Interplay of the dipole blockade and interaction-induced dephasing in Rydberg single-photon sources

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Interactions between Rydberg atoms can result in a dipole blockade for which the probability P_n to have n atomic excitations is reduced significantly when $n \geqslant 2$. Ideally, this allows one to create a single collective Rydberg excitation in an atomic ensemble with $P_1 = 1$. A single-photon source is realized by mapping this atomic excitation into a propagating light field. Even if $P_n \neq 0$ for $n \geqslant 2$, a single-photon source can be approximated if interaction-induced dephasing damps the contributions from multiply excited collective states into the preferred spatial field mode. Two quantities that can be used as figures of merit for these single-photon sources are the second-order correlation function $g^{(2)}$ associated with the phase-matched field emitted by the sample and the second-order correlation function $g^{(A)}$ associated with the atoms. Here we demonstrate that interaction-induced dephasing can lead to significant differences between $g^{(2)}$ and $g^{(A)}$ even if $P_2 \ll P_1 \approx 1$. Theoretical expressions are derived for these quantities and it is shown that $g^{(2)} \leqslant g^{(A)}$. It is also shown that there is a distinct advantage for minimizing $g^{(2)}$ and $g^{(A)}$ by using adiabatic pulsed fields rather than constant amplitude fields to excite the ensemble. These results maybe useful for optimizing Rydberg single-photon sources.

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I. INTRODUCTION

Proposed originally by Lukin *et al.* [1], the dipole blockade has become an important component of quantum information protocols involving Rydberg atoms. When fully operational, the dipole blockade inhibits all but a single collective Rydberg excitation in an ensemble of atoms following excitation by pulsed optical field(s) that are resonant with the ground to Rydberg transition. The strong interactions between Rydberg atoms produce level shifts that prevent multiple excitations in the ensemble. Experimental confirmation of the Rydberg blockade has been reported for both two-atom [2] and many-atom [3] systems. If the blockade is not perfect, there could be more than one Rydberg excitation.

Let us denote the probability of n excitations in the ensemble by P_n. We are interested in the limit that $P_2 \ll P_1$ and assume that the incident field(s) creates at most two excitations in an ensemble of N atoms. In other words, we assume that the state vector can be written as

$$|\psi(t)\rangle = c_0(t)|g\rangle + \sum_{j=1}^{N} c_j(t)|j\rangle e^{-i\omega_0 t} + \sum_{\substack{j,j'=1 \\ j'>j}}^{N} c_{jj'}(t)|jj'\rangle e^{-2i\omega_0 t}, \tag{1}$$

where $|g\rangle = |1\ldots1\ldots1\rangle$ is the state with all atoms in their ground states, $|j\rangle = |1\ldots2_j1\ldots1\rangle$ is the state with atom j in state 2 and all the other atoms in their ground states, $|jj'\rangle = |1\ldots2_j1\ldots2_{j'}1\ldots1\rangle$ is the state with atoms j and j' in state 2 and all the other atoms in their ground states, and ω_0 is the transition frequency between states 2 and 1. We neglect all spatial phase factors since they will not contribute in the phase-matched direction. The initial condition is that the ensemble is in the ground state before the fields are applied. For the state vector given in Eq. (1),

$$P_0(t) = |c_0(t)|^2, \tag{2a}$$

$$P_1(t) = \left|\sum_{j=1}^{N} c_j(t)\right|^2, \tag{2b}$$

$$P_2(t) = 2\left|\sum_{\substack{j,j'=1 \\ j'>j}}^{N} c_{jj'}(t)\right|^2. \tag{2c}$$

The limit of at most two excitations can be achieved in two ways. If the driving field is sufficiently weak to ensure that

$$\sqrt{N}A \ll 1, \tag{3}$$

where A is the pulse area, then the average number of excitations in the ensemble is much less than unity. We refer to this as the perturbation theory limit. Alternatively, for a maximum effective Rabi frequency Ω of the field(s) and a characteristic Rydberg-Rydberg interaction shift Δ that satisfy

$$\frac{\sqrt{N}\Omega}{|\Delta|} \ll 1, \tag{4}$$

the blockade limit is approached, in which the number of double excitations is much less than unity. If neither of these inequalities hold, it is necessary to include terms beyond the doubly excited states.

