#### Journal of Atomic, Molecular, Condensate & Nano Physics

Vol. 6, No. 3, pp. 161–167, 2015 ISSN 2349-2716 (online); 2349-6088 (print) Published by RGN Publications



# Ultracold Rubidium Atoms Excited to Rydberg Levels

Research Article

D. Ciampini<sup>1,2,3</sup>, O. Morsch<sup>2</sup> and E. Arimondo<sup>1,2</sup>

- <sup>1</sup> Dipartimento di Fisica 'E. Fermi', Università di Pisa, Lgo. Pontecorvo 3, 56127 Pisa, Italy
- <sup>2</sup> INO-CNR, Dipartimento di Fisica 'E. Fermi', Università di Pisa, Italy
- <sup>3</sup> CNISM UdR, Dipartimento di Fisica 'E. Fermi', Università di Pisa, Italy

**Abstract.** Ultracold atomic gases excited to strongly interacting Rydberg states are a promising system for quantum simulations of many-body systems. The dipole blockade of Rydberg excitations is a hallmark of the strong interactions between atoms in these high-lying quantum states. We have measured the Rydberg excitation for rubidium ultracold atoms in magneto-optical traps and for Bose-Einstein condensates loaded into quasi one-dimensional traps. One of the consequences of the dipole blockade is the suppression of fluctuations in the counting statistics of Rydberg excitations. We have obtained experimental results on the dynamics and the counting statistics of Rydberg excitations of ultra-cold Rubidium atoms both on and off resonance, which exhibit sub- and super-Poissonian counting statistics, respectively. We have found strongly bimodal counting distributions in the off-resonant regime.

**Keywords.** Ultracold atoms; Rydberg collective excitations; Dipole blockade; Counting statistics; Sub- and super-Poissonian distributions

**PACS.** 67.85.-d; 42.50.Ct; 03.75.Kk; 32.80.Ee

**Received:** April 8, 2015 Accepted: September 26, 2015

Copyright © 2015 D. Ciampini, O. Morsch and E. Arimondo. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

#### 1. Introduction

The atomic excitation to Rydberg levels has been intensively studied since tunable laser sources provided to the experimentalists the perfect tool to reach a single highly excited atomic level. Rydberg atoms are known to exhibit long lifetimes, huge electric dipole

<sup>\*</sup>Corresponding author: ennio.arimondo@unipi.it

moments, strong and long-range interatomic interactions, offering various possibilities for their manipulation and interaction control. The ultracold gas technology has opened new perspectives in the investigation, and manipulation, of regimes with controlled strong atomic interactions. The merge of Rydberg physics with ultracold atomic technology has produced an intense activity, motivated by both fundamental scientific research and applications. Ultra-cold atoms excited to high-lying Rydberg states have been shown in recent years to be promising candidates for experimental implementations of quantum computation and quantum simulation. The strong and controllable interactions between Rydberg atoms imply that fast two-qubit quantum gates and quantum-simulations of many-body Hamiltonians can, in principle, be efficiently realized. Dipole blockade is the central ingredient for the above Rydberg interest. In this process, the resonant excitation of an atom to a Rydberg state is inhibited if another, already excited, atom is very close, i.e., less than the so-called blockade radius away.

We have recently investigated the Rydberg excitation of rubidium atoms in different operating conditions [1–3]. In those experiments the strong interactions between Rydberg atoms manifest themselves through a reduction or an increase of the fluctuations in the number of excitations, evidencing the occurrence of different regimes. We characterize our system through the full counting statistics of the excitation events, similarly to the methods used in condensed matter physics to unveil correlations in electronic transport processes. In the regime of off-resonant excitation, we find that the behaviour of the system depends strongly on the detuning and the sign of the interaction. When the excitation is performed in the dissipative regime, we observed strongly bimodal counting distributions, denoting the presence of processes as intermittency, or phase coexistence, or bistability.

## 2. Sketch of Rydberg Physics

Rydberg atoms are atoms excited to high energy states, described by the principal quantum number n. The large distance of the electron from the atomic core determines most of the physical properties of Rydberg atoms. For instance the radius of an excited atom scales like  $n^2$  and can hence become very large (for n = 100 about one  $\mu$ m) compared to the atom in a ground state (order of nm). Rydberg atoms can stay in the excited state for long periods of time, for n = 100 almost one millisecond effective lifetime. They are sensitive even to small electric fields via the Stark effect. Their advantage, for several experiments over neutral ground state atoms is the high polarizability that scales like  $n^7$ .

In our set-up a coherent two photon excitation produces a transfer from the 5S ground state to a Rydberg state via the intermediate 6P state. Rydberg atoms are sensitive to electric fields. The large separation between an electron and its positive core is responsible for the weak binding energy. Therefore, to ionize Rydberg atoms relatively weak electric fields are needed. This sensitivity is used in the detection system where excited atoms are ionized and then the positive core accelerated toward a detecting device.

