Coincidence counts and stimulated emission resulting from weak pulsed-field-atom interactions

P. R. Berman[®] and A. Kuzmich

Physics Department, University of Michigan, Ann Arbor, Michigan 48109-1040, USA

(Received 18 September 2019; revised manuscript received 21 February 2020; accepted 21 February 2020; published 16 March 2020)

The interaction of a single-photon pulse of radiation with an atom or an ensemble of atoms is studied using a source-field approach. The atom-field interaction is weak insofar as it can be treated in lowest, nonvanishing order of perturbation theory. The output field intensity and second-order correlation function of the field are calculated. It is shown that, even when any modification of the atomic dynamics produced by the incident field is neglected, as a function of the time delay τ between the incident field pulse and the field radiated by the atom(s), there is a "bump" in the time-integrated second-order correlation at $\tau = 0$. The increase in coincidence counts for $\tau = 0$ can be interpreted in terms of Hanbury Brown and Twiss-type interference. By looking at the field intensity, we show that the bump has no direct relation with stimulated emission—it occurs even when the input field is attenuated. In recent down conversion experiments, an increase in coincidence counts has been attributed to stimulated emission—we comment on the validity of such an interpretation in light of our results.

DOI: 10.1103/PhysRevA.101.033824

I. INTRODUCTION

Any study of atom-field interactions would not be complete without the inclusion of spontaneous and stimulated emission. In his 1917 paper [1], Einstein introduced his famous A and B coefficients, with the A coefficient associated with spontaneous emission and the B coefficient with either absorption or stimulated emission (both referred to as "changes of state due to irradiation" by Einstein). Although the underlying origin of spontaneous and stimulated emission is well understood, there does not seem to be a universal definition as to what constitutes stimulated emission. Most definitions relate to processes in which atom-field interactions lead to an increase in the field intensity of an input field, although some are somewhat more restrictive insofar as they require the increase in the field intensity to occur in the same spatiotemporal mode of the input field [2]. Moreover, when the incident field drives atomic transitions having frequency ω_0 , stimulated emission is often referred to as a process in which each excited atom imparts an extra energy of $\hbar\omega_0$ to the field. Clearly, both of these criteria apply to stimulated emission in the Jaynes-Cummings model [3], since there is only one spatial field mode. They also apply (approximately) when a π pulse is incident on a two-level atom that is prepared in its excited state, provided the pulse duration is much less than the lifetime of the excited state. In that case, the average energy in the field is increased by $\hbar\omega_0$ and the spatiotemporal form of the output field mode, while not exactly the same as that of the input field, can approximate it to a high degree. In general, however, the stimulated emission that occurs when an arbitrary pulse is incident on an atom or atoms prepared with a population inversion does not produce an output field mode that is in the same spatiotemporal mode as the input. We might point put that, even in chiral wave guides [4], when an *n*-photon pulse incident on an atom in its excited state results in an (n + 1)-photon output pulse, the output field mode is

never in *exactly* the same spatiotemporal mode as the input field.

Atom-field interactions result not only in a change in the intensity of the incident field, they also lead to an output field whose second-order correlation function differs from that of the incident field. The most dramatic situation occurs when a single-photon probe pulse is incident on a two-level atom prepared in an arbitrary initial state at t = 0. Clearly, the second-order correlation function vanishes for the incident field since it corresponds to a single-photon state. However, for times t > 0, the output field state has a two-photon component. As a result, the second-order correlation function of the output field does not vanish, in general. In a recent experiment [5] involving phase-matched emission from an ensemble of three-level atoms, the second-order time-integrated correlation function of the output field was measured as a function of the delay τ between the input probe pulse and the atomic emission. An increase in the second-order time-integrated correlation function that occurred for $\tau = 0$ was attributed to constructive Hanbury Brown and Twiss (HBT) [6] or Hong-Ou-Mandel (HOM) [7] interference. Both HBT and HOM fall into the general category of two-photon interference, a subject that has been discussed extensively by many authors [8]. We are concerned here with HBT interference associated with pulsed fields from independent sources-in such cases, HBT interference is a type of intensity-intensity interference that can occur only for overlapping pulses.

