

# Pattern formation of quantum jumps with Rydberg atoms

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(Dated: February 8, 2012)

We study the nonequilibrium dynamics of quantum jumps in a one-dimensional chain of atoms. Each atom is driven on a strong transition to a short-lived state and on a weak transition to a metastable state. We choose the metastable state to be a Rydberg state so that when an atom jumps to the Rydberg state, it inhibits or enhances jumps in the neighboring atoms. This leads to rich spatiotemporal dynamics that are visible in the fluorescence of the strong transition. It also allows one to dissipatively prepare Rydberg crystals.

PACS numbers:

*Introduction.* Pattern formation is a universal phenomenon in nonequilibrium systems [1]. In the presence of driving and dissipation, a system may spontaneously form sustained spatial structures. Pattern formation is usually studied in classical systems, such as fluids, chemical reactions, and granular media. But can a many-body quantum system exhibit pattern formation? This is not obvious, because adding dissipation to a quantum system usually leads to decoherence, so one might not expect any interesting behavior. Also, quantum mechanics is linear, while pattern formation in classical systems requires nonlinearity.

In this paper, we show that a quantum system of Rydberg atoms can form spatial patterns in its fluorescence. A Rydberg atom is an atom excited to a high principal quantum number  $n$ , and there is a strong dipole-dipole interaction between nearby Rydberg atoms. This interaction leads to a wide range of physics, including quantum information processing [2–5], quantum phase transitions [6–9], thermalization of closed quantum systems [6, 10, 11], nonlinear optics [12–15], and dissipative physics [16, 17]. Here, we show that Rydberg atoms also support pattern formation.

Our idea is based on quantum jumps of a three-level atom [18–20]. It is well known that an atom driven strongly to a short-lived state and weakly to a metastable state occasionally jumps to and from the metastable state. The jumps are visible in the fluorescence of the strong transition, which exhibits distinct bright and dark periods [21–23]. Quantum jumps are a good example of how a quantum system far from equilibrium (due to laser driving and spontaneous emission) can have nontrivial dynamics.

Here, we consider a lattice of many three-level atoms, and we let the metastable state be a Rydberg state, so that a jump of one atom affects its neighbors' jumps via the dipole-dipole interaction. This leads to rich spatiotemporal dynamics in a one-dimensional chain of atoms. Depending on the experimental parameters, there is a variety of behaviors. The patterns are observable by imaging the fluorescence of the strong transition. Our scheme can also be used to make Rydberg crystals [6–9].

Previous works studied correlated quantum jumps of atoms in the context of the Dicke model [24, 25]. They

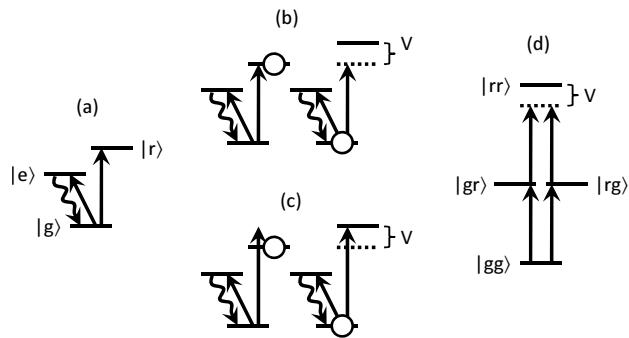


FIG. 1: (a) An atom has a ground state  $|g\rangle$ , short-lived excited state  $|e\rangle$ , and metastable state  $|r\rangle$ , which is chosen to be a Rydberg state. One observes the spontaneous emission from  $|e\rangle$ . (b) The  $|g\rangle \leftrightarrow |r\rangle$  transition is originally on resonance ( $\Delta_r = 0$ ), but when one atom is in  $|r\rangle$ , the other atom is off resonance. (c) The  $|g\rangle \leftrightarrow |r\rangle$  transition is originally off resonance ( $\Delta_r = V$ ), but when one atom is in  $|r\rangle$ , the other atom is on resonance. (d) When  $\Delta_r = 0$ ,  $|rr\rangle$  is weakly coupled to the other states. Note that (b) and (d) are equivalent.

concluded that cooperative effects are very difficult to see experimentally, because the interatomic distance must be much smaller than a wavelength. In contrast, the strong Rydberg interaction here allows the interatomic distance to be much longer than a wavelength. Thus, the atoms develop strong correlations while being individually resolvable.

*Results for a single atom.* We first review quantum jumps in a single atom [18–20]. Consider an atom with three levels: ground state  $|g\rangle$ , short-lived excited state  $|e\rangle$ , and metastable state  $|r\rangle$  [Fig. 1(a)]. We choose the metastable state to be a Rydberg state since Rydberg states have long lifetimes [26]. A laser drives the strong transition  $|g\rangle \leftrightarrow |e\rangle$ , while another drives the weak transition  $|g\rangle \leftrightarrow |r\rangle$ . The strong transition acts as a measurement of whether or not the atom is in  $|r\rangle$ . When the atom is not in  $|r\rangle$ , the atom is repeatedly excited to  $|e\rangle$  and spontaneously emits photons. Occasionally the atom is excited to  $|r\rangle$  and stays there, and the fluorescence from the strong transition turns off. Eventually, the atom returns to  $|g\rangle$ , and the fluorescence turns back on. Thus, the fluorescence signal of the strong transition

exhibits bright and dark periods, and the occurrence of a dark period implies that the atom is in  $|r\rangle$ .

The Hamiltonian for a single atom is ( $\hbar = 1$ )

$$H = \frac{\Omega_e}{2}(|g\rangle\langle e| + |e\rangle\langle g|) + \frac{\Omega_r}{2}(|g\rangle\langle r| + |r\rangle\langle g|) - \Delta_e|e\rangle\langle e| - \Delta_r|r\rangle\langle r|, \quad (1)$$

where  $\Delta_e$  and  $\Omega_e$  are the laser detuning and Rabi frequency of the strong transition, while  $\Delta_r$  and  $\Omega_r$  are the corresponding quantities for the weak transition. In the absence of spontaneous emission, Eq. (1) would completely describe the system. However, the excited states have lifetimes given by their linewidths,  $\gamma_e$  and  $\gamma_r$ .