Dipole-dipole interactions in Rydberg states are the result of the interactions between two atoms with permanent dipole moments. For a system consisting of two atoms A and B, separated by the distance  $\vec{R}$ , The amplitude of dipole-dipole interaction is typically written in the following form:  $V_{dd} = C_3/R^3$ , with the  $C_3$  coefficient determining the interaction amplitude. In absence of

permanent dipole moment the Rydberg atoms interact through van-der-Waals interactions to be written as  $V_{vdW} = C_6/R^6$ .

Dipole-dipole and van-der-Waals interactions between Rydberg atoms give rise to a phenomenon called dipole blockade. The blockade of excitation to the Rydberg state occurs when two atoms are in the same excited state and the strong dipole interaction between them causes a symmetrical energy shift. If the energy shift is larger than the  $\Omega$  Rabi frequency and the laser frequency resolution, the excitation laser frequency is out of resonance with the shifted state. For the collective excitation of large number of atoms, the blockade, i.e., the single atomic excitation, takes place within a volume with radius given by  $r_b = [C_6/(\hbar\Omega)]^{1/6}$ . The blockade regime corresponds to an atomic collective behaviour. The ground state of the system is described by  $|\Psi^{(N,0)}\rangle = |g_1,g_2,\ldots,g_N\rangle$ , and the collective state with one excitation in the arbitrary  $|e_i\rangle$  is given by:

$$|\Psi^{(N,1)}\rangle = \frac{1}{\sqrt{N}} \sum_{i=(1,N)} |g_1, g_2, \dots, e_i, \dots, g_N\rangle$$
 (2.1)

supposing the same phase for all atoms. In reality the atoms separated by more than one wavelength experience different phases of the excited light. This collective system of N atoms behaving as a single atom is called superatom.

## 3. Experimental Realization

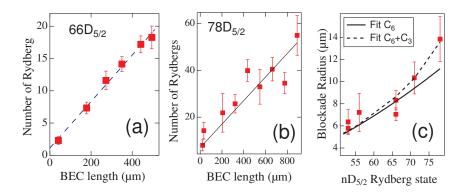
The Pisa experiment is based on the <sup>87</sup>Rb Rydberg excitation either in small clouds of ultracold atoms in a *Magneto-Optical Trap* (MOT) or in Bose-Einstein condensates. Bose-Einstein condensates of up to 10<sup>5</sup> <sup>87</sup>Rb atoms are created using a two-step evaporation protocol with a TOP-trap and a crossed optical dipole trap. Once condensation is reached, the condensate is then allowed to expand inside the (now one-dimensional) dipole trap, whose horizontal alignment has been optimized in order to avoid a centre-of-mass motion of the condensate along the trap direction.

The atoms are excited to the nS or  $nD_{5/2}$  Rydberg states using a two-photon excitation process with detuning 2GHz from the intermediate 6P level, with first step a blue laser at at  $420\,\mathrm{nm}$ , and the second step at  $1013\,\mathrm{nm}$ . For our experimental conditions, the two-photon Rabi frequency (for atoms in the highest laser intensity region) is  $\Omega/2\pi = 50 - 900\,\mathrm{kHz}$ . For the condensates in 1D geometry, the blue laser has a waist comparable to the second step laser. After the excitation laser pulse an electric field is applied for  $2\,\mu\mathrm{s}$  in order to field ionize the Rydberg atoms and to accelerate the resulting ions towards a channeltron. The overall detection efficiency is  $\eta \approx 40\%$ . The excitation-detection cycle is repeated 500 times. From the channeltron individual counts the histograms and full counting statistics (i.e., the moments of the counting distribution) are calculated.

#### 4. Results

#### 4.1 1D Dipole Blockade

We have demonstrated the realization of one-dimensional chains of collective Rydberg excitations in Rb Bose-Einstein condensates [2]. The highly elongated atomic clouds created by the method



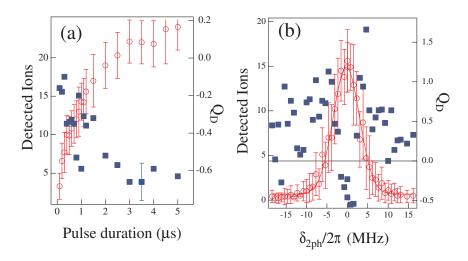
**Figure 1.** Rydberg excitation in an 1D expanded condensate. For  $66D_{5/2}$  state, in (a), and of  $78D_{5/2}$ , in (b), number of Rydberg excitations for an excitation pulse of  $1\mu$ s duration as a function of the condensate length. (c) Measured blockade radius  $r_b$  as a function of the principal quantum number n. The continuous and dashed lines are the theoretically predicted value assuming a total laser linewidth of  $300\,\mathrm{kHz}$ . A better quality fit is obtained by supposing that owing to the presence of a weak electric field the dipole-dipole interaction contains a weak  $C_3$  contribution.