There are essentially two ways to measure the effectiveness of the blockade. The first is a direct measurement of the number of Rydberg excitations [3]. In doing so, one can construct the second-order correlation function of the atomic ensemble at zero time delay, defined by

$$g^{(A)}(t)=\frac{\langle\hat{n}^2\rangle}{\langle\hat{n}\rangle^2}-\frac{1}{\langle\hat{n}\rangle}=\frac{\sum_{j,j'=1}^{N}\langle\sigma_+^{(j)}\sigma_+^{(j')}\sigma_-^{(j)}\sigma_-^{(j')}\rangle}{\left(\sum_{j=1}^{N}\langle\sigma_+^{(j)}\sigma_-^{(j)}\rangle\right)^2}, \quad (5)$$

where

$$\hat{n}=\sum_{j=1}^{N}\sigma_+^{(j)}\sigma_-^{(j)} \quad (6)$$

and $\sigma_\pm^{(j)}$ are raising and lowering operators for atom j associated with the Rydberg transition. For the state vector given in Eq. (1) [4],

$$g^{(A)}(t)=\frac{2P_2(t)}{[P_1(t)+2P_2(t)]^2}\approx\frac{2P_2(t)}{[P_1(t)]^2}. \quad (7)$$

An alternative method to probe the blockade is to apply a readout pulse that leads to phase-matched emission from the sample [4]. In this case, the second-order correlation function at zero time delay, $g^{(2)}(t)$, of the phase-matched emission can serve as a measure of the effectiveness of the blockade. Clearly, $g^{(2)}(t)=g^{(A)}(t)=0$ if $P_2=0$. You might think that $g^{(2)}(t)\approx g^{(A)}(t)\to 0$ if $P_2\ll P_1\approx 1$, but this is not necessarily true. In fact, the purpose of this paper is to demonstrate that interaction-induced dephasing can lead to significant differences between $g^{(2)}(t)$ and $g^{(A)}(t)$ even if $P_2\ll P_1\approx 1$. In effect, we want to calculate $g^{(2)}(T)$ and $g^{(A)}(T)$, where T is the duration of the excitation pulse. We shall assume that the readout pulse is applied shortly after the excitation pulse(s) and is sufficiently short to avoid any dephasing following the excitation.

In general, the correlation function $g^{(2)}(T)$ can also be used as a figure of merit in applications involving single-photon sources. Rydberg-based single-photon sources can be divided into two distinct protocols. The first involves the excitation of symmetric collective states in the regime of the Rydberg blockade, followed by a linear mapping of the collective atomic state into a light pulse, resulting in $g^{(2)}(T)=g^{(A)}(T)=0$ with unit efficiency [1,4]. The second involves an excitation pulse whose duration T is sufficiently short to result in $g^{(A)}(T)\approx g^{(2)}(T)\approx 1$. The excitation is followed by a storage period T_s during which time interaction-induced dephasing results in $g^{(2)}(T+T_s)\to 0$ for sufficiently long T_s, with a maximum efficiency of $1/e$ [5,6]. The value of $g^{(A)}(T)$ is unchanged during the storage period, $g^{(A)}(T+T_s)=g^{(A)}(T)$. Some of the first realizations of both cold-atom- and hot-atom-based Rydberg sources have been carried out using the excitation-induced dephasing [7]. The Rydberg blockade limit can be approached if larger principal quantum numbers of the Rydberg levels are used or if the size of the ensemble is reduced [8]. Recently, optimized single-photon sources were used to demonstrate high-purity single-photon wave packets and high-fidelity photonic quantum logic [9,10].

The difference between $g^{(A)}$ and $g^{(2)}$ can be important when considering schemes that are designed to produce single-photon sources immediately following the excitation pulse. The true measure of a single-photon source is to have $g^{(2)}(T)\to 0$, which is achieved for a perfect blockade. However, it is rarely possible to produce a perfect Rydberg blockade or to limit the number of excitations in the sample to a single excitation. That is, to obtain observable signals, one is often required to work in regimes when there is a nonvanishing probability to have two excitations in the sample. Once there is even a small probability to have two excitations in the sample, the values of $g^{(2)}(T)$ and $g^{(A)}(T)$ can differ significantly.

You may ask why the values of $g^{(2)}(t)$ and $g^{(A)}(t)$ differ. The difference can be traced to the state vector given in Eq. (1). If all the $c_{jj'}(t)$ were equal, the state vector would be symmetric on the interchange of particles and one would find that $g^{(2)}(t)=g^{(A)}(t)$. However, in the presence of Rydberg-Rydberg interactions the $c_{jj'}(t)$ are *not* equal, depending on the positions of the atoms. As such, when averaging over the interatomic separations, the resulting interaction-induced dephasing is different for $g^{(2)}(t)$ and $g^{(A)}(t)$. Although there are many papers devoted to the state dynamics of the symmetric states in the Rydberg blockade, there are far fewer that explore the role of nonsymmetric states [11]. We will see that, in the presence of interaction-induced dephasing, $g^{(2)}(t)=|\langle f(t)\rangle|^2$, while $g^{(A)}(t)=\langle|f(t)|^2\rangle$, where $f(t)$ is some function that depends on interatomic separations and the average is over interatomic separations. Clearly, $g^{(A)}(t)\leqslant g^{(2)}(t)$.