In the rest of paper, we make the following assumptions on the parameters. To avoid power-broadening on the strong transition, we choose to work in the low-intensity limit,  $\Omega_e \ll \gamma_e$ ; this choice is clarified later. For convenience, we set  $\Delta_e = 0$ , although it may be experimentally useful to set  $\Delta_e < 0$  for continuous laser cooling [27]. We also set  $\gamma_r = 0$ , since the lifetime of the Rydberg state scales as  $n^3$  and hence can be chosen to be arbitrarily long [26]. It is straightforward to extend the analysis to nonzero  $\Delta_e$  and  $\gamma_r$ .

Well-defined jumps appear in the fluorescence signal when a bright period consists of many photons while a dark period consists of the absence of many photons. For a single atom, this happens when  $\Omega_r \ll \Omega_e^2/\gamma_e$  in the case of  $\Delta_r = 0$  [19]. The transition rate from a dark period to a bright period is [20]

$$\Gamma^{D \rightarrow B}(\Delta_r) = \frac{\gamma_e \Omega_e^2 \Omega_r^2}{16\Delta_r^4 + 4\Delta_r^2(\gamma_e^2 - 2\Omega_e^2) + \Omega_e^4}, \quad (2)$$

and the rate from a bright period to a dark period is

$$\Gamma^{B \rightarrow D}(\Delta_r) = \frac{\gamma_e^2 + 4\Delta_r^2}{\gamma_e^2 + 2\Omega_e^2} \Gamma^{D \rightarrow B}(\Delta_r), \quad (3)$$

where  $B$  and  $D$  denote bright and dark periods. Both rates are maximum when  $\Delta_r = 0$  since the strength of the weak transition is maximum there. When  $\Delta_r = 0$ , both rates are approximately  $\gamma_e \Omega_r^2 / \Omega_e^2$ . This depends inversely on  $\Omega_e$ , because increasing  $\Omega_e$  is equivalent to measuring the atomic state more frequently; this inhibits transitions to and from  $|r\rangle$ , similar to the quantum Zeno effect [28].

*Many-body model.* Now we consider a one-dimensional chain of  $N$  three-level atoms. The atoms are coupled via the dipole-dipole interaction between their Rydberg states. In the absence of a static electric field, the interaction decays with the sixth power of distance for long distances [5]; hence, we assume nearest-neighbor interaction. The Hamiltonian is

$$H = \sum_i \left[ \frac{\Omega_e}{2}(|g\rangle\langle e|_i + |e\rangle\langle g|_i) + \frac{\Omega_r}{2}(|g\rangle\langle r|_i + |r\rangle\langle g|_i) - \Delta_r|r\rangle\langle r|_i + V|r\rangle\langle r|_i \otimes |r\rangle\langle r|_{i+1} \right], \quad (4)$$

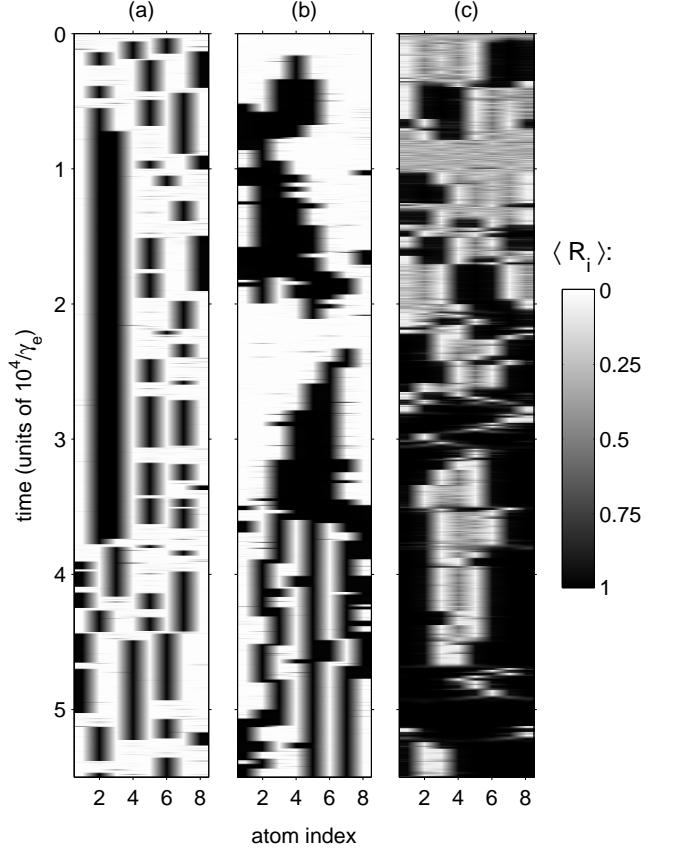


FIG. 2: Quantum trajectory simulation of chain of  $N = 8$  atoms with periodic boundary conditions. The Rydberg population of each atom is plotted vs. time, using color scheme on the right. White color means that the atom is bright and not in the Rydberg state. Black color means that the atom is dark and in the Rydberg state. (a)  $\Omega_e = 0.2\gamma_e$ ,  $\Omega_r = 0.005\gamma_e$ ,  $\Delta_r = 0$ ,  $V = 0.1\gamma_e$ . (b)  $\Omega_e = 0.1\gamma_e$ ,  $\Omega_r = 0.005\gamma_e$ ,  $\Delta_r = V = 0.1\gamma_e$ . (c)  $\Omega_e = \Omega_r = 0.1\gamma_e$ ,  $\Delta_r = 0$ ,  $V = 0.3\gamma_e$ .

where  $V$  is the nearest-neighbor interaction and we assume periodic boundary conditions. The interatomic distance is assumed to be large enough so that the fluorescence from each atom is resolvable *in situ* on a camera [29].

To demonstrate the rich spatiotemporal dynamics of the many-body system, Fig. 2 shows simulations of a chain of  $N = 8$  atoms, generated using the method of quantum trajectories [30, 31]. Each trajectory simulates a single experimental run. We have plotted the Rydberg population of each atom, i.e., the expectation value of  $R_i \equiv |r\rangle\langle r|_i$ . The atoms undergo quantum jumps, and the Rydberg interaction clearly leads to spatial correlations in the fluorescence.

