INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **39** (2006) L233–L239 doi:10.1088/0953-4075/39/11/L02

LETTER TO THE EDITOR

Strong interaction effects on the atom counting statistics of ultracold Rydberg gases

C Ates[1], T Pohl[2], T Pattard[1] and J M Rost[1]

[1] Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Straße 38, D-01187 Dresden, Germany
[2] ITAMP, Harvard-Smithsonian Center for Astrophysics, 60 Garden Street, Cambridge, MA 02138, USA

E-mail: cenap@mpipks-dresden.mpg.de

Received 24 March 2006, in final form 27 March 2006
Published 2 May 2006
Online at stacks.iop.org/JPhysB/39/L233

Abstract

Based on simple rate equations for the Rydberg excitation process, we are able to model microscopically the dynamics of Rydberg excitation in ensembles of a large number of ultracold atoms, which is beyond the capabilities of fully *ab initio* approaches. Our results for the distribution of Rydberg atom numbers are in good agreement with recent experimental data, confirming the quenching of the distribution caused by Rydberg–Rydberg interactions.

(Some figures in this article are in colour only in the electronic version)

Recently, the effect of 'dipole blockade' has been suggested as a way towards controlling Rydberg excitation in a cold atomic gas and using ensembles of Rydberg atoms for quantum information processing [1]. The essential idea behind this blockade effect is the fact that highly excited Rydberg atoms, due to their large dipole moments, are strongly interacting with each other even at distances typical for dilute gases trapped and accumulated in a standard magneto-optical trap (MOT). This interaction shifts the doubly excited state of two Rydberg atoms out of (two-photon) resonance with an excitation laser tuned to the single-atom excitation frequency, thereby suppressing the excitation of further Rydberg atoms in the neighbourhood of an excited atom. While the original proposal is based on a dipole–dipole interaction between the Rydberg states, the same mechanism of course also works for other types of interaction. In fact, the first experimental demonstrations of an interaction-induced suppression of Rydberg excitation have been achieved in systems with a van der Waals interaction rather than a dipolar one [2–4].

The experiments [2, 3] have demonstrated the excitation suppression by investigating the fraction of atoms that can be excited as a function of the laser intensity and of the density of ground-state atoms, respectively. The recent study of Cubel Liebisch *et al* [4], on the other hand, has investigated the statistics of the Rydberg excitation. In this work, the effect of interatomic interactions on the excitation process has been linked to the quenching of the distribution of Rydberg atom numbers in a sequence of repeated experiments with the same initial-state parameters. In the present letter, we focus on a theoretical modelling of this latter

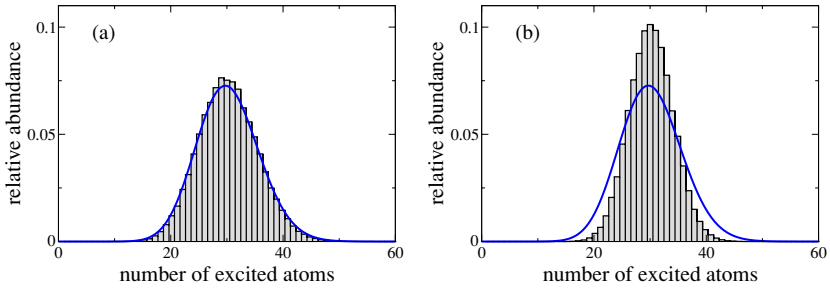

Figure 1. Calculated distribution of Rydberg atom numbers for the case of negligible interaction, $n = 45$ (a), compared to that of a system with strong interactions, $n = 85$ (b). The solid lines show Poissonian distributions with the same $\langle N_e \rangle$.

experiment, whereas first numerical simulations of the experiments [2, 3] have been reported in [2, 5].

In the experiment [4], Rb atoms have been collected in a MOT and excited to nD Rydberg states with a two-step excitation scheme via the 5P intermediate state. For each principal quantum number n, the intensity of the Rydberg excitation laser was adjusted to yield the same average number of about 30 Rydberg atoms after the excitation pulse. After the pulse was over, the number of Rydberg atoms was determined by field ionizing them and counting the number of electrons arriving at a micro-channel plate (MCP) detector. Each experiment was repeated up to 5000 times, resulting in a probability distribution for the Rydberg atom number, the atom counting statistics (figure 2 of [4]; cf our calculated results in figure 1). If the interaction between Rydberg atoms is negligible, i.e. for sufficiently low gas densities or principal quantum numbers, all Rydberg excitations in the gas are independent of each other. The resulting distribution is close to a Poissonian distribution, with the deviation from a Poissonian being due only to the finite Rydberg excitation probability (see below). At higher principal quantum numbers, on the other hand, the atom number distribution was seen to become highly sub-Poissonian and the quenching of the distribution was ascribed to the interatomic interactions in the gas which modify the dynamics of the excitation process.

