms with a Monte Carlo technique. Our results reproduce the Rydberg blockade effect. However, we demonstrate that an Autler-Townes double peak structure in the two-step excitation scheme, which occurs for moderate pulse lengths as used in the experiment, can give rise to an antiblockade effect. It is observable in a lattice gas with regularly spaced atoms. Since the antiblockade effect is robust against a large number of lattice defects it should be experimentally realizable with an optical lattice created by CO2 lasers.

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The prediction of a blockade effect in the Rydberg excitation of ultracold gases due to long-range interaction [1] has sparked several experiments which have demonstrated the blockade effect under different circumstances. In [2] Rydberg excitation to n = 30–80 was achieved by a single photon of a pulsed UV laser from the 5s Rb ground state. In [3, 4] a two-step scheme is used where a strong pump laser drives the 5s-5p transition of Rb atoms while a tunable second laser excites from 5p to a Rydberg nl state. In all three cases a suppression of the excitation has been observed as a function of increasing laser intensity or/and density of the gas, i.e., as a function of increasing effective interaction between the atoms which shifts the energy levels out of resonance with the laser light.

However, the two different excitation schemes (single UV pulse and two-step excitation) have a very different nature and may lead to dramatic differences in the blockade behavior. In fact, as we will show below, the two-step scheme (see Fig. 1) may even induce an antiblockade effect due to the form of the single Rydberg atom population \( P_e(t, \Delta) \) as a function of the detuning \( \Delta \) from resonant excitation. This antiblockade effect will only be uncovered by a structured ultracold gas, e.g., a lattice gas with atoms regularly placed on a 3-dimensional lattice. Yet, the condition on the regular arrangement is not too strict: even with 20% lattice defects the antiblockade is still visible.

The key to the antiblockade effect is the excitation dynamics of the final Rydberg state \( nl \) in a three-level system with the transition between level \( |g\rangle \) (ground state) and level \( |m\rangle \) (intermediate state) driven by a strong optical transition with Rabi frequency \( \Omega \gg \omega \), the Rabi frequency which drives the transition between level \( |m\rangle \) and the Rydberg level \( |e\rangle \), see Fig. 1.

Experimentally, the intermediate level \( |m\rangle \) decays with a rate \( \Gamma \gg \omega \), large compared to the upper Rabi frequency \( \omega \). Under the conditions \( \Omega \gg \omega \) and \( \Gamma \gg \omega \) and a sufficiently long laser pulse the optical Bloch equations [5] for transitions in this three-level system reduce to a rate equation for a two-level system which contains the upper state \( |e\rangle \) and an effective lower state \( |g\rangle \). The reason for this simplification lies in the strong decay \( \Gamma \) which damps out coherences relatively fast and ultimately allows for the rate description which directly gives the probability \( P_e \) for an atom to be in the upper Rydberg state \( |e\rangle \).

\[
P_e(t, \Delta) = P_\infty(\Delta) \left( 1 - \exp \left[ -\frac{\gamma(\Delta) t}{P_\infty(\Delta)} \right] \right), \quad (1)
\]

where \( P_\infty = P_e(t \rightarrow \infty, \Delta) \) is the steady-state occupation of \( |e\rangle \) and \( \gamma(\Delta) \) is the rate of populating \( |e\rangle \) for short times \( t \).

Typical Rydberg populations as a function of detuning \( \Delta \) are shown in the left part of Fig. 2 which also demonstrates that, for the given Rabi frequencies and decay rate, a pulse length of \( t \geq 0.5 \mu s \) is enough to make the description with a rate equation applicable.