There are different types of collective dynamics depending on the parameters. In Fig. 2(a)-(b),  $\Omega_r \ll \Omega_e^2/\gamma_e$ , so each atom by itself would exhibit quantum jumps. In Fig. 2(a) ( $\Delta_r = 0$ ), a dark period usually does not spread to the neighboring atoms. But once in

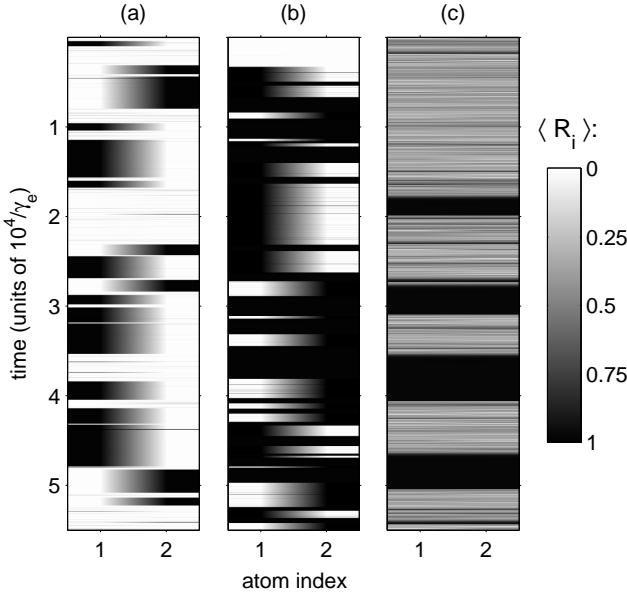


FIG. 3: Quantum trajectory simulation of  $N = 2$  atoms. The Rydberg population of each atom is plotted vs. time, using color scheme on the right. White color means that the atom is bright and not in the Rydberg state. Black color means that the atom is dark and in the Rydberg state. Parameters are the same as in Fig. 2: (a)  $\Omega_e = 0.2\gamma_e$ ,  $\Omega_r = 0.005\gamma_e$ ,  $\Delta_r = 0$ ,  $V = 0.1\gamma_e$ . (b)  $\Omega_e = 0.2\gamma_e$ ,  $\Omega_r = 0.005\gamma_e$ ,  $\Delta_r = V = 0.1\gamma_e$ . (c)  $\Omega_e = \Omega_r = 0.1\gamma_e$ ,  $\Delta_r = 0$ ,  $V = 0.3\gamma_e$ .

a while, there are two dark atoms in a row ( $BDBB$ ) and they both stay dark for a long time. In Figs. 2(b) ( $\Delta_r = V$ ), there are two types of behavior. Sometimes, dark periods spread across the chain so that many consecutive atoms are dark at the same time. Other times, an alternating  $DBDB$  pattern appears. Over time, the system switches between these two types of behaviors. In Fig. 2(c) ( $\Omega_r = \Omega_e$ ,  $\Delta_r = 0$ ), the atoms tend to turn dark or bright in groups of two or three, and sometimes all the atoms are dark. The existence of jumps here is surprising because a single atom would not exhibit jumps for these parameters.

To understand the results for  $N = 8$ , it is instructive to consider the simpler case of  $N = 2$  atoms. Figure 3 shows quantum trajectory simulations for  $N = 2$ ; note the similarity with Fig. 2. We have analytically solved the  $N = 2$  case. The details are in the supplemental material [32], and here we summarize the results and provide intuition. There are two general cases: (i)  $\Omega_r \ll \Omega_e^2/\gamma_e$  and (ii)  $\Omega_r = \Omega_e$ ,  $\Delta_r = 0$ , distinguished by whether a single atom would exhibit jumps.

*Case of  $\Omega_r \ll \Omega_e^2/\gamma_e$ .* For these parameters, an atom by itself would exhibit jumps. Let the two atoms be labelled 1 and 2. If atom 1 is in  $|r\rangle$ , then according to Eq. (4), atom 2 effectively sees a laser detuning of  $\Delta_r - V$ . But if atom 1 is not in  $|r\rangle$ , then atom 2 sees the original detuning  $\Delta_r$ . Whether atom 1 is in  $|r\rangle$  depends on whether it is in a dark period. This suggests that the

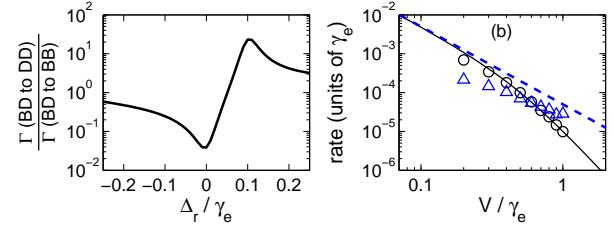


FIG. 4: Statistics for  $N = 2$  atoms. (a) Ratio of  $\Gamma^{BD \rightarrow DD}$  to  $\Gamma^{BD \rightarrow BB}$  for  $\Omega_e = 0.2\gamma_e$ ,  $\Omega_r = 0.005\gamma_e$ ,  $V = 0.1\gamma_e$ . (b) Jump rates for  $\Omega_r = \Omega_e = 0.1\gamma_e$ ,  $\Delta_r = 0$ .  $\Gamma^{DD \rightarrow BB}$ : analytical result (black, solid line) and numerical data (black circles).  $\Gamma^{BB \rightarrow DD}$ : analytical upper bound (blue, dashed line) and numerical data (blue triangles).