The deviation of the atom counting statistics from a Poissonian distribution can be quantified by the so-called Mandel Q-parameter,

$$Q = \frac{\langle N_e^2 \rangle - \langle N_e \rangle^2}{\langle N_e \rangle} - 1, \tag{1}$$

where N_e is the number of Rydberg atoms and $\langle\,\rangle$ denotes the average over the probability distribution as usual. For a Poisson distribution, $Q = 0$. In general, not much can be said about the value of Q in the present context without explicitly calculating (or measuring) the full counting statistics. However, two limiting cases can be treated exactly. First, for a pure Fock (number) state where $\langle N_e^2 \rangle = \langle N_e \rangle^2$, $Q_F = -1$. In the present context of Rydberg excitation of an atomic gas, such a state can never be reached exactly. However, it includes the artificial limiting case of a 'full blockade' due to infinitely strong interaction, where one atom is excited with probability 1, and excitation of this atom completely suppresses the excitation of any other atom. (In the case of a finite probability P_{noex} that no Rydberg atom at all is excited by the laser pulse, the limit of infinitely strong interaction is given by $Q_\infty = P_{noex} - 1$.) In the other limit of negligible interaction between the atoms, the dynamics of each atom is independent of that of the other atoms. Assuming that the excitation laser covers the whole sample and acts on each atom in the same way, the excitation probability $P_e = \langle N_e \rangle / N_A$ is the same for all

atoms. In this case, $Q_0 = -P_e$. Hence, as stated above, in the case of non-interacting atoms the deviation of the atom counting statistics from a Poisson distribution is simply determined by the excitation probability P_e, i.e. the finite ratio of the number N_e of excited atoms to the number N_A of atoms in the gas cloud. Moreover, as is the case in the opposite limit of strong interaction, the value of Q does not depend on the time evolution of the excitation process, but just on the final number of excited atoms.

In the intermediate range of finite interaction strength, no exact statements about the value of Q can be made from the elementary considerations above. It seems reasonable to assume that in this regime a transition must take place from the $(Q = 0)$-limit to the $(Q = -1)$-limit as the interaction strength is increased. However, in order to make more quantitative statements, a numerical simulation of the corresponding experiment is required. To this end, we have performed simulations using a microscopic approach based on a rate equation model for the single-atom excitation dynamics. The approach will be described in more detail elsewhere [6]. Briefly, one can solve, for a single, non-interacting atom, the optical Bloch equations of the three-level system corresponding to the two-step excitation scheme used in the experiments [3, 4]. One can further show that the Rydberg excitation dynamics of an atom can be well described by a simple rate equation for an effective two-level system under the conditions of these experiments[3]. We then use a Monte Carlo procedure to model the full system of interacting atoms, where the van der Waals interaction between the atoms leads to a—time- and space-dependent—shift of the detuning of each atom from resonance with the excitation laser and hence to a change in excitation probability. In this way, we can model the evolution of systems of several thousand atoms, which is clearly beyond the capabilities of quantum mechanical *ab initio* approaches [5], within a few minutes of CPU time. Moreover, as we simulate the full system, we obtain information about spatial correlations as well as, our focus in the present letter, statistical properties of the Rydberg excitation, which is not possible with the mean-field approach pioneered in [2].