In this paper we analyze the atom field dynamics in the limit that the driving field creates at most two excitations in the ensemble. We consider the combined action of the dipole blockade and interaction-induced dephasing during the time period in which the excitation field acts. We shall see that in the regime that we study there can be significant differences between $g^{(A)}$ and $g^{(2)}$.

II. CALCULATION OF $g^{(2)}$ AND $g^{(A)}$

The second-order correlation of the phase-matched emission is given by

$$g^{(2)}(t)=B(t)/I(t)^2, \quad (8)$$

where

$$I(t)=\langle S_+S_-\rangle \quad (9)$$

is proportional to the field intensity and

$$B(t)=\langle S_+S_+S_-S_-\rangle. \quad (10)$$

The ladder operators $S_\pm$ are defined as

$$S_\pm=\frac{1}{\sqrt{N}}\sum_{j=1}^{N}\sigma_\pm^{(j)}. \quad (11)$$

In writing these expressions, we have assumed that the raising and lowering operators for the transition on which the signal is emitted are proportional to those of the Rydberg transition.

We first imagine that the state vector is fully symmetric upon interchange of particles, that is,

$$|\psi\rangle=p_0(t)|P_0\rangle+p_1(t)e^{-i\omega_0 t}|P_1\rangle+p_2(t)e^{-2i\omega_0 t}|P_2\rangle, \quad (12)$$

where

$$|P_0\rangle = |g\rangle, \tag{13a}$$

$$|P_1\rangle = \frac{1}{\sqrt{N}}\sum_{j=1}^{N}|j\rangle, \tag{13b}$$

$$|P_2\rangle = \sqrt{\frac{2}{N(N-1)}}\sum_{\substack{j,j'=1\\ j'>j}}^{N}|jj'\rangle. \tag{13c}$$

From Eqs. (1), (12), and (13), it is clear that for the fully symmetric state

$$c_0(t) = p_0(t), \tag{14a}$$

$$c_j(t) = p_1(t)/\sqrt{N}, \tag{14b}$$

$$c_{jj'}(t) = p_2(t)\sqrt{\frac{2}{N(N-1)}}. \tag{14c}$$

Using

$$S_-|P_q\rangle = \sqrt{q}\sqrt{\frac{N-q+1}{N}}|P_{q-1}\rangle, \tag{15a}$$

$$S_+|P_q\rangle = \sqrt{q+1}\sqrt{\frac{N-q}{N}}|P_{q+1}\rangle, \tag{15b}$$

we can calculate

$$I(t) = P_1(t) + 2\frac{N-1}{N}P_2(t), \tag{16a}$$

$$B(t) = 2\frac{N-1}{N}P_2(t), \tag{16b}$$

$$g^{(2)}(t) = \frac{2\frac{N-1}{N}P_2(t)}{\left[P_1(t)+2\frac{N-1}{N}P_2(t)\right]^2}, \tag{16c}$$

$$g^{(A)}(t) = \frac{2P_2(t)}{[P_1(t)]^2}, \tag{16d}$$

where $P_n = |p_n|^2$. Note that if $N \gg 1$ and $P_2 \ll P_1$, then

$$g^{(2)}(t) \approx \frac{2P_2(t)}{[P_1(t)]^2} = g^{(A)}(t). \tag{17}$$

Thus, in the limit that $N \gg 1$ and $P_2 \ll P_1$, any difference between $g^{(2)}$ and $g^{(A)}$ arises from the the fact that, owing to atom-atom interactions, the state vector cannot be expressed solely in terms of fully symmetric states.

We next consider the state vector given in Eq. (1), which is not restricted to fully symmetric states. It follows from Eqs. (1), (8), (10), (11), and (5) that

$$I(t) = \frac{1}{N}\left|\sum_{j=1}^{N}c_j(t)\right|^2 + \frac{1}{N}\sum_{j=1}^{N}\left|\sum_{j'\neq j=1}^{N}\tilde{c}_{jj'}(t)\right|^2, \tag{18a}$$

$$B(t) = \left|\frac{2}{N^2}\sum_{\substack{j,j'=1\\ j'>j}}^{N}c_{jj'}(t)\right|^2, \tag{18b}$$