Although stimulated emission involves a change in the field *intensity* and does not directly relate to the second-order correlation function, there can be correlations between the two processes. In fact, several authors [9] explain an increase in coincidence counts observed in down-conversion experiments (equivalent to the nonvanishing of the second-order correlation function) in terms of stimulated emission processes. You might then ask, "Is the increase in coincidence counts a *consequence* of stimulated emission?" To help clarify some of

these issues, we consider a number of simple physical systems in which a single photon probe pulse is incident on an atom (or ensemble of atoms) that can itself radiate a pulsed field, even in the absence of the probe field. In each case we calculate the spatiotemporal intensity of the output field to see if it matches that of the incident pulse. Moreover, we calculate the time-integrated second-order correlation function for the total output field. We see that there can be an increase in the timeintegrated second-order correlation function when the input pulse overlaps with the field radiated from the atom(s), even though stimulated emission is either absent or negligible in the cases to be considered. This increase is interpreted as arising from HBT-like interference.

Specifically, we look at three scenarios. In the first, a single-photon pulse is incident on a two-level atom prepared in an arbitrary initial state. As a function of the time delay τ between the incident probe field and the atomic emission, we show that there can be a twofold increase in the timeintegrated second-order correlation function, even in situations where the probe field intensity is *reduced* as a result of the atom-field interaction. In the second scenario, we consider a single-photon pulse incident on a three-level atom that is driven by a classical pump field in a Raman configuration. This level scheme offers several advantages. Owing to offresonant driving by the pump field, Raman emission occurs only for times when the pump field interacts with the atom. The probe field always experiences stimulated emission in this scenario. To make some connection with the type of calculations used to explain down conversion experiments, we use both a source-field approach [10] and a state-vector approach to obtain the output field intensity. We find that subtle problems arise in the state vector approach. In particular, when the state vector is evaluated in lowest nonvanishing order of perturbation theory (as is typically done in analyses of down conversion experiments [9]), a spurious term appears. It is necessary to include higher-order corrections to the state vector to recover the correct result. Moreover, the spatiotemporal dependence of the amplified output field is not the same as the input field. Finally, we analyze a scenario in which classical fields and a single-photon pulse are incident on an atomic ensemble, resulting in phase-matched emission. In this case, we show that any stimulated emission depends linearly on the atomic density, whereas the second-order correlation function varies as the square of the atomic density. In all cases, we adopt a one-dimensional model for the incident pulsed field and assume that a *weak-coupling* approximation is valid—atom-field interactions are treated in lowest-order perturbation theory.

II. SINGLE-PHOTON PULSE INCIDENT ON A TWO-LEVEL ATOM

Consider a single atom fixed at the origin having a J = 0ground state and a J = 1 excited state. The ground state eigenket is denoted by $|1\rangle$ and the m = 0 sublevel of the excited state eigenket by $|2\rangle$, with the frequency separation of the levels equal to ω_0 . A single-photon pulse having central frequency $\bar{\omega} = \bar{k}c \approx \omega_0$ is incident on the atoms. The state vector associated with the atom-pulse system at time t = 0can be written as

$$|\psi(0)\rangle = \sum_{k} (\beta_1 |1\rangle + \beta_2 |2\rangle) b_k |1_k\rangle, \tag{1}$$

where $|1_k\rangle$ is the eigenket associated with a single photon in mode k, β_1 and β_2 are initial atomic state amplitudes, and b_k is the initial field state amplitude for mode k. We have made a paraxial approximation and neglected diffraction. That is, we assume that the initial pulse has cross-sectional area A, polarization \mathbf{u}_z and propagates in the \mathbf{u}_x direction (the **u**'s are unit vectors). A lossless, 50:50 beam splitter having crosssectional area A is located on the X axis a distance X_B from the atom and splits the output field into fields propagating in the X and Y directions. Detectors are placed on the X and Y axes an equal distance $D \ll X_B$ from the beam splitter. Coincidences are recorded of a photo count at one of the detectors at time t_1 and the other at time t_2 . Our system is intended to model the collection mode of a single-mode fiber.