jump rates for atom 2 are the same as for a single atom [Eqs. (2)-(3)], except with an effective detuning that depends on whether atom 1 is in a bright or dark period at the moment. In the supplemental material [32], we use a more careful analysis to show that this is indeed correct in the limit of small  $\Omega_r$ . Thus, the transition rates for two atoms are

$$\Gamma^{BB \rightarrow BD}(\Delta_r) = \Gamma^{BB \rightarrow DB}(\Delta_r) = \Gamma^{B \rightarrow D}(\Delta_r) \quad (5)$$

$$\Gamma^{BD \rightarrow BB}(\Delta_r) = \Gamma^{DB \rightarrow BB}(\Delta_r) = \Gamma^{D \rightarrow B}(\Delta_r) \quad (6)$$

$$\Gamma^{BD \rightarrow DD}(\Delta_r) = \Gamma^{DB \rightarrow DD}(\Delta_r) = \Gamma^{B \rightarrow D}(\Delta_r - V) \quad (7)$$

$$\Gamma^{DD \rightarrow BD}(\Delta_r) = \Gamma^{DD \rightarrow DB}(\Delta_r) = \Gamma^{D \rightarrow B}(\Delta_r - V) \quad (8)$$

An insightful quantity is the ratio  $\Gamma^{BD \rightarrow DD}/\Gamma^{BD \rightarrow BB}$ , which indicates how often the atoms are in  $DD$  relative to  $BB$ . As shown in Fig. 4(a), the ratio is minimum at  $\Delta_r = 0$  and maximum at  $\Delta_r = V$ .

The minimum at  $\Delta_r = 0$  is due to the blockade effect: when atom 1 is in  $|r\rangle$ , it shifts the Rydberg level of atom 2 off resonance so that atom 2 is prevented from jumping to  $|r\rangle$  [Fig. 1(b)]. Thus, the atoms switch between  $BB$ ,  $BD$ , and  $DB$ ; they are almost never in  $DD$ . In other words, there is at most one dark atom at a time [Fig. 3(a)].

The maximum at  $\Delta_r = V$  is due to the opposite effect: the laser is originally off-resonant, but when atom 1 happens to jump to  $|r\rangle$ , it brings the Rydberg level of atom 2 on resonance, encouraging atom 2 to jump to  $|r\rangle$  [Fig. 1(c)]. Thus, the atoms switch between  $DD$ ,  $BD$ , and  $DB$ ; they are almost never in  $BB$ , except for the initial transient. When the atoms are in  $DD$ , there is an equal chance to go to  $BD$  or  $DB$ . Thus, the dark spot appears to do a random walk between the two atoms [Fig. 3(b)].

The above considerations also apply to larger  $N$ . In general, the transition rates for atom  $i$  are given by Eqs. (2)-(3) but with an effective detuning that depends on its neighbors' current state:  $\Delta_r - V(\langle R_{i-1} \rangle + \langle R_{i+1} \rangle)$ . When  $\Delta_r = 0$ , the blockade effect prevents dark periods from spreading [Fig. 2(a)]. However, if there happens to be two dark atoms in a row ( $BDBB$ ), the dark atoms are effectively off resonance and are unlikely to turn bright; thus, they stay dark for a long time. On

the other hand, when  $\Delta_r = V$ , the anti-blockade effect encourages dark periods to spread across the chain [Fig. 2(b)]. However, sometimes an alternating pattern develops (*DBDB*); this is because when an atom is bright while both neighbors are dark, it is effectively off resonance and is unlikely to turn dark.

*Case of  $\Omega_r = \Omega_e$ ,  $\Delta_r = 0$ .* For these parameters, an atom by itself would not exhibit jumps because of the absence of a weak transition. The existence of jumps for two atoms is solely due to the dipole-dipole interaction, which causes  $|gr\rangle \leftrightarrow |rr\rangle$  and  $|rg\rangle \leftrightarrow |rr\rangle$  to become off-resonant and thus weak transitions [Fig. 1(d)]. Since  $|rr\rangle$  is metastable, occasionally the system jumps to and from  $|rr\rangle$ . When the system is in  $|rr\rangle$ , the atoms do not fluoresce. When the system is not in  $|rr\rangle$ , the wavefunction rapidly oscillates among the other eigenstates so that both atoms fluoresce from  $|e\rangle$ . Thus, the system switches between *BB* and *DD* [Fig. 3(c)]. The rates are [32]

$$\Gamma^{DD \rightarrow BB} = \frac{\gamma_e \Omega^4}{2V^2(\gamma_e^2 + 4V^2)} \quad (9)$$

$$\Gamma^{BB \rightarrow DD} \leq \frac{\Omega^4}{2\gamma_e V^2}, \quad (10)$$

where  $\Omega \equiv \Omega_r = \Omega_e$ . The inequality for  $\Gamma^{BB \rightarrow DD}$  is due to incomplete knowledge of the wave function after a photon emission. Equations (9)-(10) agree well with quantum trajectory simulations [Fig. 4(b)]. Both rates are inversely related to  $V$ , since the weak transitions become weaker as  $V$  increases. The condition for well-defined jumps is roughly  $\Omega \ll 2V$ .

A larger chain seems to have similar behavior [Fig. 2(c)]. The atoms tend to turn dark or bright simultaneously with their neighbors. However, the dynamics are more complex due to the presence of two neighbors.

*Experimental considerations.* Our scheme can be implemented using atoms trapped in an optical lattice. For example, one can use  $^{87}\text{Rb}$ , which has a strong  $5S - 5P$  transition with linewidth  $\gamma_e/2\pi = 6$  MHz [27]. Suppose one chooses the  $60S$  Rydberg state, which can be reached via a two-photon transition. For a lattice spacing of

$7 \mu\text{m}$ , the nearest-neighbor interaction is  $V = 0.2\gamma_e$  [33]. The lifetime of that Rydberg state is  $250 \mu\text{s}$  at  $0 \text{ K}$  [34]; in other words,  $\gamma_r \approx \gamma_e/10^4$ . Transitions due to black-body radiation can be minimized by working at cryogenic temperatures. Also, the  $nS$  states have negligible losses from trap-induced photoionization [35, 36]. The trapping of Rydberg atoms in optical lattices was recently demonstrated in Refs. [36, 37].

There is an important constraint on the experimental parameters: the interaction  $V$  should be much less than the trap depth, or else the repulsive interaction will push the atoms out of the lattice. Since a trap depth of 10 MHz is possible [36], we require  $V \ll \gamma_e$ . Then to avoid broadening the strong transition [19] and smearing out the effect of  $V$ , we choose  $\Omega_e \ll \gamma_e$ , as stated above.