$$g^{(2)}(t) = \frac{\left|\frac{2}{N^2}\sum_{\substack{j,j'=1\\ j'>j}}^{N}c_{jj'}(t)\right|^2}{\left(\frac{1}{N}\left|\sum_{j=1}^{N}c_j(t)\right|^2 + \frac{1}{N}\sum_{j=1}^{N}\left|\sum_{j'\neq j=1}^{N}\tilde{c}_{jj'}(t)\right|^2\right)^2}, \tag{18c}$$

$$g^{(A)}(t) = \frac{2\sum_{\substack{j,j'=1\\ j'>j}}^{N}\left|\tilde{c}_{jj'}(t)\right|^2}{\left(\sum_{j=1}^{N}\left|c_j(t)\right|^2\right)^2}, \tag{18d}$$

where $\tilde{c}_{jj'} = c_{j'j}$ if $j' < j$.

The problem then reduces to finding values of c_j and $c_{jj'}$ for a given excitation scheme and atom-atom interaction. We take as our Hamiltonian in an interaction representation

$$H = \hbar\chi(t)\sum_{j=1}^{N}(\sigma_+^{(j)} + \sigma_-^{(j)}) + \sum_{\substack{j,j'=1\\ j'>j}}^{N}\hbar\Delta_{jj'}\sigma_2^{(j)}\sigma_2^{(j')}, \tag{19}$$

where $\chi(t)$ (assumed real) is one half of the effective Rabi frequency that drives the Rydberg transition, $\Delta_{jj'}$ is a Rydberg-Rydberg interaction shift, and $\sigma_2^{(j)}$ is the excited-state population operator of atom j. The incident field is assumed to resonantly drive the transition. The second term in Eq. (19) contributes to both the dipole blockade and the interaction-induced dephasing and leads to differences between $g^{(A)}$ and $g^{(2)}$.

With this Hamiltonian, the equations of motion for the state amplitudes are

$$\dot{c}_0 = -i\chi(t)\sum_{j=1}^{N}c_j, \tag{20a}$$

$$\dot{c}_j = -i\chi(t)c_0 - i\chi(t)\sum_{j'\neq j=1}^{N}\tilde{c}_{jj'}, \tag{20b}$$

$$\dot{c}_{jj'} = -i\chi(t)(c_j + c_{j'}) - i\Delta_{jj'}c_{jj'}. \tag{20c}$$

In general these equations are difficult to solve; however, when the inequality in Eq. (3) (perturbation theory) or in Eq. (4) is satisfied (dipole blockade), an analytic solution can be obtained. In carrying out the calculations we assume that $N \gg 1$, so that we can replace

$$\sum_{\substack{j,j'=1\\ j'>j}}^{N} \approx \frac{1}{2}\sum_{\substack{j,j'=1\\ j'\neq j}}^{N} \equiv \frac{1}{2}\sum_{j,j'}. \tag{21}$$

In what follows, we suppress the condition $j' \neq j$ in any sums over j and j'; it is implicit.

A. Perturbation theory

In perturbation theory, to ensure that $c_0 \approx 1$, it is necessary that

$$\sqrt{N}A(t) \ll 1, \tag{22}$$

where

$$A(t) = 2\int_{-\infty}^{t}\chi(t')dt'. \tag{23}$$

In this limit,

$$c_j(t) \approx -i\int_{-\infty}^{t}\chi(t')dt',$$

$$c_{jj'}(t) \approx -2\int_{-\infty}^{t}\chi(t')e^{-i\Delta_{jj'}(t-t')}dt'\int_{-\infty}^{t'}\chi(t'')dt'',$$

and

$$g^{(2)}(t) = \frac{\left|\frac{2}{N^2}\sum_{j,j'}\int_{-\infty}^{t}\chi(t')e^{-i\Delta_{jj'}(t-t')}dt'\int_{-\infty}^{t'}\chi(t'')dt''\right|^2}{\left|\int_{-\infty}^{t}\chi(t')dt'\right|^4}, \tag{24}$$

$$g^{(A)}(t) = \frac{\sum_{j,j'}\left|2\int_{-\infty}^{t}\chi(t')e^{-i\Delta_{jj'}(t-t')}dt'\int_{-\infty}^{t'}\chi(t'')dt''\right|^2}{N^2\left|\int_{-\infty}^{t}\chi(t')dt'\right|^4}. \tag{25}$$

If $\Delta_{jj'}=0$, then $g^{(2)}=g^{(A)}=1$, the result for a product state. The time t in these expressions refers to a time just following the excitation pulse(s).