With this geometry, the rate of coincidence counts, normalized to the field intensity at each of the detectors, is given approximately by the second-order correlation function, defined by

$$g^{(2)}(X_B, t_1, t_2) = \frac{\langle E_{-}(X_B, t_1)E_{-}(X_B, t_2)E_{+}(X_B, t_2)E_{+}(X_B, t_1)\rangle}{\langle E_{-}(X_B, t_1)E_{+}(X_B, t_1)\rangle\langle E_{-}(X_B, t_2)E_{+}(X_B, t_2)\rangle},$$
(2)

where $E_+(X, t) = [E_-(X, t)]^{\dagger}$ is the positive frequency component of the field operator at position X at time t. It has been assumed implicitly that the field amplitudes can be taken as constant over the area of the beam splitter allowing us to evaluate all fields on the X axis. We are also interested in the time-integrated correlation function, defined as

$$g^{(2)}(X_B) = \frac{\int dt_1 \int dt_2 \langle E_-(X_B, t_1) E_-(X_B, t_2) E_+(X_B, t_2) E_+(X_B, t_1) \rangle}{\left[\int dt \langle E_-(X_B, t) E_+(X_B, t) \rangle \right]^2}.$$
(3)

In effect, this expression corresponds to the normalized, timeintegrated number of coincidence counts measured at the detectors.

To calculate $g^{(2)}$, we use source-field theory [10] and write the field operator as

$$E_{+}(X_{B},t) = E_{+}(t_{r}) = E_{+}^{(0)}(t_{r}) + E_{+}^{(\text{Source})}(t_{r}), \qquad (4)$$

where

$$E_{+}^{(0)}(t) = i \sum_{k} \left(\frac{\hbar\omega_{k}}{2\epsilon_{0}AL}\right)^{1/2} a_{k}e^{-i\omega_{k}t}$$
$$\approx i \left(\frac{\hbar}{\mu}\right)^{1/2} \sqrt{\frac{\gamma_{2}'c}{L\omega_{0}}} \sum_{k} \sqrt{\omega_{k}}a_{k}e^{-i\omega_{k}t}, \qquad (5)$$

$$E_{+}^{(\text{Source})}(t) = i \frac{\hbar \gamma_2'}{\mu} \sigma_{-}(t) e^{-i\omega_0 t}, \qquad (6)$$

where μ is the dipole matrix element between the ground and excited state (assumed real and positive), a_k is a lowering operator for the field mode k, $\omega_k = kc$, $\sigma_-(t)$ is an atomic lowering operator in an interaction representation,

$$t_r = t - X_B/c \tag{7}$$

is a retarded time, and AL is the quantization volume. The quantity γ'_2 is defined by

$$\gamma_{2}' = \frac{\omega_{0}\mu^{2}}{2\hbar\epsilon_{0}Ac} = \frac{3\pi}{2}\frac{c^{2}}{\omega_{0}^{2}A}\gamma_{2},$$
(8)

where γ_2 is the spontaneous decay rate on the 2–1 transition. The ratio γ_2'/γ_2 represents the fraction of spontaneous emission that goes into the mode volume of the detector. The weak coupling approximation essentially boils down to the assumption that $\gamma_2'/\gamma_2 \sim 1/k_0^2 A \ll 1$.

We assume that only those values of k in Eq. (5) that are sharply peaked about $k = \bar{k} \approx k_0$ contribute significantly when expectation values are taken. As a consequence, we can replace $\sqrt{\omega_k}$ by $\sqrt{\omega_0}$ in Eq. (5). The correlation functions are given by

$$g^{(2)}(t_1, t_2) = \frac{\langle E_-(t_1)E_-(t_2)E_+(t_2)E_+(t_1)\rangle}{\langle E_-(t_1)E_+(t_1)\rangle\langle E_-(t_2)E_+(t_2)\rangle} \tag{9}$$

and

$$g^{(2)} = \frac{\int dt_1 \int dt_2 \langle E_-(t_1)E_-(t_2)E_+(t_2)E_+(t_1) \rangle}{[\int dt \langle E_-(t)E_+(t) \rangle]^2}, \quad (10)$$

where all times appearing in these expressions are retarded times. From this point onwards, unless noted otherwise, all times t that appear in equations correspond to the retarded time $t - X_B/c$. That is, for a beam splitter at $X = X_B$, the second-order correlation function $g^{(2)}(X_B, t_1, t_2)$ depends only on the retarded times $t_{1,2} - X_B/c$. Similarly the field intensity $I(X_B, t)$ at the detector is a function only of the retarded time $t - X_B/c$ and will be written as I(t).