Quantum jumps can also be used to prepare a Rydberg crystal, which is a regular arrangement of Rydberg atoms among ground-state atoms [6–9]. Rydberg crystals are important for studying many-body physics and generating non-classical light, but are nontrivial to make. Here, one would let the atoms jump to and from the Rydberg state while monitoring the fluorescence until the desired crystal is obtained. The parameters can be optimized for the desired crystal type. This is in the spirit of recent works that use dissipation to prepare nontrivial quantum states [38, 39].

*Conclusion.* Thus, quantum jumps of Rydberg atoms lead to interesting spatiotemporal dynamics. The next step is to see what happens in larger systems, especially in higher dimensions: does the Rydberg interaction lead to long-range patterns in a large system? It would also be interesting to see what happens with other interaction types (long range or anisotropic) or when the atoms are free to move instead of being fixed on a lattice. Finally, one should look for other quantum many-body systems that exhibit pattern formation. Perhaps nonequilibrium quantum systems share some universal features as is the case with nonequilibrium classical systems [1].

We thank H. Häffner and H. Weimer for useful discussions. This work was supported by NSF Grant No. DMR-1003337.

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## SUPPLEMENTAL MATERIAL

This supplement provides details of the jump-rate calculations. Appendix A reviews the derivation for one atom. We essentially reproduce the derivation in Refs. [1–3], because we need to refer back to it later, and it is convenient to see it in our notation. Appendix B and C derive the results for two atoms: Appendix B covers the case of  $\Omega_r \ll \Omega_e^2/\gamma_e$ , while Appendix C covers the case of  $\Omega_r = \Omega_e$ ,  $\Delta_r = 0$ . In general, we use the “quantum-trajectory” approach, which is based on the wave function, to account for spontaneous emission. An alternative approach is to use the density matrix as in Refs. [4, 5].

### Appendix A: Review of one-atom case

When an atom exhibits quantum jumps, the fluorescence signal has bright periods, in which the photons are closely spaced in time, and dark periods, in which no photons are emitted for a while. The goal is to calculate the transition rate from a bright period to a dark period and vice versa. The important quantity is the time interval between successive emissions [1]. During a bright period, the intervals are short, but a dark period is an exceptionally long interval. Suppose one has the function  $P_0(t)$ , which is the probability that the atom has not emitted a

photon by time  $t$ , given that it emitted at time 0.  $P_0(t)$  decreases monotonically as  $t$  increases. When the parameters are such that there are well-defined quantum jumps,  $P_0(t)$  decreases rapidly to a small value for small  $t$ , but has a long tail for large  $t$ . This reflects the fact that the time between emissions is usually short (bright period), but once in a while it is very long (dark period). Note that each emission is an independent event, due to the fact that the wave function always returns to  $|g\rangle$  after an emission.

We write  $P_0(t) = P_{\text{short}}(t) + P_{\text{long}}(t)$  to separate the short and long time-scale parts. The long tail is given by  $P_{\text{long}}(t) = p \exp(-\Gamma^{D \rightarrow B} t)$ , where  $p$  is the probability that a given interval is long enough to be a dark period, and  $\Gamma^{D \rightarrow B}$  is the transition rate from a dark period to a bright period. In other words,  $1/\Gamma^{D \rightarrow B}$  is the average duration of a dark period.

To calculate  $P_0(t)$ , we follow the evolution of the wave function  $|\psi(t)\rangle$ , given that the atom has not emitted a photon yet. This is found by evolving  $|\psi(t)\rangle$  with a non-Hermitian Hamiltonian  $H_{\text{eff}} = H - i\frac{\gamma_e}{2}|e\rangle\langle e|$ . The non-Hermitian term accounts for the population that emits a photon, hence dropping out of consideration [1]. Thus,  $P_0(t) = \langle\psi(t)|\psi(t)\rangle$ .

In the basis  $\{|g\rangle, |e\rangle, |r\rangle\}$ , the matrix form of  $H_{\text{eff}}$  is

$$H_{\text{eff}} = \begin{pmatrix} 0 & \frac{\Omega_e}{2} & \frac{\Omega_r}{2} \\ \frac{\Omega_e}{2} & -\frac{i\gamma_e}{2} & 0 \\ \frac{\Omega_r}{2} & 0 & -\Delta_r \end{pmatrix}. \quad (\text{A1})$$

We want to solve the differential equation  $i\frac{d}{dt}|\psi(t)\rangle = H_{\text{eff}}|\psi(t)\rangle$  given the initial condition  $|\psi(0)\rangle = |g\rangle$ . The general solution is  $|\psi(t)\rangle = \sum_n c_n e^{-i\lambda_n t} |u_n\rangle$ , where  $\lambda_n$  and  $|u_n\rangle$  are the eigenvalues and eigenvectors of  $H_{\text{eff}}$ , and  $c_n$  are determined from the initial condition  $|g\rangle = \sum_n c_n |u_n\rangle$ .

We calculate the eigenvalues and eigenvectors perturbatively in  $\Omega_r$ , which is assumed to be small. (Note that since  $H_{\text{eff}}$  is non-Hermitian, perturbation theory is different from the usual Hermitian case [6].) All three eigenvalues have negative imaginary parts, which leads to the nonunitary decay. It turns out that the imaginary part of one of the eigenvalues, which we call  $\lambda_3$ , is much less negative than the other two. This means that the  $|u_1\rangle$  and  $|u_2\rangle$  components in  $|\psi(t)\rangle$  decay much faster than the  $|u_3\rangle$  component. After a long time without a photon emission,  $|\psi(t)\rangle$  contains only  $|u_3\rangle$ . Thus,  $\lambda_3$  corresponds to the long tail of  $P_0(t)$ .