We take

$$\Delta_{jj'} = \frac{C}{\left|\mathbf{r}_j-\mathbf{r}_{j'}\right|^6}. \tag{26}$$

Going over to continuum variables and setting $\mathbf{r}=\mathbf{r}_j-\mathbf{r}_{j'}$, it is possible to write $g^{(2)}(t)$ as

$$g^{(2)}(t) = \frac{\left|2\int drW(r)\int_{-\infty}^{t}\chi(t')e^{-iC_6(t-t')/r^6}dt'\int_{-\infty}^{t'}\chi(t'')dt''\right|^2}{\left|\int_{-\infty}^{t}\chi(t')dt'\right|^4} \tag{27}$$

and $g^{(A)}(t)$ as

$$g^{(A)}(t) = \frac{\int drW(r)\left|2\int_{-\infty}^{t}\chi(t')e^{-iC_6(t-t')/r^6}dt'\int_{-\infty}^{t'}\chi(t'')dt''\right|^2}{\left|\int_{-\infty}^{t}\chi(t')dt'\right|^4}, \tag{28}$$

where $W(r)$ is the nearest-neighbor distribution. For $W(r)$, we take a uniform spherical distribution of atoms for which [12]

$$W(r) = 3\frac{r^2}{R^3}-\frac{9}{4}\frac{r^3}{R^4}+\frac{3}{16}\frac{r^5}{R^6}, \qquad 0\leqslant r\leqslant 2R. \tag{29}$$

We consider two pulse shapes, a constant amplitude pulse and a Gaussian pulse amplitude,

$$\chi(t) = \chi_1\Theta(t)\Theta(T-t), \tag{30}$$

$$\chi(t) = \chi_2 e^{-t^2/T_p^2}, \tag{31}$$

where $\Theta(t)$ is a Heaviside function.

B. Dipole blockade limit

The second case for which an analytic approximation is valid is when

$$\sqrt{N}\left|\frac{\chi(t)}{\Delta_{jj'}}\right| \ll 1, \tag{32}$$

allowing us to treat $c_{jj'}$ in perturbation theory. In this limit, it follows from Eqs. (20) that

$$c_0(t) = \cos[\sqrt{N}A(t)/2], \tag{33a}$$

$$c_j(t) = -i\frac{\sin[\sqrt{N}A(t)/2]}{\sqrt{N}}, \tag{33b}$$

$$c_{jj'}(t) \approx -\frac{2\chi(t)}{\Delta_{jj'}}c_j = 2i\chi(t)\frac{\sin[\sqrt{N}A(t)/2]}{\sqrt{N}\Delta_{jj'}}, \tag{33c}$$

such that

$$g^{(2)}(t) \approx \frac{\left|\sum_{j,j'}\tilde{c}_{jj'}(t)\right|^2}{\left|\sum_j c_j(t)\right|^4} = \frac{4\chi(t)^2\left|\sum_{j,j'}\frac{1}{\Delta_{jj'}}\right|^2}{N^3\sin^2\left[\sqrt{N}A(t)/2\right]}$$

$$= \frac{4\chi(t)^2\left|\int drW(r)r^6/C_6\right|^2}{\sin^2\left[\sqrt{N}A(t)/2\right]} \tag{34}$$

and

$$g^{(A)}(t) = \frac{\sum_{j,j'}\left|\tilde{c}_{jj'}(t)\right|^2}{\left[\sum_j\left|c_j(t)\right|^2\right]^2} = \frac{4\chi(t)^2\sum_{j,j'}\left|\frac{1}{\Delta_{jj'}}\right|^2}{\sin^2\left[\sqrt{N}A(t)/2\right]}$$

$$= \frac{4\chi(t)^2\int drW(r)r^{12}/C_6^2}{\sin^2[\sqrt{N}A(t)/2]}. \tag{35}$$

Recall that $\sum_{j,j'}$ excludes $j=j'$. For the limits considered in this paper, it follows from Eqs. (27), (28), (34), and (35) that $g^{(2)}(t)\leqslant g^{(A)}(t)$.

III. CONSTANT AMPLITUDE PULSES

A. Perturbation theory limit

If $\chi(t)$ is given by Eq. (30),

$$g_1^{(2)}(T) = \frac{\left|2\chi_1^2\int drW(r)\int_0^T e^{-iC_6(t-t')/r^6}dt'\int_0^{t'}dt''\right|^2}{\chi_1^4\left|\int_0^t dt'\right|^4}$$

$$= \left|2\int drW(r)\frac{1-e^{-iC_6T/r^6}+iC_6T/r^6}{(C_6T/r^6)^2}\right|^2 \tag{36}$$