described above are up to  $l = 1 \,\mathrm{mm}$  long, while their radial dimensions are on the order of few microns (radial dipole trap frequencies are around 100 Hz). Since the expected blockade radii for the Rydberg states between n = 50 and n = 80 used in our experiments range from  $5 \mu m$ to  $15\,\mu\mathrm{m}$ , this suggests that at most one Rydberg excitation fits radially into the condensate and that the total number of Rydberg excitations inside such a sample should depend on the l length. In order to test this picture we align the two Rydberg excitation lasers so as to be almost parallel to the dipole trap beam in which the condensate expands. After expanding the condensates, Rydberg atoms are created using pulses of up to  $1.5 \mu s$  duration (during which the condensate expansion is frozen and Penning and blackbody ionization were found to be negligible) and finally detected by field ionization. Figures 1(a) and 1(b) show typical results of such an experiment for excitation to the  $66D_{5/2}$  and  $78D_{5/2}$  states using a  $1\,\mu s$  excitation pulse, in which a linear increase of the number of Rydberg atoms with the length of the condensate is visible. This result agrees with the simple intuitive picture of super-atoms being stacked in a one-dimensional array of varying length. From the data in Figures 1(a) and 1(b) we can extract the mean distance between adjacent superatoms, assuming a close-packed filling of the one-dimensional atomic cloud. In Figure 1(c) the blockade radius measured in this way is shown for different Rydberg states, together with the value predicted for a pure van-der-Waals interaction following an  $n^{11/6}$  scaling. The measured values for the blockade radii of 5-15  $\mu$ m confirm our interpretation of an essentially one-dimensional chain of Rydberg excitations. For high-lying Rydberg states with n > 70 we find deviations from the theoretically expected scaling with n, which may be due to small electric background fields.

#### 4.2 Sub- and Super-Poissonian Counting Statistics

In the limit of vanishing interactions between the Rydberg atoms each excitation to a Rydberg state is independent of all the others, leading to a Poissonian excitation process. When the interactions are strong, however, the Rydberg excitation processes are highly correlated and

one expects sub-Poissonian counting statistics. All these regimes can be quantified through the Mandel Q-parameter defined as  $Q = \langle (\Delta N)^2 \rangle / \langle N \rangle - 1$ , where  $\langle (\Delta N)^2 \rangle$  and  $\langle N \rangle$  are the variance and mean, respectively, for the number N of Rydberg excitations [3]. The highly correlated character of the Rydberg excitations is demonstrated by analyzing the counting statistics of our experiments, which are strongly sub-Poissonian with observed Mandel  $Q_D = \eta Q$ -factors around -0.6, indicating actual Q-factors close to -1.



**Figure 2.** Excitation dynamics of the  $80\mathrm{S}_{1/2}$  state. In (a) detected ion number  $\langle N_D \rangle$  (red open circles) and  $Q_D$  Mandel parameter (blue squares) vs laser pulse duration for resonant excitation. In (b)  $\langle N_D \rangle$  and  $Q_D$  for a range of frequencies around the two-photon excitation frequency.

In Figure 2(a), an example of the time dependent resonant excitation dynamics to the  $80S_{1/2}$  state for cold atoms in a MOT is shown. Initially, a rapid increase in the average detected ion number is concomitant with a decrease in  $Q_D$ . After 0.5  $\mu$ s the growth in the detected ion number slows down while  $Q_D$  fluctuates around -0.6. In the long time limit the effect of a varying Rabi frequency across the sample, due to both the distribution of atoms and the spatial laser profile and line-width, cause a dephasing of the excitations, leading to a steady growth in the number of Rydberg atoms excited.

In the superatom model, the saturation of the excitation dynamics is related to the excitation of a maximum number of superatoms that fit into the volume of the MOT, leading also to a limiting minimum value for Q in a 'close-packing' geometry of blockade spheres. For  $\delta_{2ph}=0$  two-photon detuning  $Q_D$  is highly negative. Away from resonance, however, the counting statistics quickly becomes super-Poissonian with Q as large as 2-3. Our experimental observations in different operation regimes confirm that the measured positive value of the Mandel parameter off resonance is intrinsic to the excitation dynamics of the system. A theoretical model, presented in [3], and based on the Dicke coherent states reproduces well the salient features of the experiment.