To second order in  $\Omega_r$  [1, 2],

$$\lambda_3 = -\Delta_r + \frac{\Omega_r^2(-2\Delta_r + i\gamma_e)}{8\Delta_r^2 - 2\Omega_e^2 - 4i\gamma_e\Delta_r}. \quad (\text{A2})$$

To first order in  $\Omega_r$ ,

$$\begin{aligned} |u_3\rangle &= \frac{\Omega_r(-2\Delta_r + i\gamma_e)}{4\Delta_r^2 - \Omega_e^2 - 2i\gamma_e\Delta_r}|g\rangle \\ &\quad + \frac{\Omega_e\Omega_r}{4\Delta_r^2 - \Omega_e^2 - 2i\gamma_e\Delta_r}|e\rangle + |r\rangle \end{aligned} \quad (\text{A3})$$

$$c_3 = \frac{\Omega_r(-2\Delta_r + i\gamma_e)}{4\Delta_r^2 - \Omega_e^2 - 2i\gamma_e\Delta_r}. \quad (\text{A4})$$

Since  $|u_3\rangle$  consists mainly of  $|r\rangle$ , the occurrence of a dark period implies, as expected, that the atom is in  $|r\rangle$ . (However, note that the atom is not completely in  $|r\rangle$ . In fact, the dark period ends when the small  $|e\rangle$  component in  $|u_3\rangle$  decays and emits a photon [3].)

We can now construct  $P_{\text{long}}(t)$ :

$$p = |c_3|^2 \quad (\text{A5})$$

$$= \frac{\Omega_r^2(\gamma_e^2 + 4\Delta_r^2)}{16\Delta_r^4 + 4\Delta_r^2(\gamma_e^2 - 2\Omega_e^2) + \Omega_e^4} \quad (\text{A6})$$

$$\Gamma^{D \rightarrow B} = -2 \operatorname{Im} \lambda_3 \quad (\text{A7})$$

$$= \frac{\gamma_e\Omega_e^2\Omega_r^2}{16\Delta_r^4 + 4\Delta_r^2(\gamma_e^2 - 2\Omega_e^2) + \Omega_e^4}. \quad (\text{A8})$$

Then instead of finding  $P_{\text{short}}(t)$  explicitly, we use a short cut [3]. During a bright period, there is negligible population in  $|r\rangle$ , so the atom is basically a two-level atom driven by a laser with Rabi frequency  $\Omega_e$ . Thus, to lowest order in  $\Omega_r$ , the emission rate  $\Gamma_{\text{short}}$  during a bright period is the same as a two-level atom [7]:

$$\Gamma_{\text{short}} = \frac{\gamma_e\Omega_e^2}{\gamma_e^2 + 2\Omega_e^2}. \quad (\text{A9})$$

However, each emission in a bright period has a small probability  $p$  of taking a long time, in which case the bright period ends. Thus, the transition rate from a bright period to a dark period is

$$\Gamma^{B \rightarrow D} = p \Gamma_{\text{short}} \quad (\text{A10})$$

$$= \frac{\gamma_e^2 + 4\Delta_r^2}{\gamma_e^2 + 2\Omega_e^2} \Gamma^{D \rightarrow B}. \quad (\text{A11})$$

The jumps are well-defined when a bright or dark period is much longer than the typical emission time during a bright period:  $\Gamma^{B \rightarrow D}, \Gamma^{D \rightarrow B} \ll \Gamma_{\text{short}}$ . When  $\Delta_r = 0$  and  $\Omega_e \ll \gamma_e$ , this condition becomes  $\Omega_r \ll \Omega_e^2/\gamma_e$ .

## Appendix B: Two atoms, $\Omega_r \ll \Omega_e^2/\gamma_e$

When  $\Omega_r \ll \Omega_e^2/\gamma_e$ , a single atom would exhibit quantum jumps. In the case of two interacting atoms, each one still undergoes quantum jumps, but the jump rates of each depend on the current state of the other atom. The goal is to calculate, to lowest order in  $\Omega_r$ , the transition rates among the possible states:  $BB$ ,  $BD$ ,  $DB$ , and  $DD$ .

Suppose for a moment that the interaction strength  $V = 0$ . Then each atom jumps independently, and the jump rates are the same as the single-atom case [Eqs. (A8) and (A11)].

Then let  $V \neq 0$ . Due to its form, the Rydberg interaction only affects the state  $|rr\rangle$ . When the atoms are in  $BB$ ,  $BD$ , and  $DB$ , there is negligible population in  $|rr\rangle$ , so the interaction has negligible effect on the transitions among  $BB$ ,  $BD$ , and  $DB$ . So to lowest order in  $\Omega_r$ , those transition rates are the same as when  $V = 0$ . Thus, we can immediately write down:

$$\Gamma^{BB \rightarrow BD} = \Gamma^{BB \rightarrow DB} = \Gamma^{B \rightarrow D} \quad (\text{B1})$$

$$\Gamma^{BD \rightarrow BB} = \Gamma^{DB \rightarrow BB} = \Gamma^{D \rightarrow B}. \quad (\text{B2})$$

The remaining task is to calculate the transition rates that involve  $DD$ :  $\Gamma^{BD \rightarrow DD}$ ,  $\Gamma^{DB \rightarrow DD}$ ,  $\Gamma^{DD \rightarrow BD}$ , and  $\Gamma^{DD \rightarrow DB}$ .

To calculate these rates, we use an approach similar to Appendix A. Suppose the atoms are initially in  $BD$ , i.e., atom 1 is fluorescing while atom 2 is not. We are interested in the time interval between an emission by atom 1 and a subsequent emission by either atom 1 or 2. Usually the intervals are short since atom 1 is in a bright period. But once in a while, there is a very long interval, which means that atom 1 has become dark and the atoms are in  $DD$ . If the long interval ends due to an emission by atom 1, the atoms end up in  $BD$ ; if it is due to an emission by atom 2, the atoms end up in  $DB$ . We want to calculate  $P_0(t)$ , which is the probability that neither atom has emitted a photon by time  $t$ , given that atom 1 emitted at time 0 and also given that atom 2 started dark.  $P_0(t)$  has a long tail corresponding to time spent in  $DD$ .