or

$$g_1^{(2)}(\tau_1) = \left|2\int dsP(s)\frac{1-e^{-i\tau_1/s^6}+i\tau_1/s^6}{\tau_1^2/s^{12}}\right|^2, \tag{37}$$

where

$$P(s) = 3s^2-\frac{9}{4}s^3+\frac{3}{16}s^5, \qquad 0\leqslant s\leqslant 2, \tag{38a}$$

$$\tau_1 = C_6T/R^6. \tag{38b}$$

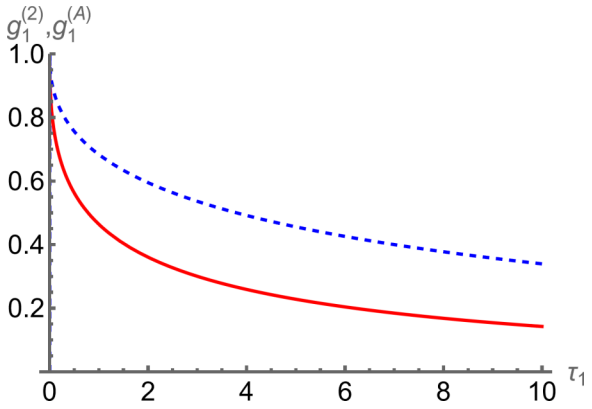

FIG. 1. Graphs of $g_1^{(2)}$ (solid red curve) and $g_1^{(A)}$ (dashed blue curve) as a function of τ_1.

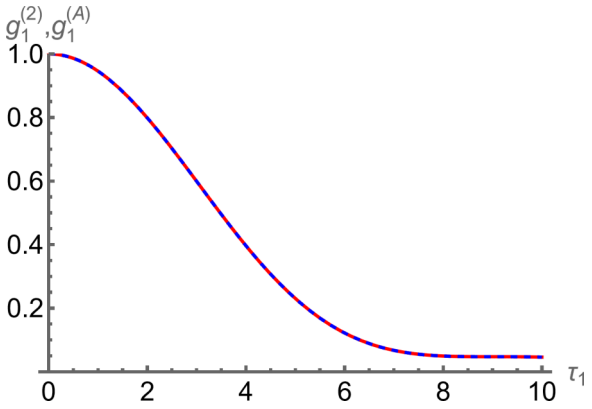

FIG. 2. Graphs of $g_1^{(2)}$ (solid red curve) and $g_1^{(A)}$ (dashed blue curve) as a function of τ_1 for equal level shifts.

It turns out you can get analytic (long) expressions for the integrals, which are not reproduced here. The quantity τ_1 is a characteristic phase shift for the uniform density distribution.

The results can be compared with those for $g^{(A)}$,

$$g_1^{(A)}(T) = \int dr W(r) \left| 2\frac{1 - e^{-iC_6T/r^6} + iC_6T/r^6}{(C_6T/r^6)^2} \right|^2,$$
$$g_1^{(A)}(\tau_1) = \int ds P(s) \left| 2\frac{1 - e^{-i\tau_1/s^6} + i\tau_1/s^6}{\tau_1^2/s^{12}} \right|^2, \tag{39}$$

for which analytic expressions are also available. Results are shown in Fig. 1 with the red solid curve giving $g_1^{(2)}(\tau_1)$ and the blue dashed curve giving $g_1^{(A)}(\tau_1)$. The characteristic decay time of $\tau_1 \approx 4$ depends to a large extent on the definition of τ_1 given in Eq. (38b), in which the phase corresponds to atoms separated by R. If $\tau_1 = 1$, most of the atoms separated by distances greater than R would have phase shifts much less than unity since the phase shift falls off as separation to the minus 6 power. It would take a value of $\tau_1 = 64$ to ensure that the phase for *all atomic* separations is greater than unity. For $\tau_1 \gg 1$,

$$g_1^{(2)} \sim \frac{16\,384}{225\tau_1^2}, \tag{40a}$$
$$g_1^{(A)} \sim \frac{4096}{15\tau_1^2}, \tag{40b}$$
$$\frac{g_1^{(2)}}{g_1^{(A)}} \sim \frac{4}{15}. \tag{40c}$$

It is interesting to plot the results for equal $\Delta_{jj'}$, even if this limit has no physical basis. We set $\Delta_{jj'} = C_6/R^6$ and replace C_6/r^6 by C_6/R^6 in Eqs. (37) and (39) to obtain

$$g_1^{(2)}(\tau_1) = g_1^{(A)}(\tau_1) = \left| 2\frac{1 - e^{-i\tau_1} + i\tau_1}{\tau_1^2} \right|^2. \tag{41}$$

As shown in Fig. 2, the two curves now overlap and have a different shape. The dipole blockade still results in values of $g_1^{(2)}(\tau_1)$ and $g_1^{(A)}(\tau_1)$ much less than unity for sufficiently large τ_1. However, it is the interaction-induced dephasing that originates from a *distribution* of $\Delta_{jj'}$ that results in $g_1^{(2)}(\tau_1) \neq g_1^{(A)}(\tau_1)$ and the more rapid decrease in $g_1^{(2)}(\tau_1)$ than in $g_1^{(A)}(\tau_1)$ for early times seen in Fig. 1.

