# Atom Counting Statistics in Ensembles of Interacting Rydberg Atoms

T. Cubel Liebisch, A. Reinhard, P. R. Berman, and G. Raithel

*FOCUS Center and Michigan Center for Theoretical Physics, Department of Physics, University of Michigan,*
*Ann Arbor, Michigan 48109, USA*

We show that the probability distributions for the number of Rydberg excitations in small ensembles of cold atoms, excited using short (100 ns) laser pulses, can be highly sub-Poissonian. The phenomenon occurs if the atom density and the principal quantum number of the excited Rydberg level are sufficiently high. Our observations are attributed to a blockade of the Rydberg atom excitation.

PACS numbers: 32.80.Rm, 03.67.−a, 34.20.Cf

Excitation of ensembles of cold atoms into Rydberg states produces collective quantum states in which Rydberg excitations are coherently distributed among all atoms. For instance, the first excited state of an $N$-atom ensemble is a state with one shared excitation, $|N, 1\rangle = (1/\sqrt{N}) \sum_{i=1}^{N} |g_1, g_2, \ldots, r_i, \ldots, g_N\rangle$, where the subscripts are atom labels, and $|g\rangle$ denotes an atom in the ground and $|r\rangle$ an atom in the Rydberg state. Because of interactions between Rydberg atoms, such as van der Waals and dipole-dipole interactions, the energy levels of the system deviate from an equidistant ladder $E_k = kE_{\text{Ryd}}$, where $k$ is the number of Rydberg excitations and $E_{\text{Ryd}}$ the energy of a single, isolated Rydberg atom. In the illustration in Fig. 1(a), the energy of the second excited state, $|N, 2\rangle$, is lowered by an interaction energy $V_{\text{Int}}$. If the laser excitation is tuned to the lowest transition $|N, 0\rangle \rightarrow |N, 1\rangle$ and the excitation bandwidth is less than $|V_{\text{Int}}|/h$, there is negligible excitation of all but the first collective excitation $|N, 1\rangle$. Such a blockade effect of Rydberg atom excitation has recently received much attention because it can be useful in quantum information processing [1,2], quantum cryptography [3], and in improving spectroscopic resolution [2]. Experimental evidence of a suppression of Rydberg excitation in large samples has been obtained [4,5]. Moreover, additional studies of Rydberg systems have been carried out to characterize the blockade [6–10] and to design a quantum phase gate [11,12]. In this work we present evidence for a blockade effect using an atom counting approach, in which the blockade manifests itself via sub-Poissonian Rydberg atom counting statistics. The statistical method differs from previous work [4,5] in that it is particularly well suited for the study of few-atom samples rather than large atom clouds. If the Rydberg blockade is to be used in quantum information processing applications, it will be important to learn how to employ the blockade in small atomic systems.

Within an extended atomic ensemble with a Rydberg-excitation blockade, it is expected that there will be multiple uniform regions in which only one collective excitation occurs; such regions are represented by the bubbles in Fig. 1(b). The radius $R_b$ of these bubbles is determined by the strength of the Rydberg-Rydberg interaction and the

laser bandwidth, which is 5 MHz in our experiment [13]. The number of Rydberg excitations in the whole ensemble approximately equals the number of bubbles that fit into the excitation volume. Thus, from one realization of the excitation process to another, the number of Rydberg excitations does not fluctuate greatly. Therefore, a Rydberg-excitation blockade is expected to result in a sub-Poissonian distribution of the number of Rydberg excitations. We observe this effect by repeated generation and measurement of Rydberg excitations in identically prepared atomic ensembles. We thereby establish a close connection between the Rydberg atom counting statistics and the presence of a blockade in the Rydberg atom excitation. A recent simulation of strongly interacting atoms by Robicheaux [14] appears to be consistent with our findings.

