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Citation for published version (APA):

Sanders, J., Jonckheere, M., & Kokkelmans, S. J. J. M. F. (2015). Sub-Poissonian statistics of jamming limits in ultracold Rydberg gases. *Physical Review Letters*, 115(4), 043002-1/5.
<https://doi.org/10.1103/PhysRevLett.115.043002>

DOI:

[10.1103/PhysRevLett.115.043002](https://doi.org/10.1103/PhysRevLett.115.043002)

Document status and date:

Published: 23/07/2015

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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- The final published version features the final layout of the paper including the volume, issue and page numbers.

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Sub-Poissonian Statistics of Jamming Limits in Ultracold Rydberg Gases

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(Received 10 April 2015; published 23 July 2015)

Several recent experiments have established by measuring the Mandel Q parameter that the number of Rydberg excitations in ultracold gases exhibits sub-Poissonian statistics. This effect is attributed to the Rydberg blockade that occurs due to the strong interatomic interactions between highly excited atoms. Because of this blockade effect, the system can end up in a state in which all particles are either excited or blocked: a *jamming limit*. We analyze appropriately constructed random-graph models that capture the blockade effect, and derive formulae for the mean and variance of the number of Rydberg excitations in jamming limits. This yields an explicit relationship between the Mandel Q parameter and the blockade effect, and comparison to measurement data shows strong agreement between theory and experiment.

DOI: 10.1103/PhysRevLett.115.043002

PACS numbers: 32.80.Rm, 02.50.Ga

Ultracold gases with atoms in highly excited states have attracted substantial interest over recent years, for example, for their potential application in quantum computing [1–3], and for the study of nonequilibrium phase transitions [4]. These atomic systems exhibit complicated spatial behavior due to strong van der Waals or dipolar interactions between neighboring atoms, which has been demonstrated through several experimental observations of reduced fluctuation in the number of excitations in ultracold gases of Rydberg atoms [5–10].

In these experiments, a laser facilitates excitation of ultracold atoms into a Rydberg state. After some time t , information on the mean and variance of the number of excited particles $X(t)$ is obtained by repeating counting experiments, and the Mandel Q parameter [11]

$$Q(t) = \frac{\text{Var}[X(t)]}{\mathbb{E}[X(t)]} - 1 \quad (1)$$

is calculated to quantify a deviation from Poisson statistics, since if $X(t)$ is Poisson distributed, $Q(t) = 0$. The experiments establish that $Q(t) < 0$ for $t > 0$, and $X(t)$ is said to have a sub-Poisson distribution.

The observed negative Mandel Q parameter is attributed to the Rydberg blockade effect [1,2]. There exist simulation techniques [12] and models based on Dicke states [6] that numerically describe the Mandel Q parameter. Besides for a one-dimensional system with reversible dynamics [13], no closed-form expression appears to be available that describes the relation between the Mandel Q parameter and the blockade effect.

Explicit formulae for the Mandel Q parameter are difficult to obtain, because the problem at hand is reminiscent of parking processes [14] and irreversible continuum random sequential adsorption problems [15], which is also where the term jamming limit comes from. The

standard two-dimensional continuum random sequential adsorption problem is that of throwing disks of radius $r > 0$ one by one randomly in a two-dimensional box, such that the disks do not overlap. Except for the one-dimensional variant, such problems are notoriously challenging to analyze due to spatial correlations. One further question is whether such stochastic processes are suited to explain effects occurring in ultracold Rydberg gases, and if so, under what conditions. This matter is discussed in Ref. [16], where a suitable stochastic process is provided based on rate equations that adequately describe the Rydberg gas when an incoherent process (such as spontaneous emission) occurs [17].

This Letter adopts the stochastic process in Ref. [16] that models the Rydberg gas, and uses it to study the Mandel Q parameter in the jamming limit which occurs when atoms only transition from the ground state to the Rydberg state. The model includes the blockade effect through so-called interference graphs, and by considering specially constructed large Erdős-Rényi (ER) random graphs [18] that retain essential features of the blockade effect, we overcome the mathematical difficulties normally involved with having a spatial component. The problem remains nontrivial though, and we point interested readers to the rigorous derivation of the necessary fluid and diffusion limits [19]. This Letter explains how to use these theoretical insights in the context of Rydberg gases through less complicated heuristic arguments, and while doing so explicitly relates the mean and variance of the number of excitations to the blockade effect.