For atoms embedded in an environment of other atoms, e.g., a gas, the Rydberg level \( |e\rangle \) of a ground state atom is shifted by \( \delta \) due to the (weak) interaction with Rydberg atoms in the neighborhood.
is given by the sum \( \delta \) running over all Rydberg atoms existing at this time and genetically close state \( n \) is easily generalized to this case where now \( A \) pair of Rydberg atoms in state \( \Gamma = 2\pi \) our Rydberg levels by appropriate scaling in induced dipole coupling \( V \), action induced shift \( \delta \) atom resonance and the detuning \( \Delta \) is given by an inter-
matic (\( \Gamma_0 \), \( H_0 \)). For a specific quantum number \( n_0 \) we may define \( \mu = \frac{\mu_{ab}}{\mu_{bb}} R_0^3 \). We have taken its value and the value for \( \delta_0 \) (\( \mu_0^2(n_0) = 843800 \) a.u., \( \delta_0 = -0.0378 \) a.u. for \( n_0 = 48 \)) from [7] and adapted to our Rydberg levels by appropriate scaling in \( n \) [8],

\[
\mu^2(n) = \mu^2(n_0) \left( \frac{n^*}{n_0} \right)^4
\]

\[
\delta_0(n) = \delta_0(n_0) \left( \frac{n^*}{n^*_0} \right)^3,
\]

where \( n^* = n - \eta \) includes the appropriate quantum defect, for the \( ns \) states of Rb \( \eta = 3.13 \). Furthermore, we excite the repulsive branch of Eq. (2) which finally defines the shift \( \delta \) resulting from a single excited atom.

However, we are interested in the many-atom case of an ultracold gas, where the laser is tuned to the single atom resonance and the detuning \( \Delta \) is given by an interaction induced shift \( \delta \) of \( |e \rangle \). Since the shift is additive it is easily generalized to this case where now \( \delta_i \) for atom \( i \) is given by the sum \( \delta_i = \sum_j \delta(|r_i - r_j|) \), with the index \( j \) running over all Rydberg atoms existing at this time and \( \delta(R) \) given in Eq. (2). Number and location of Rydberg atoms at a time can be determined by solving the rate equation Eq. (1) for each atom with a Monte Carlo technique. The result is shown in Fig. 3, namely a decreasing fraction of Rydberg atoms with increasing level shift \( \delta \), here realized through increasing excitation \( n \). This is the typical blockade effect, observed in [2, 3].

We use the simple picture as formulated in [7] for Rb. A pair of Rydberg atoms in state \( ab \) at distance \( R \) experiences a shift \( \delta(R) \) of its electronic energy due to an induced dipole coupling \( V(R) = \mu_{ad}/R^3 \) to an energetically close state \( a'b' \). It is given by the eigenvalues

\[
\delta(R) = \frac{1}{2} (\delta_0 \pm (\delta_0^2 + 4V^2)^{\frac{1}{2}})
\]

of the two-state matrix Hamiltonian with elements \( H_{11} = 0 \), \( H_{22} = \delta_0 \), \( H_{12} = H_{21} = V(R) \), where \( \delta_0 \) is the asymptotic (\( R \to \infty \)) difference between the energies of the two pairs.

The relevant neighboring pair to an excited pair \( ab = nsns \), corresponding to two atoms in state \( |e \rangle \) of Fig. 1 is \( a'b' = (n-1)p_3/n_3/2 \). For a specific quantum number \( n_0 \) we may define \( \mu^2(n_0) = \mu_{ns}(n_0-1)n^0_{ns}n_{ns} \). We have taken its value and the value for \( \delta_0 \) (\( \mu^2(n_0) = 843800 \) a.u., \( \delta_0 = -0.0378 \) a.u. for \( n_0 = 48 \)) from [7] and adapted to our Rydberg levels by appropriate scaling in \( n \) [8],

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![FIG. 2: The population \( P_e \) of the Rydberg state \( |e \rangle \) in the three-level system of Fig. 1 according to the rate equation Eq. (1) (solid) and the optical Bloch equation (dashed) for laser pulse lengths of 0.1\( \mu \)s (a,e), 0.3\( \mu \)s (b,f), 0.5\( \mu \)s (c,g), 1.0\( \mu \)s (d,h). The parameters are \( \Omega = 2\pi \cdot 4 \) MHz, \( \omega = 2\pi \cdot 0.2 \) MHz, \( \Gamma = 2\pi \cdot 6 \) MHz for the left set (a-d) and \( \Omega = 2\pi \cdot 22.1 \) MHz, \( \omega = 2\pi \cdot 0.8 \) MHz, \( \Gamma = 2\pi \cdot 6 \) MHz for the right set (e-h).](#)