We write  $P_0(t) = P_{\text{short}}(t) + P_{\text{long}}(t)$  to separate the short and long time-scale parts. The long tail is given by  $P_{\text{long}}(t) = p \exp(-2\Gamma^{DD \rightarrow BD}t)$ , where  $p$  is the probability that a given interval is long enough to be a  $DD$  period.  $2\Gamma^{DD \rightarrow BD}$  is the total transition rate out of  $DD$  since  $\Gamma^{DD \rightarrow BD} = \Gamma^{DD \rightarrow DB}$ .

To evolve the wave function in the absence of an emission, we use the non-Hermitian Hamiltonian  $H_{\text{eff}} = H - i\frac{\gamma_e}{2}(|e\rangle\langle e|_1 + |e\rangle\langle e|_2)$ , where  $H$  is the two-atom Hamiltonian. We want to solve the differential equation  $i\frac{d}{dt}\langle\psi(t)\rangle = H_{\text{eff}}|\psi(t)\rangle$  in order to find  $P_0(t) = \langle\psi(t)|\psi(t)\rangle$ .

The question now is what initial condition to use. Since atom 1 is assumed to emit at time 0, it is in  $|g\rangle$ . Also, as discussed above, during a  $BD$  period, there is very little population in  $|rr\rangle$ , so the interaction has negligible effect on the dynamics. To first order in  $\Omega_r$ , atom 2's wave function is the same as that of a single atom in a dark period [Eq. (A3)]. So the initial condition of the two-atom system is:

$$\begin{aligned} |\psi(0)\rangle &= \frac{\Omega_r(-2\Delta_r + i\gamma_e)}{4\Delta_r^2 - \Omega_e^2 - 2i\gamma_e\Delta_r}|gg\rangle \\ &\quad + \frac{\Omega_r\Omega_e}{4\Delta_r^2 - \Omega_e^2 - 2i\gamma_e\Delta_r}|ge\rangle + |gr\rangle. \end{aligned} \quad (\text{B3})$$

The general solution to the differential equation is  $|\psi(t)\rangle = \sum_n c_n e^{-i\lambda_n t} |u_n\rangle$ , where  $\lambda_n$  and  $|u_n\rangle$  are the eigenvalues and eigenvectors of  $H_{\text{eff}}$ , which is a  $9 \times 9$  matrix.  $c_n$  are determined from the initial condition  $|\psi(0)\rangle = \sum_n c_n |u_n\rangle$ .

We calculate the eigenvalues and eigenvectors perturbatively in  $\Omega_r$ . All nine eigenvalues have negative imaginary parts, which leads to the nonunitary decay. It turns out that the imaginary part of one of the eigenvalues, which we call  $\lambda_9$ , is much less negative than the other eight. This means that the other eight components of  $|\psi(t)\rangle$  decay much faster than the  $|u_9\rangle$  component. After a long time without a photon emission,  $|\psi(t)\rangle$  contains only  $|u_9\rangle$ . Thus,  $\lambda_9$  corresponds to the long tail of  $P_0(t)$ .

To second order in  $\Omega_r$ ,

$$\lambda_9 = -2\Delta_r + V + \frac{\Omega_r^2(-2\Delta'_r + i\gamma_e)}{4\Delta'^2 - \Omega_e^2 - 2i\gamma_e\Delta'_r}, \quad (\text{B4})$$

where  $\Delta'_r = \Delta_r - V$ . To first order in  $\Omega_r$ ,

$$\begin{aligned} |u_9\rangle &= \frac{\Omega_r(-2\Delta'_r + i\gamma_e)}{4\Delta'^2 - \Omega_e^2 - 2i\gamma_e\Delta'_r} |gr\rangle \\ &\quad + \frac{\Omega_e\Omega_r}{4\Delta'^2 - \Omega_e^2 - 2i\gamma_e\Delta'_r} |er\rangle \\ &\quad + \frac{\Omega_r(-2\Delta'_r + i\gamma_e)}{4\Delta'^2 - \Omega_e^2 - 2i\gamma_e\Delta'_r} |rg\rangle \\ &\quad + \frac{\Omega_e\Omega_r}{4\Delta'^2 - \Omega_e^2 - 2i\gamma_e\Delta'_r} |re\rangle + |rr\rangle \quad (\text{B5}) \\ c_9 &= \frac{\Omega_r(-2\Delta'_r + i\gamma_e)}{4\Delta'^2 - \Omega_e^2 - 2i\gamma_e\Delta'_r}. \quad (\text{B6}) \end{aligned}$$

Note that  $|u_9\rangle$  consists mainly of  $|rr\rangle$ , since it corresponds to a *DD* period.

We can now construct  $P_{\text{long}}(t)$ :

$$p = |c_9|^2 \quad (\text{B7})$$

$$= \frac{\Omega_r^2(\gamma_e^2 + 4\Delta'^2)}{16\Delta'^4 + 4\Delta'^2(\gamma_e^2 - 2\Omega_e^2) + \Omega_e^4} \quad (\text{B8})$$

$$\Gamma^{DD \rightarrow BD} = \Gamma^{DD \rightarrow DB} = -\text{Im } \lambda_9 \quad (\text{B9})$$

$$= \frac{\gamma_e\Omega_e^2\Omega_r^2}{16\Delta'^4 + 4\Delta'^2(\gamma_e^2 - 2\Omega_e^2) + \Omega_e^4} \quad (\text{B10})$$

To calculate  $\Gamma^{BD \rightarrow DD}$ , we use the short cut from Appendix A. Since atom 1 is bright, it has negligible population in  $|r\rangle$ , so its emission rate  $\Gamma_{\text{short}}$  is the same as a two-level atom [Eq. (A9)]. Each emission has probability  $p$  of being long enough to be a dark period.

$$\Gamma^{BD \rightarrow DD} = \Gamma^{DB \rightarrow DD} = p \Gamma_{\text{short}} \quad (\text{B11})$$

$$= \frac{\gamma_e^2 + 4\Delta_r^2}{\gamma_e^2 + 2\Omega_e^2} \Gamma^{DD \rightarrow BD}. \quad (\text{B12})$$

Note the similarity between Eqs. (B10) and (A8) and between Eqs. (B12) and (A11).