In our experiment, $^{85}$Rb atoms are collected in a vapor cell magneto-optical trap (MOT) and excited into $nD$ Rydberg states using two-photon excitation. The lower ($5S \rightarrow 5P$, $\lambda = 780$ nm) and the upper transitions ($5P \rightarrow nD$, $\lambda \approx 480$ nm) are excited using coincident, counter-propagating, on-resonant laser pulses of 100 ns duration. The laser pulses are formed by passing frequency-stabilized continuous-wave lasers through amplitude-modulated acousto-optic modulators. The laser beam driving the lower transition has a linewidth of 1–2 MHz and is collimated to about 3 mm, which is much larger than the
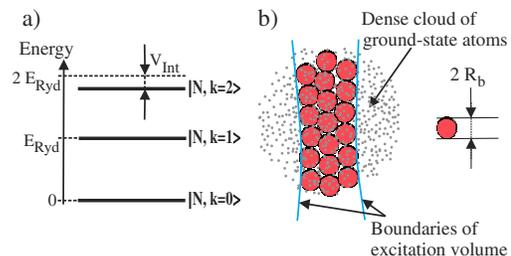


FIG. 1 (color online). (a) Lowest collective states of an ensemble of $N$ atoms containing $k$ Rydberg excitations. Rydberg-Rydberg interaction causes an energy shift $V_{\text{Int}}$ of the state $|N, k = 2\rangle$. (b) Model of an extended atomic ensemble with a Rydberg-excitation blockade of a range $R_b$. Each "bubble" represents a region in which, ideally, there exists exactly one Rydberg excitation.

MOT size. We use Rabi frequencies of the lower transition in a range from 3 to 9 MHz, as determined by Autler-Townes measurements [13]. The beam driving the upper transition is derived from a frequency-doubled 960 nm diode laser (Toptica Laser DL-100) that is locked to a pressure-tuned Fabry-Perot cavity [15] and has a linewidth of $\approx 5$ MHz (at 480 nm). To achieve sufficiently large Rabi frequencies on the upper transition, the 480 nm beam is focused into the MOT with a full width at half maximum diameter of the intensity distribution of 16 $\mu$m $\pm 1$ $\mu$m, which is much less than the MOT size. The Rayleigh length of the 480 nm beam is 1.1 mm $\pm$ 0.1 mm, which is larger than the MOT size. The MOT was run at a higher-than-usual field gradient (50 G/cm) and with quite small-diameter laser beams in order to limit the MOT diameter to 500 $\mu$m while still having a central atom density of about $5 \times 10^9$ cm$^{-3}$. In this configuration, we achieve densities of a few times $10^8$ cm$^{-3}$ of high-lying Rydberg states (principal quantum numbers $n = 54$–84) in a cylindrical volume of about 20 $\mu$m in diameter and about 500 $\mu$m in length that contains of order 1000 ground-state atoms.

The Rabi frequency on the lower transition is practically constant in this volume, while the Rabi frequency on the upper transition falls off with the distance from the beam axis. As the target Rydberg state is varied from $54D_{5/2}$ to $84D_{5/2}$, the Rabi frequency of the upper transition at a beam power of 1.2 mW varies from $\sim 5$ to $\sim 2.5$ MHz. To avoid detector saturation and ionization, which are discussed below, we typically reduce the intensity of the upper-transition beam such that only about 50 Rydberg excitations are generated in the above specified volume. Each excitation is associated with a bubble such as in Fig. 1(b). As $n$ varies from 50 to 90, the number of ground-state atoms in each bubble increases from roughly one to ten. About 700 ns after the excitation pulses, the Rydberg excitations are field ionized using an approximately linear field ionization ramp. The liberated electrons are detected using a micro-channel plate (MCP) detector, and the MCP pulses are counted. This sequence is repeated up to 5000 times. The resultant probability distributions for the counting results yield the Rydberg atom counting statistics.