For our simulations, we use parameters corresponding to those of the experiment [4]. More precisely, we assume a Gaussian ground-state density with peak density $\rho_0 = 5 \times 10^9$ cm^{-3} and width $\sigma = 500$ μm, and an excitation laser waist of 16 μm FWHM, leading to an effective excitation volume of 1.8×10^{-4} mm^3 containing $N_A \sim 900$ atoms. The Rabi frequency of the laser pumping the 5S $\rightarrow$ 5P transition is kept fixed at $\Omega_{pump} = 2\pi \times 4$ MHz, while the intensity of the Rydberg excitation laser is varied in order to achieve the desired Rydberg atom number $\langle N_e \rangle$. The interaction between two Rydberg atoms is calculated using the simplified two-state model used in [7], leading to a van der Waals-like $1/R^6$ behaviour at large distances and a dipole–dipole-like $1/R^3$ behaviour at short distances. To be concrete, the level shift Δ due to the interaction of two Rydberg atoms in nD states is given by $\Delta(R) = \frac{1}{2}\left(\Delta_0 - \sqrt{\Delta_0^2 + 4V^2(R)}\right)$, where $V(R) = -\mu_{DF}\mu_{DP}/R^3$ is the dipole coupling of the nD–nD asymptote to the energetically nearby $(n-2)$F–$(n+2)$P$_{3/2}$ molecular state and Δ_0 is the energy difference between those two asymptotes. We use the parameters of the 46D–46D $\leftrightarrow$ 48P$_{3/2}$–44F transition ($\mu_{DP} = 1548$ au, $\mu_{DF} = 1587$ au, $\Delta_0 = 0.003$ au [7]) as reference and calculate the interaction strength of other principal quantum numbers by applying appropriate scaling laws ($\mu_{DP}\mu_{DF} \sim n^4$, $\Delta_0 \sim n^{-3}$). For each set of initial conditions, 200 000 repetitions have been performed in order to have good statistics. The resulting Rydberg atom counting statistics is shown in figure 1 for the case of negligible interaction, principal quantum number $n = 45$, and for a significant amount of interaction, $n = 85$. While the distribution is close to a Poissonian in the first case, it is strongly sub-Poissonian

[3] In fact, the excitation pulse lengths used in [4] are at the border of the regime of applicability of the rate equation model since coherences have just enough time to damp out.

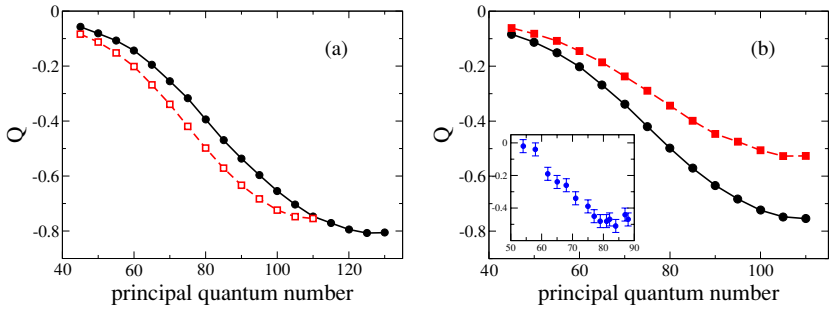

Figure 2. (a) Mandel Q-parameter (1) as a function of principal quantum number of excited Rydberg state for two different values of $\langle N_e \rangle = 30$ (filled circles) and $\langle N_e \rangle = 43$ (open squares). Results are shown only up to the principal quantum number where the Rydberg Rabi frequency Ω_{Ryd} required to excite the desired number of atoms starts to become comparable to the pump Rabi frequency Ω_{pump}, more precisely $\Omega_{Ryd}^2 = \Omega_{pump}^2/2$, where the assumptions underlying our rate equation treatment start to break down. (b) Q-parameter Q_D of the measured distribution (squares) versus that of the true distribution, Q_A (circles), demonstrating the effect of a finite detector efficiency η. For comparison, the experimental values of [4] are shown as an inset.

in the latter, in accordance with the experimental findings of [4]. More quantitatively, the Q-parameter (1) is plotted in figure 2 as a function of principal quantum number n of the Rydberg state. Since the coefficient of the Rydberg–Rydberg interaction strongly scales with n, the principal quantum number directly determines the strength of the interatomic interaction. As can be seen, there is indeed a smooth transition from the non-interacting limit $Q \lesssim 0$ towards the highly sub-Poissonian case where $Q \to -1$. Comparing our figure 2 to figure 3 of [4], there is a very good qualitative agreement between the experimental and the theoretical results. Quantitative discrepancies between the two may to some extent be attributed to the fact that the value we use for the strength of the interaction, determined from a two-state approximation, is probably not very accurate for the nD states used in the experiments. Changing this interaction strength would correspond to a compression or stretching of the abscissa in figure 2, explaining the fact that a saturation of Q is observed at smaller quantum numbers n in the experiment. Moreover, the issue of a finite detection efficiency of the MCP detector used in the experiment has been raised in [4]. In order to quantify this effect, we take it into account in our simulations by artificially introducing a detection probability η for the Rydberg atoms. As can be seen in figure 2(b), doing so modifies the Q-parameter of the observed distribution quantitatively, leading to smaller Q-values at large interaction strength. The value of Q obtained under the assumption of a 70% detection efficiency agrees very well with what is observed in the experiment [4]. Note that the relation between Q_A, the Q-parameter of the true distribution, and Q_D, the Q-parameter of the measured distribution, is not a trivial scaling $Q_D = \eta Q_A$. This is due to the fact that the two curves correspond to equal *measured* $\langle N_e \rangle$, but different *real* $\langle N_e \rangle$, hence different amount of interaction in the gas. On the other hand, the scaling relation applies for distributions with the same real $\langle N_e \rangle$.