It is assumed that the atom-input field interaction is weak, that is, to lowest order, all atom-input field interactions are neglected. In other words, the atomic operators evolve as if the input pulse was absent-the atom interacts only with

In a similar manner, using Eqs. (4)–(6), we can calculate

the vacuum field and undergoes spontaneous decay into all directions, such that

$$\sigma_{-}(t) \approx \sigma_{-}^{(0)}(t) = \sigma_{-}(0)e^{-\gamma t}\Theta(t), \qquad (11)$$

where $\gamma = \gamma_2/2$ and Θ is a Heaviside function. It is then a simple matter to calculate the dimensionless intensity at the beam splitter as

$$I_N^{(0)}(t) = \frac{I^{(0)}(t)}{\hbar\omega_0\gamma_2} = \frac{2\epsilon_0cA}{\hbar\omega_0\gamma_2} \langle E_-(t)E_+(t)\rangle$$
$$= \frac{c}{\gamma_2 L} \left|\sum_k e^{-i(\omega_k - \bar{\omega})t} b_k\right|^2 + \frac{\gamma_2'}{\gamma_2} |\beta_2|^2 e^{-\gamma_2 t} \Theta(t), \quad (12)$$

where Eqs. (5), (6), and (8) were used. Although the atom undergoes spontaneous emission in all directions, only a fraction γ_2'/γ_2 of this emission goes into the mode volume of the detector.

We transform to continuous variables using the prescription

$$\sum_{k} \to \frac{L}{2\pi} \int_{-\infty}^{\infty} dk; \quad b_k \to \left(\frac{2\pi}{L}\right)^{1/2} b(k) \qquad (13)$$

to obtain

$$I_{N}^{(0)}(t) = \frac{1}{\gamma_{2}} |f(t)|^{2} + \frac{\gamma_{2}'}{\gamma_{2}} |\beta_{2}|^{2} e^{-\gamma_{2} t} \Theta(t), \qquad (14)$$

where

$$f(t) \approx \sqrt{\frac{c}{2\pi}} \int_{-\infty}^{\infty} dk \, b(k) e^{-i\left(k-\bar{k}\right)ct}$$
(15)

is normalized such that

$$\int_{-\infty}^{\infty} dt |f(t)|^2 = 1.$$
 (16)

The quantity $|f(t)|^2$ is the temporal profile of the average field intensity measured at the detectors at time $t + X_B/c$ and $|b(k)|^2$ is proportional to the spectral density of the pulse. To this order of approximation, the field intensity is simply the sum of the intensities of the incident field and the field radiated by the atom. Stimulated emission or absorption of the incident field is absent since we have neglected interactions between the incident probe field and the atom. The second term in Eq. (14) is of order γ'_2/γ_2 smaller than the first and can be neglected.

$$\langle E_{-}(t_{1})E_{-}(t_{2})E_{+}(t_{2})E_{+}(t_{1})\rangle = \left(\frac{\hbar\omega_{0}}{2\epsilon_{0}A}\right) \left(\frac{\hbar\gamma_{2}'}{\mu}\right)^{2} \frac{|\beta_{2}|^{2}}{\gamma_{2}c} [|f(t_{1})|^{2}|w(t_{2})|^{2} + |f(t_{2})|^{2}|w(t_{1})|^{2} + 2\operatorname{Re}[f^{*}(t_{1})w(t_{1})f(t_{2})w^{*}(t_{2})]], \quad (17)$$
where

$$w(t) = \sqrt{\gamma_2} e^{-\gamma t} \Theta(t) \tag{18a}$$

and Eqs. (5), (6), and (8) were used. Note that w(t) has been defined such that

$$\int_{-\infty}^{\infty} dt |w(t)|^2 = 1.$$
 (19)

Combining Eqs. (9), (10), (12), and (17), we obtain

$$g^{(2)}(t_1, t_2) = \frac{\gamma_2'}{\gamma_2} |\beta_2|^2 \frac{|f(t_1)|^2 |w(t_2)|^2 + |f(t_2)|^2 |w(t_1)|^2 + 2\operatorname{Re}[f^*(t_1)w(t_1)f(t_2)w^*(t_2)]}{|f(t_1)|^2 |f(t_2)|^2}$$
(20)

and

with t_0

$$g^{(2)} = 2\frac{\gamma'_2}{\gamma_2}|\beta_2|^2 \left(1 + \left|\int_{-\infty}^{\infty} dt f(t)w^*(t)\right|^2\right).$$
(21)