We consider a gas of ultracold atoms in an excitation volume $V \subseteq \mathbb{R}^3$, and we assume that each particle has its own distinct position. Each particle can go from a ground state to a Rydberg state, and a particle in the Rydberg state prevents neighboring particles from also entering the Rydberg state. The density of particles is assumed to be ρ ,

and the number of excitable particles N within any region $A \subset V$ to be Poisson distributed with parameter ρA . This implies in particular that in the absence of blockade effects, the number of excited particles within the excitation volume, $X(t)$, will be Poisson distributed, as is the case in experiments [5–10]. It also implies that the particles are uniformly distributed at random over the excitation volume.

The blockade effect will be modeled using the notion of a blockade radius r . This is in line with simulations and measurements of pair correlation functions between atoms in the Rydberg state, which show a sharp cutoff when plotted as a function of the distance between the atoms [20,21]. Particles within a distance $r > 0$ are considered *neighbors* of each other, and neighbors each block the other if excited. We denote the number of neighbors of a particle $i = 0, 1, \dots, N$ within its blocking volume $V_{b,i}$ by B_i . As a consequence of these assumptions, the number of neighbors of particle i is also Poisson distributed. Specifically,

$$\mathbb{P}[B_i = b] = \frac{(\rho V_{b,i})^b e^{-\rho V_{b,i}}}{b!}, \quad b = 0, 1, \dots, \quad (2)$$

if $V_{b,i}$ is fully contained within V .

We will study the number of excitations by examining the asymptotic behavior of large ER random graphs. Each vertex of such a graph will represent one particle, so its set of vertices is $\mathcal{V} = \{1, \dots, N\}$. We draw an edge between two particles i and j if we consider i and j to be neighbors (particles that would block one another). One can construct an ER random graph by considering every pair of vertices (i, j) once, and drawing the edge between i and j with probability p , independent from all other edges. In order to deduce information on $X(t)$ through examining the ER random graph, we need to match the ER random graph model to the physical system, and we will do so by counting and matching the number of neighbors. Matching the models has to be done via the number of neighbors, because there is *no such notion as a physical position of a particle* in an ER random graph. This principle, in fact, makes this mathematical model tractable.

The number of neighbors $B_{\text{ER},i}$ of a particle i in the ER random graph is binomially distributed, $B_{\text{ER},i} \sim \text{Bin}(N-1, p)$, so that for $b = 0, 1, \dots, N-1$, $\mathbb{P}[B_{\text{ER},i} = b] = \binom{N-1}{b} p^b (1-p)^{N-1-b}$, and $\mathbb{E}[B_{\text{ER},i}] = (N-1)p$. When setting $p = c/N$ where c is some constant, we see that as $N \rightarrow \infty$, the distribution converges to a Poisson distribution,

$$\lim_{N \rightarrow \infty} \mathbb{P}[B_{\text{ER},i} = b] = \frac{c^b e^{-c}}{b!}, \quad b = 0, 1, \dots \quad (3)$$

Comparing Eq. (3) to Eq. (2), we note that the limiting distribution is the same if the average number of neighbors in the ER random graph, c , is related to the density and blockade volume as $c = \rho V_b$. By setting $c = \rho V_b$, we ensure that the particles in the ER random graph have the same distribution

FIG. 1 (color online). (left) A spatial Poisson point process in which neighbors within radius r block each other is used to choose appropriate parameters for (right) an ER random graph so that the particles have the same distribution of number of neighbors as $N \rightarrow \infty$. Note that this identification procedure only matches the distribution of the number of neighbors, and does not entail a mapping between the specific particles in both models.

of number of neighbors as in the spatial problem when the number of particles $N \rightarrow \infty$. Figure 1 summarizes the construction.

Let us now describe the spatial dynamics, illustrated in Fig. 2. At time $T_0 = 0$ the laser is activated, and from that point onward excitations can occur. At a time $T_1 > T_0$, the first particle, 1, excites and enters the Rydberg state. Because of the Rydberg blockade, particle 1 will subsequently prevent all other particles within a radius r from also becoming excited. Later, at a time $T_2 > T_1$, a second particle, 2, excites, which cannot be within distance r of particle 1. Particle 2 from that point onward also blocks particles within a distance r of itself. This process continues until some finite time $T_{X(\infty)} < \infty$ when all particles are either blocked or excited. The random number of excited particles $1 \leq X(\infty) \leq N$ is then detected.