In Fig. 2(a), we show a typical Rydberg atom counting statistics for excitation into the $84D_{5/2}$ Rydberg level. A comparison between the measurement result [histogram in Fig. 2(a)] and a Poissonian distribution with the same average [line in Fig. 2(a)] shows that the Rydberg atom counting statistics is sub-Poissonian. The degree of sub-Poissonian character is a measure of blockade effectiveness. As a quantitative parameter characterizing the width of the distribution we use the Mandel $Q$ parameter, defined as the variance in the count distribution divided by the mean number of counts minus one, $Q = \frac{\langle N^2 \rangle - \langle N \rangle^2}{\langle N \rangle} - 1$ [16]. Values of $Q > 0$ correspond to super-Poissonian, $Q = 0$ to Poissonian, and $Q < 0$ to sub-Poissonian distributions. The distribution in Fig. 2(a) is found to have a $Q$ value of $-0.51 \pm 0.04$, which qualifies as highly sub-Poissonian.
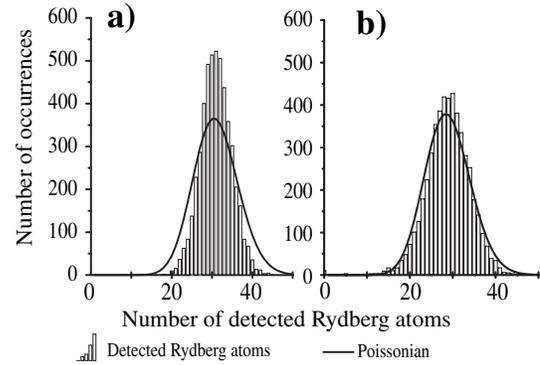


FIG. 2. (a) The histogram shows the probability distribution of the number of detected $84D_{5/2}$ Rydberg atoms, obtained from 5000 realizations of the experiment. A highly sub-Poissonian $Q$ value of $-0.51 \pm 0.04$ is found. To make the sub-Poissonian character of the measured distribution more apparent, the histogram is compared with a Poissonian with the same average (line). (b) Reference experiment at a Rydberg atom density that is $\sim 100$ times lower than in (a). A close-to-Poissonian $Q_D$ value of $-0.08 \pm 0.04$ is found.

It is noted that the $Q$ value of the *detected* number of Rydberg atoms, henceforth referred to as $Q_D$, is not the same as the $Q$ value of the number of Rydberg atoms present in the sample before detection, referred to as $Q_A$. While $Q_D$ is what we measure, $Q_A$ actually is of higher fundamental interest. If each Rydberg atom produces a count with a fixed probability $\eta$, which is a physical property of the utilized MCP detector, then $Q_D = \eta Q_A$. Since $Q_A \geq -1$, our measurement result $Q_D = -0.51$ implies that $\eta \geq 0.51$. An $\eta$ value of 0.51 is at the lower limit of the range of typically specified MCP detection efficiencies for low-energy electrons. The range $0.5 \lesssim \eta \lesssim 0.85$ given in Ref. [17] corresponds to $-1 \lesssim Q_A \lesssim -0.60$ for a measured $Q_D = -0.51$. It is concluded that with certainty $Q_A \leq -0.51$, and that $Q_A \leq -0.60$ for the likely case that the $\eta$ value of the MCP installed in our system lies within a typical range.

In the low-density reference experiment shown in Fig. 2(b), the diameter of the upper-transition ($5P_{3/2} \rightarrow 84D_{5/2}$) laser beam was increased by about a factor of 10 while adjusting its intensity to yield an average of 30 detected Rydberg atoms. The excitation volume in the reference experiment was about a factor 100 larger than under the conditions of Fig. 2(a), and the Rydberg atom density about a factor 100 lower. The reference experiment yields a Rydberg atom counting statistics that is close to Poissonian, with a $Q_D$ value of $-0.08 \pm 0.04$. This result reflects the fact that the average Rydberg-atom separation $R$ in the reference experiment is a factor of order 4 larger than under the conditions of Fig. 2(a). Consequently, van-der-Waals and dipole-dipole interactions—which scale as $1/R^6$ and $1/R^3$, respectively—do not contribute significantly to the results of the reference experiment [Fig. 2(b)]. There, the observed counting statistics is determined by the variation in ground-state atom number and the low excita-

tion probability of an isolated atom rather than by the number of "bubbles" contained in the excitation volume (as suggested in Fig. 1).