The terms involving $f(t)w^*(t)$ can be interpreted as constructive HBT interference, with the maximum value of $g^{(2)}$ obtained if f(t) = w(t). The origin of this interference term can be traced to terms in Eq. (17) of the form

$$\langle E_{-}^{(0)}(t_{1})E_{-}^{(\text{Source})}(t_{2})E_{+}^{(0)}(t_{2})E_{+}^{(\text{Source})}(t_{1})\rangle \text{ or } \langle E_{-}^{(\text{Source})}(t_{1})E_{-}^{(0)}(t_{2})E_{+}^{(\text{Source})}(t_{2})E_{+}^{(0)}(t_{1})\rangle,$$

which are nonvanishing only for overlapping pulses. In effect, HBT terms of this nature are responsible for all the interference effects discussed in this paper. To examine the qualitative dependence of $g^{(2)}$ on pulse characteristics, we choose an exponentially decreasing pulse envelope,

$$f_{\exp}(t) = \sqrt{\Gamma} e^{-i\Delta t} e^{-\Gamma(t-t_0)/2} \Theta(t-t_0), \qquad (22)$$

$$= -X_0/c > 0$$
 and

$$\Delta = \bar{\omega} - \omega_0. \tag{23}$$

This corresponds to a pulse having bandwidth Γ and central frequency $\bar{\omega}$, whose wave front is located a distance X_0 to the left of the atom at t = 0. In that case,

$$g^{(2)} = 2\frac{\gamma_2'}{\gamma_2} |\beta_2|^2 \left(1 + \frac{4\Gamma\gamma_2 e^{-\gamma_2 t_0}}{(\Gamma + \gamma_2)^2 + 4\Delta^2} \right).$$
(24)

There is a factor of two "bump" in $g^{(2)}$ when $\Delta = 0$; $t_0 = 0$; $\Gamma = \gamma_2$, compared to the case when either $\gamma_2 t_0 \gg 1$ or $|\Delta| \gg \gamma_2$ or $\Gamma \gg \gamma_2$. Corrections to $g^{(2)}$ resulting from atom-input field interactions have been neglected are smaller by a factor of order $\gamma'_2/\gamma_2 \ll 1$. Thus, the bump in $g^{(2)}$ can be explained entirely as a result of constructive HBT interference.

Field intensity

You might ask whether the bump is correlated with stimulated emission; that is, are the optimal conditions for producing the bump the same as those for maximizing the field intensity in the forward direction. To answer this question, we must calculate how the field intensity given in Eq. (14) is modified by atom-input field interactions. The Hamiltonian in an interaction representation is

$$H^{I}(t) = \hbar g \sum_{k} [\sigma_{+}(t)a_{k}(t)e^{-i(\omega_{k}-\omega_{0})t} - a_{k}^{\dagger}(t)\sigma_{-}(t)e^{i(\omega_{k}-\omega_{0})t}],$$
(25)

where

$$g = -i \left(\frac{\omega_0}{2\hbar\epsilon_0 AL}\right)^{1/2} \mu = -i \sqrt{\gamma_2' \frac{c}{L}}$$
(26)

is a coupling constant and $a_k(t)$ is an interaction representation Heisenberg operator. Using standard techniques [10], we find that the time evolution equation for σ_+ is

$$\dot{\sigma}_{+} = ig \sum_{k} a_{k}^{\dagger} [\sigma_{22}(t) - \sigma_{11}(t)] e^{i(\omega_{k} - \omega_{0})t} - \gamma \sigma_{+}(t)$$

$$\approx ig \sum_{k} a_{k}^{\dagger} [2\sigma_{22}(0)e^{-\gamma_{2}t}\Theta(t) - 1] e^{i(\omega_{k} - \omega_{0})t} - \gamma \sigma_{+}(t), \quad (27)$$

where $a_k^{\dagger} \equiv a_k^{\dagger}(0)$. Including the lowest-order corrections to $\sigma_+(t)$ produced by the probe field, we find

$$\sigma_{+}(t) \approx \sigma_{+}(0)e^{-\gamma t}\Theta(t) + ig\sum_{k} a_{k}^{\dagger} \int_{0}^{t} dt' e^{i(\omega_{k}-\omega_{0})t'}e^{-\gamma(t-t')} \times [2\sigma_{22}(0)e^{-\gamma_{2}t'} - 1]\Theta(t).$$
(28)