### Appendix C: Two atoms, $\Omega_r = \Omega_e$ , $\Delta_r = 0$

For these parameters, a single atom would not exhibit quantum jumps. The existence of jumps for two atoms is solely due to the interaction. To calculate the jump rates, we use an approach similar to Appendices A and B, but there are some important differences.

We are interested in the time intervals between photon emissions of either atom. We want to calculate  $P_0(t)$ , which is the probability that neither atom has emitted a photon by time  $t$ , given that atom 1 emitted at time 0. (Alternatively, one could let atom 2 emit at time 0.) We write  $P_0(t) = P_{\text{short}}(t) + P_{\text{long}}(t)$  to separate the short and long time-scale parts. As in Appendix B, we want to solve the differential equation  $i\frac{d}{dt}|\psi(t)\rangle = H_{\text{eff}}|\psi(t)\rangle$  in order to find  $P_0(t) = \langle\psi(t)|\psi(t)\rangle$ .

Before discussing what initial condition to use, we first calculate the eigenvalues  $\lambda_n$  and eigenvectors  $|u_n\rangle$  of  $H_{\text{eff}}$ . We define  $\Omega \equiv \Omega_r = \Omega_e$  and do perturbation theory in  $\Omega$ , which is assumed to be small. As in Appendix B, all nine eigenvalues have negative imaginary parts, which leads to the nonunitary decay. The imaginary part of one of the eigenvalues, which we call  $\lambda_9$ , is much less negative than the other eight. This means that the other eight components of  $|\psi\rangle$  decay much faster than the  $|u_9\rangle$  component. Thus,  $\lambda_9$  corresponds to the long tail of  $P_0(t)$ . To fourth order in  $\Omega$ ,

$$\lambda_9 = V + \frac{\Omega^2}{2V} + \frac{\Omega^4(2V - i\gamma_e)}{4V^2(\gamma_e^2 + 4V^2)}. \quad (\text{C1})$$

To first order in  $\Omega$ ,

$$|u_9\rangle = \frac{\Omega}{2V} |gr\rangle + \frac{\Omega}{2V} |rg\rangle + |rr\rangle, \quad (\text{C2})$$

which consists mainly of  $|rr\rangle$ , reflecting the fact that if both atoms have not emitted for a while, they are in a *DD* period.

Now it turns out that the real parts of the other eight eigenvalues have very different values, which causes the wave function to oscillate rapidly among the eight eigenvectors. Thus, after atom 1 emits a photon, the short time-scale behavior consists of rapid oscillation among the eight eigenvectors, and each atom's  $|e\rangle$  population fluctuates a lot. The time scale of the oscillation is faster than the typical photon emission rate, so both atoms are equally likely to emit next. Thus, the atoms can either be in *BB* or *DD*. When in *BB*, both atoms emit, and the time interval between emissions is relatively short. But once in a while, it takes a very long time for the next photon to be emitted, which means that the atoms are in *DD*. Once the long interval ends, the atoms go back to *BB*.

The rapid oscillation during *BB* makes it impossible to choose a unique initial condition  $|\psi(0)\rangle$ , because each time atom 1 emits, atom 2's wave function is different. To account for this ignorance, we let atom 2's wave function be completely arbitrary:

$$|\psi(0)\rangle = a_1 |gg\rangle + a_2 |ge\rangle + a_3 |gr\rangle. \quad (\text{C3})$$

Normalization requires  $|a_1|^2 + |a_2|^2 + |a_3|^2 = 1$ , but  $a_1, a_2, a_3$  are otherwise unknown. Despite the incomplete knowledge, we can still obtain a useful bound on  $\Gamma^{BB \rightarrow DD}$ .

The general solution to the differential equation  $i\frac{d}{dt}|\psi(t)\rangle = H_{\text{eff}}|\psi(t)\rangle$  is  $|\psi(t)\rangle = \sum_n c_n e^{-i\lambda_n t}|u_n\rangle$ , where  $c_n$  are determined from the initial condition  $|\psi(0)\rangle = \sum_n c_n|u_n\rangle$ . To first order in  $\Omega$ ,

$$c_9 = a_3 \frac{\Omega}{2V}. \quad (\text{C4})$$

Given the above results, we can now construct  $P_{\text{long}}(t) = p \exp(-\Gamma^{DD \rightarrow BB} t)$ , where  $p$  is the probability that a given interval is long enough to be a *DD* period, and  $\Gamma^{DD \rightarrow BB}$  is the transition rate from *DD* to *BB*:

$$p = |c_9|^2 \leq \frac{\Omega^2}{4V^2} \quad (\text{C5})$$

$$\Gamma^{DD \rightarrow BB} = -2 \operatorname{Im} \lambda_9 \quad (\text{C6})$$

$$= \frac{\gamma_e \Omega^4}{2V^2(\gamma_e^2 + 4V^2)}. \quad (\text{C7})$$

The inequality for  $p$  reflects the incomplete knowledge of

the initial wave function.

To calculate  $\Gamma^{BB \rightarrow DD}$ , we have to first calculate  $\Gamma_{\text{short}}$ , which is the total emission rate of both atoms during a *BB* period. We approximate  $\Gamma_{\text{short}}$  using the emission rate in the absence of the  $|g\rangle \leftrightarrow |r\rangle$  transition, like in Eq. (A9):

$$\Gamma_{\text{short}} \approx \frac{2\gamma_e \Omega^2}{\gamma_e^2 + 2\Omega^2}. \quad (\text{C8})$$

However, since the  $|g\rangle \leftrightarrow |r\rangle$  transition is not weak, the above approximation to  $\Gamma_{\text{short}}$  is usually an upper bound. Now we can calculate:

$$\Gamma^{BB \rightarrow DD} = p \Gamma_{\text{short}} \leq \frac{\Omega^4}{2\gamma_e V^2}. \quad (\text{C9})$$

The jumps are well-defined when a *BB* period consists of many emissions while a *DD* period consists of the absence of many emissions:  $\Gamma^{BB \rightarrow DD}, \Gamma^{DD \rightarrow BB} \ll \Gamma_{\text{short}}$ . Roughly speaking, this happens when

$$\Omega \ll 2V. \quad (\text{C10})$$

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