We have investigated the dependence of the $Q_D$ values on $n$. For each data point in Fig. 3, the powers of the excitation lasers were adjusted such that the average number of counted Rydberg atoms was ~30. For $n \lesssim 58$, the $Q_D$ values approach zero. Thus, for $n \lesssim 58$ the excitation process appears to be random. In the range $58 \lesssim n \lesssim 77$, the $Q_D$ value drops from 0 to $-0.5$ and levels out at $\approx -0.5$ for $n \gtrsim 77$. We attribute this behavior to an excitation blockade in the Rydberg atom excitation process that becomes increasingly important for larger $n$. Figure 3 demonstrates the transition between the uncorrelated domain ($n \lesssim 58$) and the blockade domain ($n \gtrsim 77$).

The observed blockade originates most likely from a few near-elastic resonances of the type $|n, l\rangle \otimes |n, l\rangle \rightarrow |n', l'\rangle \otimes |n'', l''\rangle$ with large matrix elements $\langle n, l|r|n', l'\rangle$ and $\langle n, l|r|n'', l''\rangle$ ($|n, l\rangle$ is the initial state). For simplicity, we assume that only one such coupled state is important and that it differs in energy by $\Delta E$ from the initial state. The perturbed energy levels of a pair of atoms in Rydberg states, located at a separation $R_{\mathrm{Ryd}}$, are obtained by diagonalizing a $2 \times 2$ matrix with values $-\Delta E/2$ and $+\Delta E/2$ on the diagonal and a value of order $n^4/R_{\mathrm{Ryd}}^3$ on the off-diagonal elements [6]. There, the off-diagonal elements account for the dipole-dipole interaction. The bubble diameter $2R_{\mathrm{b}}$ of Fig. 1(b) is approximately obtained by equating the interaction-induced level shift with the laser linewidth $\delta\nu_{\mathrm{L}}$, i.e., by $\sqrt{(\Delta E/2)^2 + (n^4/(2R_{\mathrm{b}})^3)^2} - \Delta E/2 \approx \delta\nu_{\mathrm{L}}$. In our experiment $\delta\nu_{\mathrm{L}} \approx h \times 5$ MHz (in laboratory units). For rubidium $nD_{5/2}$ Rydberg states with $50 \lesssim n \lesssim 90$, $\Delta E$ typically is of order $h \times 100$ MHz. Using this estimate, level shifts of order $\delta\nu_{\mathrm{L}}$ require coupling terms $n^4/(2R_{\mathrm{b}})^3$ of order $h \times 20$ MHz. The corresponding bubble diameters are $2R_{\mathrm{b}} \sim 8$ $\mu$m for $n = 58$ and $2R_{\mathrm{b}} \sim 12$ $\mu$m for $n = 77$. Since the average separation between ground-state atoms is $R \sim 6$ $\mu$m, from these bubble-size estimates it appears reasonable that the block-

ade gradually develops over the observed range $58 \lesssim n \lesssim 77$.

A question of some interest is whether the Rydberg interactions in the blockaded ensemble are of dipole-dipole or van der Waals character. Following Ref. [6], we compare the off-diagonal and the diagonal elements of the above mentioned matrix and note that the interaction has dipole-dipole character if $n^4/R_{\mathrm{Ryd}}^3 > \Delta E$ and van der Waals character if $n^4/R_{\mathrm{Ryd}}^3 < \Delta E$. In the previous paragraph we have estimated $\Delta E \sim h \times 100$ MHz and, for atom pairs with separations of the order of the bubble diameter, $n^4/R_{\mathrm{Ryd}}^3 \sim h \times 20$ MHz (in laboratory units). Therefore, atom pairs that are close enough to be "just about blockaded" mostly interact via van der Waals interaction, regardless of $n$. On the other hand, in the deeply blockaded domain there exist many atom pairs at distances $\ll 2R_{\mathrm{b}}$, for which the interaction takes dipole-dipole character. For instance, for $n = 84$ we estimate a bubble diameter of $2R_{\mathrm{b}} \sim 13$ $\mu$m and a critical separation below which the interaction leans towards dipole-dipole character of about 8 $\mu$m. In this case, a fraction of $(8/13)^3 \approx 25\%$ of the blockaded atom pairs are blockaded by dipole-dipole interaction. It is noted that all estimates in this and the previous paragraph are strictly qualitative. The exact values of $\Delta E$ and the dipole-dipole interaction matrix element can shift the balance towards either side. Also, the assumption that there exists a single, dominant dipole-dipole interaction channel is not expected to hold in generality.