![FIG. 3: The fraction of excited Rydberg atoms \( f_e \) as a function of increasing excitation \( n \) and for different laser pulse length of 1\( \mu \)s (solid), 2\( \mu \)s (dotted), and 5\( \mu \)s (dashed). The density of the ultracold gas is \( \rho = 8 \times 10^9 \) cm\(^{-3} \), and the parameters are close to those of the experiment [3], \( \Omega = 2\pi \cdot 22.1 \) MHz, \( \omega = 2\pi \cdot 0.8 \) MHz, \( \Gamma = 2\pi \cdot 6 \) MHz.](#)

The parameters of Fig. 3 are close to those of the experiment [3]. However, for those parameters, the single atom excitation probability \( P_e(t, \Delta) \) differs qualitatively from the one shown on the left part of Fig. 2. It has a double peak structure due to an Autler-Townes splitting induced by the strong driving \( \Omega \), as can be seen on the right part of Fig. 2 with maxima at \( \Delta = \pm \Delta_m \). Due to the wide distribution of mutual atomic distances \( R \) in a gas the characteristic double peak excitation profile with

![Diagram showing population intervals](#)
a peak separation of $\Delta \approx \Omega$ does not have an effect on $f_e$ as shown in Fig. 3.

To make the effect of the double peak in the excitation profile visible, one needs a situation where the distribution of mutual atomic distances of the atom is restricted. This can be achieved, e.g., with a lattice gas, i.e., with atoms regularly spaced on a three dimensional lattice. The fraction of excited atoms on a simple cubic lattice with a lattice constant $a = 5\mu m$ is shown in Fig. 4. One can see the known Rydberg blockade but on top of it an antiblockade effect, i.e., an enhanced fraction of excited atoms for certain values of $n$. A closer look at Fig. 4 reveals that there are even satellite peaks next to the main peaks. This pattern of peaks is easily understood if one analyzes the geometry of the atoms placed on a cubic lattice (see Fig. 5). The shift in the Rydberg level of a ground state atom is dominantly induced by a Rydberg atom located on the next lattice site at a distance $a$. Hence we may write for the shift $\delta(a_i; n_i)$ that is shown in Fig. 4. As experimentally also done, by changing the laser intensity we can reach different $n$ keeping the Rabi frequency $\omega$ constant. Clearly, with different $n_i$ the (optimum) shift $\Delta_m$ can be achieved by a Rydberg atom at a different distance $a_i$, so that in general

$$\delta(a_i; n_i) = \Delta_m.$$  \hspace{1cm} (4)

The obvious candidates in a cubic lattice apart from the nearest neighbor at $a_1 = a$ are the neighbors along the diagonal at $a_2 = \sqrt{2}a$ and along the cubic diagonal at $a_3 = \sqrt{3}a$. If one calculates the corresponding quantum numbers $n_i$ from Eq. (4), one would predict \( \{n_1, n_2, n_3 \} = \{65, 78, 87\} \). This differs at most by one quantum of excitation from the numerical values of Fig. 3, which are $\{65, 77, 86\}$ for the shortest pulse length (1$\mu$s). Of course, for the longer pulses the interaction is stronger with a considerable amount of excited atoms. This background of excited atoms leads to a general shift of the peaks towards lower $n$.

In addition to the main peaks described, satellite peaks can be seen to the left of the main peaks. They correspond to a situation where two Rydberg atoms on each side of a central atom contribute half to the shift of the excited level of the central atom, see Fig. 5. Of course, for this enhancement less interaction is necessary than in the nearest neighbor enhancement. Therefore these satellite peaks appear at smaller $n$, to the left of the main peaks. Since double the number of Rydberg atoms is necessary, the satellite peaks are roughly only half the height of the main peaks in the linear regime of low concentration of Rydberg atoms (short laser pulse, solid curve in Fig. 3).