When this is substituted into Eqs. (6) and (4), and the sums converted to integrals, we obtain

$$I_{N}(t) = I_{N}^{(0)}(t) + \frac{\gamma_{2}'}{\gamma_{2}} \bigg(f(t) \int_{0}^{t} dt' f^{*}(t') e^{-\gamma(t-t')} \times [2|\beta_{2}|^{2} e^{-\gamma_{2}t'} - 1] + \text{c.c.} \bigg),$$
(29)

where

$$I_N^{(0)}(t) = \frac{1}{\gamma_2} |f(t)|^2 + \frac{\gamma_2'}{\gamma_2^2} |\beta_2|^2 |w(t)|^2.$$
(30)

For the specific choice of f(t) given in Eq. (22),

$$I_{N}(t) = \frac{\Gamma}{\gamma_{2}} e^{-\Gamma(t-t_{0})} \Theta(t-t_{0}) + \frac{\gamma_{2}'}{\gamma_{2}} e^{-\gamma_{2}t} \Theta(t) + \frac{8\Theta(t-t_{0})\Gamma\gamma_{2}'|\beta_{2}|^{2}}{\gamma_{2}[(\Gamma+\gamma_{2})^{2}+4\Delta^{2}]} \\ \times \begin{bmatrix} (\gamma_{2}+\Gamma)\{\cos\left[\Delta(t-t_{0})\right]e^{-\gamma(t+t_{0})}e^{-\Gamma(t-t_{0})/2} - e^{-\gamma_{2}t}e^{-\Gamma(t-t_{0})}\} \\ +2\Delta\sin\left[\Delta(t-t_{0})\right]e^{-\gamma(t+t_{0})}e^{-\Gamma(t-t_{0})/2} \end{bmatrix} \\ - \frac{4\Theta(t-t_{0})\Gamma\gamma_{2}'}{\gamma_{2}[(\Gamma-\gamma_{2})^{2}+4\Delta^{2}]} \begin{bmatrix} (\Gamma-\gamma_{2})\{\cos\left[\Delta(t-t_{0})\right]e^{-(\gamma+\Gamma)(t-t_{0})/2} - e^{-\Gamma(t-t_{0})}\} \\ +2\Delta\sin\left[\Delta(t-t_{0})\right]e^{-(\gamma+\Gamma)(t-t_{0})/2} \end{bmatrix}.$$
(31)

Even if the atom is excited initially ($\beta_2 = 1$) and the probe field is chosen so that its spatiotemporal profile mirrors that of the atomic emission ($\Gamma = \gamma_2$, $\Delta = 0$, $t_0 = 0$), the output field intensity,

$$I_{N}(t) \sim e^{-\gamma_{2}t}\Theta(t) + \frac{\gamma_{2}'}{\gamma_{2}}e^{-\gamma_{2}t}\Theta(t) + 2\frac{\gamma_{2}'}{\gamma_{2}}\Theta(t)e^{-\gamma_{2}t}[2(1-e^{-\gamma_{2}t})-\gamma_{2}t],$$
(32)

does not match that of the input probe field.

The (dimensionless) total energy deposited into the detector mode is

$$W_N = \gamma_2 \int_0^\infty I_N(t)dt = 1 + \frac{\gamma_2'}{\gamma_2} |\beta_2|^2 + \frac{\gamma_2'}{\gamma_2} \frac{4\gamma_2}{(\Gamma + \gamma_2)^2 + 4\Delta^2} [2\Gamma |\beta_2|^2 e^{-\gamma_2 t_0} - (\gamma_2 + \Gamma)].$$
(33)

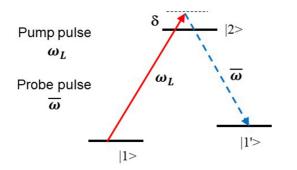


FIG. 1. Level scheme for Raman excitation. The central frequency of the pump pulse is $\omega_L = \omega_{21} + \delta$ and that of the probe pulse is $\bar{\omega} = \omega_L - \omega_{1'1}$.