In figure 2, we also compare the behaviour of Q for different Rydberg laser intensities leading to a different average excitation of $\langle N_e \rangle = 30$ and $\langle N_e \rangle = 43$, respectively. As discussed above, the value of Q in the interaction-free low-n limit depends on the ratio $\langle N_e \rangle / N_A$, i.e. on the average number of excited atoms. This different low-n limit of Q in the two cases is immediately apparent from the figure. In the opposite limit of strong interaction, the curve corresponding to a larger number of Rydberg atoms seems to flatten out at smaller

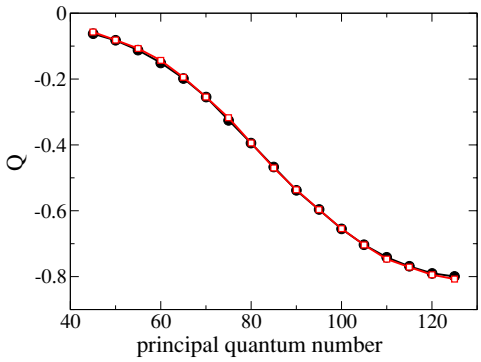

Figure 3. Q-parameter as a function of principal quantum number of excited Rydberg state for two different excitation pulse profiles (open squares: constant Rydberg Rabi frequency, full circles: $\sin^2$-envelope). For details, see the text.

n and at a higher value of Q, which would be in agreement with the qualitative picture that in this interaction-dominated regime the number $N_A/N_e - 1$ of 'blockaded atoms' per excited atom, and hence the interaction-induced suppression of excitation, is smaller for larger $\langle N_e \rangle$.

As stated in the caption of figure 2, results are shown there only up to the principal quantum number where $\Omega_{Ryd}^2 = \Omega_{pump}^2/2$. At this point, the assumptions underlying the rate equation treatment start to break down [6]. While this specific cut-off is due to the rate equation model used in the present simulations we note, however, that the curves shown in figure 2 can *in principle* not be continued to arbitrarily high Rydberg principal quantum numbers. This is not merely an artefact of the numerical treatment, but applies also to the experiment. The reason is the nonlinear dependence of the effective Rydberg excitation rate on the Rydberg Rabi frequency in the two-step excitation scheme used in [4]. Contrary to a single-step excitation such as used, e.g., in [2], increasing Ω_{Ryd} while keeping all other parameters constant results in a maximum of the effective excitation rate for a finite Ω_{Ryd}, beyond which it decreases again. Consequently, for large interaction strength the corresponding interaction-induced detuning from resonance cannot be compensated by increasing the Rydberg Rabi frequency accordingly.

As mentioned above, the pulse lengths of $\approx$100 ns used in [4] are at the border of the regime where our rate equation model accurately describes the evolution of the Rydberg level population for a single, non-interacting atom. However, in a numerical simulation it is possible to use longer excitation pulses of 2 μs, with the intensity of the Rydberg excitation laser adjusted so as to yield the same average number of Rydberg atoms. In such a pulse regime, the rate equation model accurately describes the single-atom dynamics. Within the limit of validity of our rate equation model, $\Omega_{Ryd} \lesssim \Omega_{pump}/\sqrt{2}$ for both pulses, such a stretching of the pulse does not visibly change the statistical properties of the excitation process, i.e. the Q-value. Moreover, we have gone further and investigated the influence of the excitation pulse profile on the statistics. In figure 3, we show the Q-parameter for two different scenarios, namely constant Rydberg Rabi frequency on the one hand and a $\sin^2$-envelope of the Rabi frequency on the other. As can be seen, both excitation schemes lead to the same statistical properties of the Rydberg number distribution. Hence, we speculate that the quenching of this distribution is indeed purely an effect of the interatomic interaction, and probably to a large extent independent of the precise properties of the excitation process, such as the temporal profile of the excitation pulse or interatomic coherences.