A lattice with characteristic spacing of $a \sim 5\mu m$ can be produced by CO$_2$ lasers, but probably can only be filled leaving a significant amount of lattice defects. Therefore, we have simulated a situation with 20% random lattice defects. The defects hardly diminish the contrast of the antiblockade effect in the fraction of excited atoms (Fig. 4(b)), since the antiblockade relies dominantly on contributions from pairs of close neighbors: If a neighbor is missing due to a lattice defect this does neither lead to antiblockade nor to blockade and therefore does not spoil $f_e$. The large tolerance with respect to lattice defects makes an experimental verification of the antiblockade feasible, depending on the appropriate

![FIG. 4: The fraction of excited Rydberg atoms $f_e$ as a function of increasing excitation $n$ for atoms regularly arranged in a 3-dimensional simple cubic lattice with lattice constant $5\mu m$ ($\rho = 8 \cdot 10^9$ cm$^{-3}$) with parameters as in Fig. 3 (a) perfect filling of lattice sites, (b) 20% lattice defects (i.e., empty lattice sites).](image)

![FIG. 5: Arrangement of Rydberg and ground state atoms leading to antiblockade peaks and satellite peaks.](image)
combination of Rabi frequencies, pulse length and decay rate $\Gamma$. The corresponding “phase diagram” with the boundary between the single peak (blockade) and double peak (antiblockade) domain is shown in Fig. 6. The rate equation Eq. (1) reveals that for large $\Omega \gg \omega$, the exponential is suppressed and the steady state probability $P_\infty$ dominates, giving rise to a single peak structure. On the other hand, for small $\gamma t / P_\infty$ the Rydberg population is governed by $\gamma$ which gives rise to a double peak structure. The rate $\gamma$ as well as $P_\infty$ take a relatively simple form in the limit $\omega \ll \Gamma \ll \Omega$, namely

$$\gamma = \frac{\Gamma \omega^2 / \Omega^2}{2(1 - 4\Delta^2 / \Omega^2)^2}, \quad P_\infty = \frac{1}{1 + 8\Delta^2 / \Omega^2}. \quad (5)$$

The coefficients Eq. (5) reveal a universal condition for the transition from the double to the single peak structure of $P_c(t, \Delta)$, defined by $\partial^2 P_c(t, \Delta) / \partial \Delta^2 |_{\Delta=0} = 0$ which can be written as

$$g_0 = 2 \ln(1 + g_0) \quad (6)$$

with $g_0 = \Gamma t \omega^2 / \Omega^2$. Equation (6) is easily solved by iteration to give $g_0 = 2.513$. Hence, for $\Omega \gg \omega$ we expect a linear line $\omega = \alpha \Omega$ separating the two regimes, where $\alpha^2 = g_0 / (t \Gamma)$ which is indeed the case (dashed lines in Fig. 6). Note also, that Eq. (5) clearly demonstrates the transient character of the double peak structure which vanishes for long laser pulses. Yet, experimentally accessible parameters, e.g., in [13], realize exactly the transient regime and therefore provide the conditions to see the antiblockade.

To summarize, we have derived a rate equation for the population of Rydberg states in ultracold gases by a two-step excitation process. The rate describes very well the structure of the Rydberg excitation in a single atom when compared to the exact solution of the Bloch equations including a non-trivial transient Autler-Townes splitting in the Rydberg population for certain parameters.

The validity of the rate equation has allowed us to formulate the many-body excitation dynamics of Rydberg states in an ultracold gas without a mean-field approximation and for a realistic number of atoms as a random Monte Carlo process. We can reproduce the observed Rydberg blockade effect observed previously and also its effect on the atom counting statistics as measured in [14], but in addition we have identified an antiblockade effect due to the Autler-Townes splitted Rydberg population. We predict that this antiblockade effect can be seen in an experiment with a gas trapped in an optical lattice created by a CO₂ laser since the antiblockade effect is robust even against a large number of lattice defects. In the (realistic) limit of a very small upper Rabi frequency $\omega$ we could show that the formation of the double or single peak structure in the Rydberg population is determined by a universal parameter which allows a simple navigation in parameter space consisting of the two Rabi frequencies, the decay rate $\Gamma$ of the intermediate level and the pulse length, to achieve the desired peak structure in the single-atom Rydberg excitation probability.

\[\text{References}\]

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