The change in the incident pulse energy is of order $\gamma'_2/\gamma_2 \ll 1$. Note that if $\Delta = 0$; $|\beta_2|^2 = 1$; $t_0 = 0$; $\Gamma = \gamma_2$ (conditions to optimize the coincidence count bump), the interference term vanishes! In fact, to maximize the intensity you need to take $\Delta = 0$; $|\beta_2|^2 = 1$; $t_0 = 0$; $\Gamma = 3\gamma_2$, conditions which leads to a much smaller coincidence count rate than the optimal one. As long as $\rho_{22}(0) = |\beta_2|^2 \neq 0$, the coincidence count bump is maximal if $\Delta = 0$; $t_0 = 0$; $\Gamma = \gamma_2$, regardless of whether or not the interference term is positive (stimulated emission) or negative (absorption).

However, the bump height in W_N resulting from stimulated processes when the pulses overlap is equal to the corresponding bump height in $g^{(2)}$. Although the increase in $g^{(2)}$ when the pulses overlap in this case is not *caused* by stimulated emission, it is correlated with the increased intensity produced by stimulated processes. In some sense, the increase in coincidence counts is not classic HBT insofar as absorption or stimulated emission accompanies the increase in coincidence counts, but any modification of $g^{(2)}$ produced by absorption or stimulated emission is negligibly small.

III. SINGLE-ATOM RAMAN CONFIGURATION

Next we consider the Raman scheme shown in Fig. 1 for a three-level atom having states $|1\rangle$, $|1'\rangle$, and $|2\rangle$. States $|1\rangle$ and $|1'\rangle$ have the same parity, which is opposite that of state $|2\rangle$. A classical pulsed field having central frequency ω_L drives the 1–2 transition in an atom that is located at X = 0. This pump field is detuned from the atomic transition by an amount $\delta = \omega_L - \omega_{21} \gg \gamma_2$, where γ_2 is the excited state decay rate. In the absence of any input fields, there is Raman emission centered at frequency $\bar{\omega} = \omega_L - \omega_{1'1}$. An input single-photon pulse field is incident along the X axis, whose central frequency is taken to be equal to $\bar{\omega}$. This physical system, while not the same as that encountered in down-conversion experiments, shares many of its properties.

As in the previous section, we use an effective 1-D Hamiltonian which, in the interaction representation, can be taken as

$$H^{I}(t) = \hbar[\chi(t)\sigma_{21}(t)e^{-i\delta t} + \chi^{*}(t)\sigma_{12}(t)e^{i\delta t}] + \hbar g_{1} \sum_{k} \sigma_{21'}(t)a_{k}(t)e^{-i(\omega_{k}-\omega_{21'})t} - a_{k}^{\dagger}(t)\sigma_{1'2}(t)e^{i(\omega_{k}-\omega_{21'})t},$$
(34)

where $\chi(t)$ is one half the Rabi frequency associated with the classical pump field that drives the 1–2 transition, the $\sigma(t)$'s are raising or lowering interaction representation Heisenberg operators,

$$g_1 = -i \left(\frac{\omega_{21'}}{2\hbar\epsilon_0 AL}\right)^{1/2} \mu_{21'} = -i \sqrt{\gamma_{2,1'}^{\prime} \frac{c}{L}}$$
(35)

and

$$\gamma_{2,1'}' = \frac{\omega_{21'}\mu_{21'}^2}{2\hbar\epsilon_0 Ac} = \frac{3\pi}{2} \frac{1}{k_{21'}^2 A} \gamma_{2,1'},$$
(36)

where $\mu_{21'}$ is a dipole moment matrix element (assumed real and positive). The quantity $\gamma_{2,1'}$ is the spontaneous decay rate on the 2–1' transition and $\gamma'_{2,1'}/\gamma_{2,1'}$ is the fraction of spontaneous emission on the 2–1' transition that goes into the mode volume of the detector, which itself is matched to the mode volume of the single-photon input probe pulse. Terms involving vacuum field interactions on the 2–1 transition have not been included. Such terms give rise to Rayleigh scattering of the pump field, which is not of interest here. The corresponding source-field result in this 1D model is

$$E_{+}(t) = E_{+}^{(0)}(t) + E_{+}^{(\text{Source})}(t), \qquad (37)$$

where

$$E_{+}^{(0)}(t) = i \frac{\hbar}{\mu_{21'}} \sqrt{\frac{c}{L} \gamma_{2,1'}'} \sum_{k} a_k e^{-i\omega_k t},$$
 (38)

$$E_{+}^{(\text{Source})}(t) = i \frac{\hbar}{\mu_{21'}} \gamma_{2,1'}' \sigma_{1'2}(t) e^{-i\omega_{21'}t}.$$
 (39)

Recall that all times are actually retarded times.