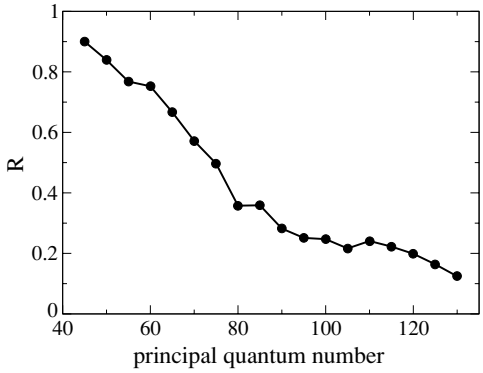

Figure 4. Asymmetry of the Rydberg atom number distribution for $\langle N_e \rangle = 30$, parameterized by the parameter R (equation (2)).

As a final point, we address the possibility of quantifying the counting statistics beyond the second moment of the distribution entering the Q-parameter. From the physical basis of the excitation blockade, one would intuitively expect a stronger quenching of the distribution for Rydberg atom numbers $N_e > \langle N_e \rangle$ than for $N_e < \langle N_e \rangle$, since the interaction-induced blockade will inhibit the excitation of a large number of Rydberg atoms but not that of a small number. Hence, one might expect an asymmetry of the counting statistics beyond the 'natural' asymmetry of the Poisson distribution due to the finite value of $\langle N_e \rangle$. To quantify this effect, we define the parameter

$$R = \frac{\langle (N_e - \langle N_e \rangle)^3 \rangle}{\langle N_e^2 \rangle - \langle N_e \rangle^2} \tag{2}$$

which contains the third moment of the atom number distribution and hence is a measure for its asymmetry. By construction $R = 1$ for a Poisson distribution and $R = 1 - 2P_e$ for non-interacting atoms. In figure 4, it is shown as a function of principal quantum number n. As expected, R is found to decrease monotonically as the strength of the interaction between the atoms increases, thereby demonstrating the changing asymmetry of the atom number distribution. So far, experimental evidence for this change has not been presented. It might in fact be obtained from the distributions measured in [4]. We note, however, that we found the third moment of the distribution entering R to fluctuate much more strongly than the variance entering Q, so that averaging over a significantly larger number of realizations was necessary to obtain converged results. Hence, the 5000 realizations measured in [4] might not be sufficient to observe a clear trend.

In summary, we have developed a theoretical description of Rydberg excitation in ultracold atomic gases which can be easily implemented numerically. Our approach, based on a rate equation description of the single-atom dynamics and a Monte Carlo treatment of the interacting system, is particularly well suited for the microscopic simulation of large ensembles of atoms. We have focused on the properties of the Rydberg atom counting statistics as measured in [4]. The results of our simulations agree well with the findings of the experiment, clearly demonstrating the change in the statistics due to the interaction between the atoms. Taking into account a reasonable estimate for the counting efficiency of the detector used in the experiment, quantitative agreement with the measured Q-values in the strongly interacting limit has been obtained. The fact that the transition to a sub-Poissonian distribution takes place at higher quantum numbers n in the simulation can be attributed to a simplified expression for the

interaction potential which probably underestimates the strength of the interaction. In addition to the simulation of the experiment [4], we have investigated the influence of the temporal profile of the excitation pulse on the Rydberg number statistics. For the case considered ($\sin^2$-envelope of the Rydberg Rabi frequency), we have found the statistics to be independent of the pulse shape, i.e. entirely determined by the average number of Rydberg atoms in the final state. Moreover, we have gone beyond the parameterization of the distribution by its width, measured by Mandel's Q-parameter, and demonstrated its asymmetry using the parameter R defined in (2). As might be expected from the physical basis of the excitation blockade, larger numbers of Rydberg atoms are suppressed more strongly than smaller numbers.

Acknowledgments

We would like to thank Georg Raithel and his group for sending us their experimental data in electronic form.

References

[1] Lukin M D, Fleischhauer M, Côté R, Duan M L, Jaksch D, Cirac J I and Zoller P 2001 *Phys. Rev. Lett.* **87** 037901
[2] Tong D, Farooqi S M, Stanojevic J, Krishnan S, Zhang Y P, Côté R, Eyler E E and Gould P L 2004 *Phys. Rev. Lett.* **93** 063001
[3] Singer K, Reetz-Lamour M, Amthor T, Marcassa L G and Weidemüller M 2004 *Phys. Rev. Lett.* **93** 163001
[4] Cubel Liebisch T, Reinhard A, Berman P R and Raithel G 2005 *Phys. Rev. Lett.* **95** 253002
[5] Robicheaux F and Hernández J V 2005 *Phys. Rev.* A **72** 063403
[6] Ates C, Pohl T, Pattard T and Rost J M 2006 in preparation
[7] Li W, Tanner P J and Gallagher T F 2005 *Phys. Rev. Lett.* **94** 173001