The spatial dynamics are mimicked when building the ER random graph through an exploration algorithm [22] as follows. An unaffected particle $\circ$ is chosen uniformly at random. It becomes excited $\bullet$, and simultaneously a random subset of unaffected particles become blocked $\blacklozenge$, to which we draw an edge. This repeats until all particles are excited or blocked, and a jamming limit has been constructed. The Supplemental Material defines the exploration algorithm appropriately [23].

To derive the Mandel Q parameter, we need a stochastic recursion for the number of unaffected particles U_m at each m th moment an excitation occurs. This is obtained when considering that when the $(m+1)$ th excitation occurs, the

FIG. 2 (color online). (left) A random first particle excites. (middle) Subsequently, random second and third particles excite. (right) The process continues until all particles are either blocked or excited, and the resulting state is a jamming limit.

number of unaffected particles decreases by (i) the one particle that excites, and (ii) a random number of unaffected particles that each is a neighbor of the new excitation with probability p and thus now become blocked. Conditional on there being $N = n$ particles in the excitation volume, we thus have

$$U_{m+1} = U_m - 1 - \text{Bin}(U_m - 1, p), \quad U_0 = n. \quad (4)$$

We will now analyze this stochastic recursion, and identify the moment τ the number of unaffected particles is zero, i.e., $U_\tau = 0$. Precisely at this moment, we have that the number of excitations $X(\infty) = \tau$.

The Supplemental Material details the following steps [23]. From Eq. (4), we obtain a closed-form expression for $\mathbb{E}[U_m]$ by invoking the tower property and giving an induction argument. Through decomposition, we subsequently obtain an expression for $\text{Var}[U_m]$. When scaling the probability of being neighbors as $p = c/n$, the mean and variance converge to fluid limits, which can be seen by letting $f \in [0, 1]$, and proving that as $n \rightarrow \infty$,

$$\frac{\mathbb{E}[U_{[fn]}]}{n} \rightarrow u(f), \quad \frac{\text{Var}[U_{[fn]}]}{n} \rightarrow v(f). \quad (5)$$

Here, $[\dots]$ denotes rounding to the nearest integer, and the fluid limits are $u(f) = e^{-cf} - (1 - e^{-cf})/c$, and $v(f) = \{e^{-cf}(1 - e^{-cf})[(1 + 2c)e^{-cf} - 1]\}/(2c)$. Note that these fluid limits are rigorously proven in Ref. [19].

Consider now Fig. 3 (left) and the following steps. The process U_m hits zero when $m \approx f^*n$, with $f^* = \ln(1 + c)/c$ being the solution to $u(f^*) = 0$. Therefore,

$$\mathbb{E}[X(\infty)|N] \approx f^*n = \frac{n \ln(1 + c)}{c}. \quad (6)$$

To approximate the variance, calculate $u'(f)$ and note that $u'(f^* + \varepsilon) \approx -1$ for sufficiently small ε . Since U_m is probably near 0 for $m \approx f^*n$, the fluctuations in $X(\infty)$ will thus be of the order of $\sqrt{\text{Var}[U_{[f^*n]}]}$. Hence,

FIG. 3. (left) The fluid limit $u(f)$ together with a sample path of U_m/n for $n = 50$. The dashed line indicates the tangent at f^* , and the arrows indicate the typical fluctuations. (right) Histogram of $X(\infty)$ in a two-dimensional random sequential adsorption problem with precisely $n = 800$ particles, together with the probability density function of a normal distribution with mean $n \ln(1 + c)/c$ and variance $nc/[2(1 + c)^2]$.

$$\text{Var}[X(\infty)|N] \approx \text{Var}[U_{[f^*n]}] \approx v(f^*)n = \frac{nc}{2(1 + c)^2}. \quad (7)$$

Invoking the central limit theorem, we have for large fixed n that the number of excitations is approximately normal distributed with mean $n \ln(1 + c)/c$ and standard deviation $\sqrt{nc/[2(1 + c)^2]}$. This limit result, Eq. (6), and Eq. (7) are formally established by deriving diffusion limits in [19].