Varying the frequency of the upper-transition ($5P_{3/2} \rightarrow nD_{5/2}$) laser, we studied the width of the Rydberg-excitation lines as a function of the intensity of the upper-transition laser. Over the full range of intensities we were able to realize with our laser, the FWHM linewidth was observed to slightly increase from $\approx 12$ MHz to $\approx 14$ MHz. This result is in strong contrast to Ref. [5], where substantial line broadening is reported. We attribute this difference to the absence of secondary effects in our experiment, as discussed in the next paragraph. We have further observed that the average number of detected Rydberg atoms versus the intensity of the upper-transition laser exhibits a saturation behavior similar to that reported in Ref. [4], which has been attributed to a Rydberg-excitation blockade. Finally, we have investigated how the $Q_D$ values depend on the detuning of the upper-transition laser from the center of the Rydberg-excitation line. On both sides of the line the $Q_D$ values increase from about $-0.5$ to zero, and the increase occurs more rapidly on the blue-detuned side than on the red-detuned side. A full understanding of the line shapes and the dependence of $Q_D$ on detuning awaits a theoretical analysis.

While the high susceptibility of Rydberg atoms to external fields and other Rydberg atoms makes the blockade effect possible, it also leads to experimental challenges in distinguishing the blockade from other effects. Long excitation pulses inevitably cause Rydberg atom ionization and plasma formation when large densities of Rydberg atoms
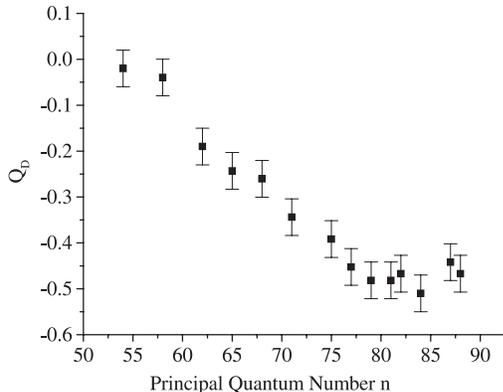


FIG. 3. $Q_D$ values as a function of principal quantum number $n$. The average number of detected Rydberg atoms is kept constant at about 30.
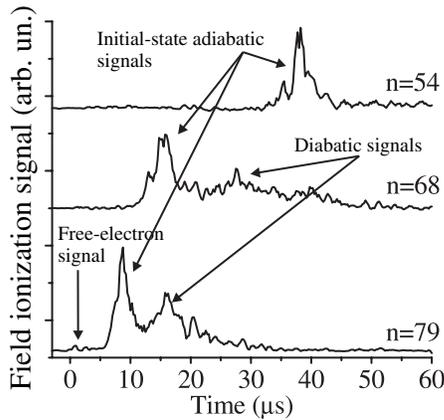
FIG. 4.    State-selective field ionization spectra taken under the same conditions as in Fig. 3 for the indicated principal quantum numbers. The linear field ionization ramp pulse starts at time $t = 0$. Only the case $n = 79$ shows a very small free-electron component.

are excited [6,18,19]. If plasma formation occurs during the course of the excitation pulse, plasma electric fields could cause line broadening and saturation of Rydberg atom excitation due to Stark shifts, thus mimicking a blockade effect. To exclude this possibility, we have verified by state-selective field ionization that between excitation and detection the number of free electrons generated is negligible (see Fig. 4). The main peaks in Fig. 4 are adiabatic field ionization signals of the initial state. The broad signals, which are only present at very high $n$, are likely to be diabatic field ionization signals (as opposed to collision-induced features) because they did not noticeably depend on the number of excited Rydberg atoms. The field ionization ramp reaches 110 V/cm, which is sufficient to ionize all initial-state atoms with $n$ as low as 42. Even if there were $l$- or $m$-changing collisions, for which we have found no indication under the conditions used, we could have detected all high-$l$ states for $n$ as low as 59. It needs to be stressed that we had to use excitation pulses as short as 100 ns in order to achieve ionization- and collision-free conditions. Moreover, transform-limited pulses with temporal widths shorter than $\sim$10 ns should not be used, because the bandwidth of these pulses would be too large to resolve the interaction-induced energy level shifts that give rise to the blockade phenomena discussed in this Letter. Therefore, the range in pulse length $\tau$ suitable for the reported experiments is limited to about $10\,\mathrm{ns} \lesssim \tau \lesssim 100\,\mathrm{ns}$.