#### 4.3 Full Counting Statistics

For off-resonant excitation, the behaviour of the system depends strongly on the detuning and the sign of the Rydberg-Rydberg interaction [1]. We have characterised the properties of our system through the full counting statistics of the excitation events, similarly to the methods recently used in condensed matter physics to unveil correlations in electronic transport processes. Evidence for the different excitation regimes is found in the histograms of the counting distributions shown in Figure 3(a) for 70S excitation. On resonance, the distribution is roughly Poissonian and it becomes sub-Poissonian in the fully blockaded regime. Away from resonance, Rydberg excitations of atoms at a distance R from an already excited atom satisfying  $C_6/R^6 = \hbar \delta_{2ph}$  are allowed and atomic pair excitations dominate the dynamics. (Notice that the  $C_6$  van-der-Waals coefficient is positive for S states.) This antiblockade resonant condition is the opposite of the blockade effect, where the interaction suppresses excitations. For positive detuning the distribution becomes bimodal with a dominant feature close to 0 excitations and another one centred around  $N_D = 12$ . Finally, for a detuning with opposite sign to that of the van-der-Waals interaction, neither single-particle nor pair excitations are resonant, leading to an overall suppression of the excitation.

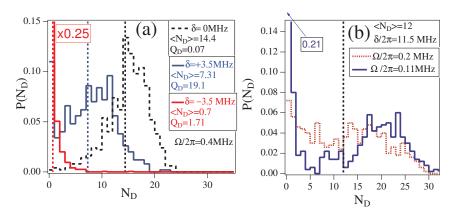


Figure 3.  $P(N_D)$  histograms vs  $N_D$ , at  $20\mu$ s short interaction time in (a) and dashed red line in (b), and a long interaction time blue solid line in (b). In (a) for positive detuning (blue line) the histogram exhibits a bimodal structure; on resonance (black dashed line) it has a single peak, and for negative detuning (red line)  $\langle N_D \rangle$  is considerably smaller. In (b) the probabilities histograms show the crossover to the dissipative regime with the counting distribution strongly bimodal at 950  $\mu$ s interaction time. As the excitation duration is increased, the Rabi frequency is decreased in order maintain  $\langle N_D \rangle$  constant.

A dissipative excitation regime is reached when several excitation-spontaneous emission cycles occur during the excitation pulse, interrupting the continuity of the excitation. In the dissipative regime the full counting distribution provides more insight into the properties of our system than an analysis of only the mean and the variance. We analyse the distribution central moments, up to the fourth order. The comparison of the counting distributions for different durations shown in Figure 3(b), where the Rabi frequency was adjusted in order to keep the  $\langle N_D \rangle$  mean value constant, suggests that in that regime the distributions become more strongly bimodal for longer excitation durations, indicating that dissipation favours the bimodality appearance.

#### 5. Conclusion

For resonant Rydberg excitation we have obtained clear evidence of sub-Poissonian counting statistics of Rydberg excitations in an ultra-cold atomic sample, which are a clear signature of many-body quantum correlations in such a system. We have characterized both the dynamics and the dependence on the detuning for the Q-parameter and reproduced the main features of both using a novel model based on the Dicke states. Through the full counting statistics we have analyzed resonant and off-resonant Rydberg excitations, both in the antiblockade regime and not, at short interaction times and in the long time dissipative regime. We have shown that the full counting statistics reveals characteristic features of the system that are not evident in the typically measured mean value or standard deviation. In future studies, the full counting statistics will be an important tool to examine in situ the spatial correlations between Rydberg excitations indirectly evidenced and for unveiling many-body effects in Rydberg excitations.

## **Acknowledgements**

The results here presented were produced by M. Bason, A. Chotia, P. Huillery, N. Malossi, J. Radogostowicz, S. Scotto, C. Simonelli, M. M. Valado and M. Viteau, past and present master students, Ph.D students and post-doc in our Laboratory, whose collaboration is gratefully acknowledged.

#### **Competing Interests**

The authors declare that they have no competing interests.

#### **Authors' Contributions**

All the authors contributed significantly in writing this article. The authors read and approved the final manuscript.

#### References

- [1] N. Malossi, M. M. Valado, S. Scotto, P. Huillery, P. Pillet, D. Ciampini, E. Arimondo and O. Morsch, Full counting statistics and phase diagram of a dissipative Rydberg gas, *Phys. Rev. Lett.* 113 (2014), 023006.
- M. Viteau, M. G. Bason, J. Radogostowicz, N. Malossi, D. Ciampini, O. Morsch and E. Arimondo, A'Rydberg excitations in Bose-Einstein condensates in quasi-one-dimensional potentials and optical lattices, *Phys. Rev. Lett.* **107** (2011), 060402.
- [3] M. Viteau, P. Huillery, M. G. Bason, N. Malossi, D. Ciampini, O. Morsch, E. Arimondo, D. Comparat and P. Pillet, Cooperative excitation and many-body interactions in a cold Rydberg gas, *Phys. Rev. Lett.* 109 (2012), 053002.