We will now compare Eqs. (6) and (7) to simulations of the mean and variance observed in the two-dimensional random sequential adsorption problem described earlier, and with periodic boundary conditions. We consider $h = 1 \mu\text{m}$, $l = w = 400 \mu\text{m}$, $r = 6.5 \mu\text{m}$, and $\rho = 5 \times 10^9 \text{cm}^{-3}$, which are typical values in magneto-optical traps, and correspond to $n \approx 800$ and $c \approx 0.664$. Figure 3 (right) shows a histogram of the number of excitations, as well as the probability density function of a normal distribution with mean $n \ln(1 + c)/c$ and variance $nc/[2(1 + c)^2]$. Compared to the simulation's outcome, the expressions differ for this set of parameters (i) 2.6% for the mean, (ii) 2.5% for the variance, and (iii) 0.015% for the Mandel Q parameter. Because the mean and variance are both overestimated, the Mandel Q parameter happens to be more accurately approximated. The errors the approximation makes can be attributed to the fact that particles in the ER random graph model have no physical position, whereas particles in two-dimensional Poisson disk throwing processes do exhibit spatial correlations. Intriguingly the random graph, which has no spatial interpretation, yields a good approximation.

It is important to understand that the results thus far are conditional on there being $N = n$ particles within the excitation volume. However, the number of particles within the excitation volume is random and Poisson distributed, specifically $N \sim \text{Poi}(\rho V)$. To obtain an unconditional expression for the mean and variance, we can utilize the tower property, $\mathbb{E}[X(\infty)] = \mathbb{E}[\mathbb{E}[X(\infty)|N]] \approx \mathbb{E}[N \ln(1 + c)/c] = \rho V \ln(1 + c)/c$, and decomposition, $\text{Var}[X(\infty)] = \mathbb{E}[\text{Var}[X(\infty)|N]] + \text{Var}[\mathbb{E}[X(\infty)|N]] \approx \mathbb{E}[Nc/(2(1 + c)^2)] + \text{Var}[N \ln(1 + c)/c] = \{c/[2(1 + c)^2] + [\ln(1 + c)/c]^2\}\rho V$. Recalling definition Eq. (1), the Mandel Q parameter in the jamming limit is therefore

$$Q(\infty) \approx \frac{c^2}{2(1 + c)^2 \ln(1 + c)} + \frac{\ln(1 + c)}{c} - 1, \quad (8)$$

which is exact in the ER case when $\rho V \rightarrow \infty$ [19]. Note that Eq. (8) only depends on the average number of neighbors c , which in fact explains observations on simulated Mandel Q parameters [12] as we discuss in Ref. [23].

Let us also discuss the time dependency of the mean number of excitations. We incorporate time dependency by assuming that every unaffected particle excites at rate λ , and specifically that $T_m - T_{m-1} \sim \text{Exp}(\lambda U_{m-1})$, which corresponds to modeling the Rydberg gas using rate

equations [17] as discussed in Ref. [16]. Under these assumptions, we obtain the time-dependent fluid limit [19]

$$\frac{\mathbb{E}[X(t)|N]}{n} \rightarrow x(t) = \lambda \int_0^t u(x(s)) ds. \quad (9)$$

After substituting $u(f) = e^{-cf} - (1 - e^{-cf})/c$ into Eq. (9), recalling that initially no particles are excited, and taking the derivative, we obtain the differential system $dx/dt = \lambda\{\exp(-cx(t)) - [1 - \exp(-cx(t))]/c\}$, with $x(0) = 0$, for $x(t)$. This differential system has as its unique solution $x(t) = \ln(1 + c - ce^{-\lambda t})/c$, and in particular, we recover the mean fraction of excitations in the jamming limit by calculating $\lim_{t \rightarrow \infty} x(t) = \ln(1 + c)/c$.

We now validate the model by comparisons with experimental data in Refs. [6,7], which requires us to incorporate the notion of a detector efficiency $\eta \in [0, 1]$ into the model. The detector efficiency η can be interpreted as being the probability that a Rydberg atom is detected. Let $I_i \sim \text{Ber}(\eta)$ denote random variables that indicate whether each i th Rydberg atom is detected. The number of detected Rydberg atoms is then given by $X_D(t) = \sum_{i=1}^{X(t)} I_i$. Assuming the $I_1, \dots, I_{X(t)}$ are independent, calculation shows that $\mathbb{E}[X_D(t)] = \eta \mathbb{E}[X(t)]$, and $\text{Var}[X_D(t)] = \eta^2 \text{Var}[X(t)] + \eta(1 - \eta) \mathbb{E}[X(t)]$, see the Supplemental Material [23]. The detected Mandel Q parameter thus reduces to $Q_D(t) = \eta Q(t)$, see also Ref. [5].