Another important aspect to be considered is that detector saturation may lead to a sub-Poissonian counting distribution, even if the underlying atom distribution were Poissonian. In our experiments, the Rydberg atoms are field ionized with field ionization pulses having ramp speeds ranging from 0.35 V/cm/$\mu$s to 1.1 V/cm/$\mu$s and are then detected by the MCP detector. The MCP pulses have a width of about 5 ns and are detected using a gated discriminator/pulse counter. The field ionization ramp speeds were chosen slow enough that, for an average

number of 30 detected Rydberg atoms, the probability that any of the MCP pulses would overlap was deemed negligible. To test this assertion experimentally, we have taken data such as those shown in Fig. 2(a) for $\sim$100, 60, 30, 20, and 10 average Rydberg counts and at $n = 84$. In this test, the average number of Rydberg counts was adjusted by varying the lower-transition Rabi frequency. The upper-transition Rabi frequency and geometrical parameters such as spot sizes were fixed. The $Q_D$ values did not change more than $\pm0.04$, with $Q_D$ values becoming slightly less negative for higher average counts. Therefore, we are certain that at an average count of 30, used in most data shown, detector saturation did not systematically reduce the measured $Q_D$ values.

Using pulsed laser excitation of cold-atom clouds, we have shown that a blockade due to Rydberg-Rydberg interactions can be used to produce collective Rydberg atom excitations characterized by a highly sub-Poissonian number-state distribution. We have estimated that the blockade is predominantly caused by van der Waals interactions. Future work will include detailed studies of the spectral structure of the Rydberg resonance line and of the dependence of $Q$ on the detuning of the excitation laser from the center of the resonance. Also, it is planned to study smaller atomic samples and interactions between such samples, as these will be appropriate for further development of this research towards applications in quantum information processing.

[1]  D. Jaksch *et al.*, Phys. Rev. Lett. **85**, 2208 (2000).
[2]  M. D. Lukin *et al.*, Phys. Rev. Lett. **87**, 037901 (2001).
[3]  M. Saffman and T. G. Walker, Phys. Rev. A **66**, 065403 (2002).
[4]  D. Tong *et al.*, Phys. Rev. Lett. **93**, 063001 (2004).
[5]  K. Singer *et al.*, Phys. Rev. Lett. **93**, 163001 (2004).
[6]  Wenhui Li, P. J. Tanner, and T. F. Gallagher, Phys. Rev. Lett. **94**, 173001 (2005).
[7]  S. M. Farooqi *et al.*, Phys. Rev. Lett. **91**, 183002 (2003).
[8]  A. L. de Oliveira *et al.*, Phys. Rev. Lett. **90**, 143002 (2003).
[9]  K. Afrousheh *et al.*, Phys. Rev. Lett. **93**, 233001 (2004).
[10]  T. J. Carroll *et al.*, Phys. Rev. Lett. **93**, 153001 (2004).
[11]  I. E. Protsenko *et al.*, Phys. Rev. A **65**, 052301 (2002).
[12]  M. Saffman and T. G. Walker, quant-ph\0502051.
[13]  B. K. Teo *et al.*, Phys. Rev. A **68**, 053407 (2003).
[14]  F. Robicheaux and J. V. Hernández, Phys. Rev. A (to be published).
[15]  E. Hansis *et al.*, Rev. Sci. Instrum. **76**, 033105 (2005).
[16]  L. Mandel, Opt. Lett. **4**, 205 (1979).
[17]  J. L. Wiza, Nucl. Instrum. Methods **162**, 587 (1979), also posted at http://www.burle.com.
[18]  W. Li *et al.*, Phys. Rev. A **70**, 042713 (2004).
[19]  A. Walz-Flannigan *et al.*, Phys. Rev. A **69**, 063405 (2004).