B. Dipole blockade limit

In the second approximation,

$$g_1^{(2)}(T) \approx \frac{4\chi_1^2 \left| \sum_{j,j'} \frac{1}{\Delta_{jj'}} \right|^2}{N^3 \sin^2(\sqrt{N}\chi_1 T)} = \frac{4N\beta_1^2 \left| \int ds P(s) s^6 \right|^2}{\sin^2(\sqrt{N}\chi_1 T)} \tag{42}$$

and

$$g_1^{(A)}(T) = \frac{4\chi_1^2 \sum_{j,j'} \left| \frac{1}{\Delta_{jj'}} \right|^2}{N \sin^2(\sqrt{N}\chi_1 T)} = \frac{4N\beta_1^2 \int ds P(s) s^{12}}{\sin^2(\sqrt{N}\chi_1 T)}, \tag{43}$$

where

$$\beta_1 = \frac{\chi_1}{C_6/R^6}, \tag{44}$$

and we have gone over into continuum variables. In this case, aside from the $\sin^2(\sqrt{N}\chi T)$ factor, both $g^{(2)}$ and $g^{(A)}$ are time independent, since we have assumed that $|\Delta_{jj'}|T \gg 1$. The integrals can be carried out to obtain

$$g_1^{(2)}(T) = \frac{16\,384N\beta_1^2}{225\sin^2(\sqrt{N}\chi_1 T)} \approx 72.8\frac{N\beta_1^2}{\sin^2(\sqrt{N}\chi_1 T)}, \tag{45a}$$
$$g_1^{(A)}(T) = \frac{4096N\beta_1^2}{15\sin^2(\sqrt{N}\chi_1 T)} \approx 273\frac{N\beta_1^2}{\sin^2(\sqrt{N}\chi_1 T)}. \tag{45b}$$

Note that in the perturbation theory limit given by Eq. (3), these reduce to

$$g_1^{(2)} \sim \frac{16\,384}{225\tau_1^2}, \tag{46a}$$
$$g_1^{(A)} \sim \frac{4096}{15\tau_1^2}, \tag{46b}$$
$$\frac{g_1^{(2)}}{g_1^{(A)}} \sim \frac{4}{15}. \tag{46c}$$

Equations (46) are valid only for times τ_1 for which $g_1^{(2)} \ll 1$ and $g_1^{(A)} \ll 1$. These results are identical to the perturbation

theory results of Eqs. (40) in the limit that $\tau_1 \gtrsim 250$. Such large values of τ_1 guarantee that only singly excited states contribute in the perturbation theory limit.

IV. GAUSSIAN PULSES

We next consider the Gaussian pulses given by Eq. (31). The excitation pulse is assumed to be adiabatic; that is,

$$\tau_2 \gg 1, \tag{47}$$

where τ_2 is defined as

$$\tau_2 = C_6 T_p / R^6. \tag{48}$$

We can dispense of the dipole blockade limit immediately. If inequality (4) holds, the $c_{jj'}$ adiabatically follow the field, implying that

$$g_2^{(2)} = g_2^{(A)} \approx 0 \tag{49}$$

following the pulse.

We are left with the perturbation theory limit. To illustrate the relevant physics, we compare only $g_2^{(A)}$ with $g_1^{(A)}$ (but not $g_2^{(2)}$ with $g_1^{(2)}$) since dephasing reduces $g_2^{(2)}$ (but not $g_2^{(A)}$) for times $t \gg T_p$, when the pulse amplitude is very small. As a consequence, we can integrate out to $t = \infty$ in calculating $g_2^{(A)}$ without introducing any significant dephasing for times $t \gg T_p$. We set

$$T_p = T/\sqrt{\pi}, \tag{50}$$

which leads to the same probability of excitation for a single atom for the constant and Gaussian pulses. For the Gaussian pulse,