The experiments in Ref. [6] were on excitation volumes said to contain $\rho V = 8 \times 10^3$ ground-state atoms, and with a reported detector efficiency of $\eta = 0.40$. Fitting

$$\mathbb{E}[X_D(t)] \approx \frac{\eta \rho V \ln(1 + c - ce^{-\lambda t})}{c} \quad (10)$$

to measurements of the number of detected excitations as a function of time (Fig. 1(a), Ref. [6]), we obtain an excitation rate of $\lambda = 14$ kHz, and average number of neighbors of $c = 2.7 \times 10^2$. Figure 4 shows strong agreement between theory and experiment.

Lastly, we will compare the model to a histogram of the number of detected dark-state polaritons in Ref. [7]. The

FIG. 4. The average number of detected excitations as a function of time, $\mathbb{E}[X_D(t)]$, fitted to the measurement data in Ref. [6], Fig. 1(a). The fit results in an excitation rate of $\lambda = 14$ kHz, and average number of neighbors of $c = 2.7 \times 10^2$.

histogram displays sub-Poissonian statistics due to a blockade effect that is a result of the dominant Rydberg character of the polaritons. Because of a partial overlap between the excitation laser and the cigar-shaped atomic cloud, we will infer the size of the excitation volume using the density $\rho = 5 \times 10^{17} \text{ m}^{-3}$ [7] as follows. The detector efficiency is reported to be $\eta = 0.4$, and the histogram has a sample mean of $\mathbb{E}[X_D(\infty)] \approx 11$. If we assume that the blockade regions are spherical, and since the blockade radius $r \approx 5 \mu\text{m}$ [7], we find that $c = \frac{4}{3} \rho \pi r^3 \approx 2.6 \times 10^2$. Using the formula for the mean number of detected Rydberg atoms, it follows that $V = c \mathbb{E}[X_D(\infty)] / [\rho \eta \ln(1 + c)] \approx 2.6 \times 10^{-15} \text{ m}^3$. The factor with which the density function of the Poisson distribution is scaled in Ref. [7], Fig. 4(a) is $n_s \approx 315$. Figure 5 now compares the appropriately scaled probability density function of a normal distribution with mean and variance as predicted by the model to the histogram in Ref. [7], Fig. 4(a). The result $Q_D \approx -0.36$ is consistent with their observation that $Q_D = -0.32 \pm 0.04$ in the density range $2 \times 10^{17} \text{ m}^{-3} < \rho < 2 \times 10^{18} \text{ m}^{-3}$.

This Letter derived closed-form expressions for the Mandel Q parameter in limiting large random graphs constructed to model the spatial problem. This approach allowed us to derive explicit formulae for the mean and variance of the number of Rydberg excitations in the jamming limit, that turn out to be functions only of the average number of neighbors within the blockade volume. The comparison to measurement data of Refs. [6,7] shows quantitative agreement between theory and experiment, and we conclude that the model captures blockade effects observed in ultracold Rydberg gases.

Interesting future research would be to further explore the approximating relation between random graphs and spatial problems, particularly because higher-dimensional continuum random sequential adsorption processes are difficult to analyze. To this end, the underlying stochastic

FIG. 5. Histogram (Fig. 4(a), Ref. [7]) of the number of detected dark-state polaritons, together with the appropriately scaled probability density function of a normal distribution with mean $\mathbb{E}[X_D(\infty)]$ and variance $\text{Var}[X_D(\infty)]$. Here, $Q_D(\infty) \approx -0.36$, and the dashed line indicates the Poisson distribution.

recursions can be generalized. In fact, the derivation in Ref. [19] covers a generalization of Eq. (4) in terms of the number of neighbors, and related more general stochastic recursions have been studied for configuration models [24]. These results can potentially be used to describe geometrical features such as an inhomogeneous density and excitation intensity in an atomic cloud, but at increased complexity. Modifying Eq. (4) would also enable analysis of off-resonant excitation effects like the growth of Rydberg aggregates [25,26]. Such approaches can further extend the use of random graphs as an approximation to particle systems that exhibit complicated interactions.

This research was financially supported by an ERC Starting Grant, as well as The Netherlands Organization for Scientific Research (NWO), and is part of the research program of the Foundation for Fundamental Research on Matter (FOM). It was supported by a Wiskundecluster STAR visitor grant. We also acknowledge the European Union H2020 FET Proactive project RySQ (Grant No. 640378). The authors are grateful for the support from Paola Bermolen, Sem Borst, Johan van Leeuwen, and Edgar Vredenbregt.

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