$$\begin{aligned} g_2^{(A)}(\tau_2) &= \frac{\int ds P(s) \left| 2\int_{-\infty}^{\infty} e^{-\tau'^2} e^{i\tau_2\tau'/s^6} d\tau' \int_{-\infty}^{\tau'} e^{-\tau''^2} d\tau'' \right|^2}{\left| \int_{-\infty}^{\infty} e^{-\tau'^2} d\tau' \right|^4} \\ &= \frac{\int ds P(s) \left| 2\int_{-\infty}^{\infty} e^{-\tau'^2} e^{i\tau_2\tau'/s^6} d\tau' [1 + \Phi(\tau')] \right|^2}{4\pi} \\ &= \int ds P(s) \left| e^{-(\tau_2/s^6)^2/4} \left[1 - \Phi\left(\frac{i\tau_2/s^6}{2\sqrt{2}} \right) \right] \right|^2, \end{aligned} \tag{51}$$

where $\Phi(\tau)$ is an error function. A comparison of $g_2^{(A)}(\tau_2)$ with $g_1^{(A)}(\sqrt{\pi}\tau_2)$ is given in Fig. 3. The red solid curve is for the Gaussian field envelope and the blue dashed curve is for a finite duration, constant field amplitude. As you can see, there does not appear to be much difference, whereas we might have expected a much smaller value with adiabatic excitation.

How can we understand these results? We have already seen that, in the perturbation theory limit, the contributions from the doubly excited state is fully damped only for large values of τ_1. That is, we should expect adiabaticity to play a significant role only when most of the atoms experience level shifts that lead to state amplitude phases that are greater than unity. This occurs only for $\tau_1 \gg 1$. Thus, it is only in this limit that we would expect the adiabaticity to lead to values of $g_2^{(A)}(\tau_2 = \tau_1/\sqrt{\pi})$ to approach 0, a result that is illustrated in Fig. 4.

V. DISCUSSION

We have shown that interaction-induced dephasing during the excitation process always results in $g^{(2)} \leqslant g^{(A)}$. For example, for a uniform density of atoms in a spherical volume, the ratio $g^{(2)}/g^{(A)}$ approaches 4/15 at long times. These observations imply that in typical realizations of Rydberg single-photon sources, the values of $g^{(A)}$ were likely much

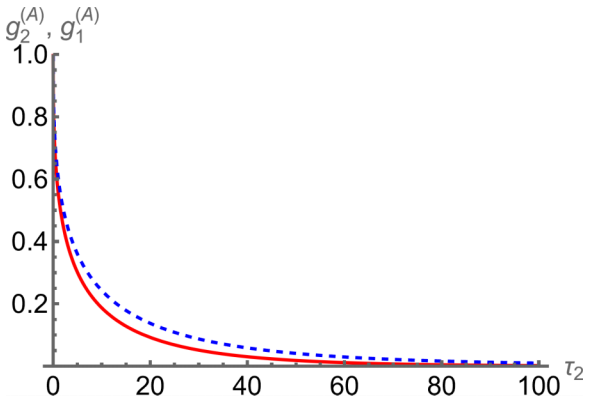

FIG. 3. Graphs of $g_2^{(A)}(\tau_2)$ (solid red curve) and $g_1^{(A)}$ $(\sqrt{\pi}\tau_2)$ (dashed blue curve) as a function of τ_2.

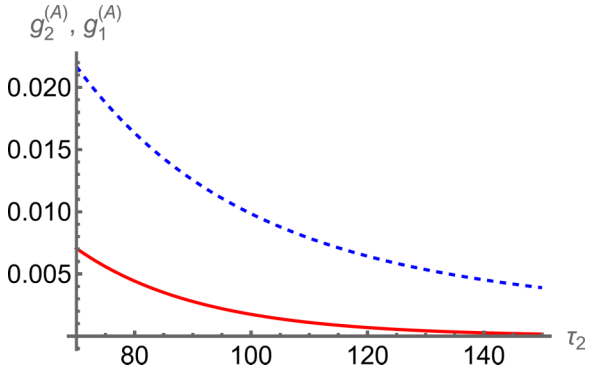

FIG. 4. Graphs of $g_2^{(A)}(\tau_2)$ (solid red curve) and $g_1^{(A)}$ $(\sqrt{\pi}\tau_2)$ (dashed blue curve) as a function of τ_2.

larger than the observed values of $g^{(2)}$ due to suppression of the latter by the interaction-induced dephasing. We have also shown that it is desirable to use pulsed adiabatic excitation to produce a Rydberg-based single-photon source having the lowest value of $g^{(2)}(0)$. Our discussion has been limited to a uniform spherical density, but the results will be qualitatively similar for other density distributions, although the ratio $g^{(2)}/g^{(A)}$ at long times can differ. For example, a Gaussian atomic density results in this ratio being $g^{(2)}/g^{(A)} \to 0.102$.

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