A. Equations of motion

Perturbation theory is used and only those terms are retained that lead to nonzero values when expectation values are taken with an initial state vector

$$\psi(0)\rangle = \sum_{k} b_k a_k^{\dagger} |1;0\rangle, \qquad (40)$$

where $|1;0\rangle$ is the eigenket for the atom to be in level 1 and the field to be in its vacuum state. Moreover, we set

$$\sigma_{11}(t) \approx \sigma_{11}(0) = 1.$$
 (41)

The classical pump pulse is taken to be a smooth pulse starting at t = 0 and ending at time T, with $\delta T \gg 1$ and $\delta \gg \gamma_2$. As such the excited state amplitude adiabatically follows the classical pulse and

$$\sigma_{21}(t) \approx \frac{\chi^*(t)e^{i\delta t}}{\delta}.$$
 (42)

The equations of motion for the other operators that will be needed are

$$\dot{\sigma}_{1'2}(t) = -i\chi(t)e^{-i\delta t}\sigma_{1'1}(t);$$
(43a)
$$\dot{\sigma}_{1'1}(t) = ig_1 \sum a_k e^{-i(\omega_k - \omega_{21'})t}\sigma_{21}(t)$$

$$\approx ig_1 \frac{\chi^*(t)}{\delta} \sum_k a_k e^{-i(\omega_k - \bar{\omega})t}.$$
 (43b)

In these expressions, terms have been neglected that correspond to any off-resonant driving of the 1'-2 transition by the probe field in the absence of the classical field.

To evaluate $g^{(2)}$, we neglect all atom-probe field interactions. In that case,

$$\sigma_{1'2}(t) \approx \frac{\chi(t)}{\delta} e^{-i\delta t} \sigma_{1'1}(0).$$
(44)

Then the calculation proceeds exactly as before [note that $\sigma_{21'}(t)\sigma_{1'2}(t) \sim \sigma_{11'}(0)\sigma_{1'1}(0) = \sigma_{11}(0) \approx 1$] and we find

$$g^{(2)}(t_1, t_2) = \frac{\gamma'_{2,1'}}{\Gamma_R} \frac{|f(t_1)|^2 |w_R(t_2)|^2 + |f(t_2)|^2 |w_R(t_1)|^2 + 2\operatorname{Re}\left[f^*(t_1)w_R(t_1)f(t_2)w_R^*(t_2)\right]}{|f(t_1)|^2 |f(t_2)|^2},$$
(45)

where

$$w_R(t) = \sqrt{\Gamma_R} \frac{\chi(t)}{\delta} \Theta(t), \qquad (46)$$

$$\Gamma_R^{-1} = \int_0^\infty dt \, \frac{|\chi(t)|^2}{\delta^2},\tag{47}$$

and $f(t_1)$ is defined in Eq. (15). Note that, as defined,

$$\int_{-\infty}^{\infty} dt |w_R(t)|^2 = 1.$$
 (48)

The time-integrated correlation function is

$$g^{(2)} = 2\frac{\gamma'_{2,1'}}{\Gamma_R} \left(1 + \left| \int_0^\infty dt \, f(t) w_R^*(t) \right|^2 \right). \tag{49}$$

Clearly, if you match the temporal profile of the input pulse with that of the classical field, you will get twice the coincidence counts when the pulses overlap compared to the case when they do not overlap. Thus, just as in the previous case, the increase in coincidence counts can be attributed to HBT interference.

C. Field Intensity

Although the increase in coincidence counts is not linked directly to stimulated emission, we would like to see to what extent stimulated emission is present in this Raman configuration. To do so we must calculate the field intensity. Such a calculation can be carried out in two ways—the easy way and the hard way.

1. Source-field approach

The easy way is to use the source-field approach to evaluate

$$I(t) = 2\epsilon_0 c A \langle [E_-^{(0)}(t) + E_-^{(\text{Source})}(t)] [E_+^{(0)}(t) + E_+^{(\text{Source})}(t)] \rangle,$$
(50)

where the field operators are defined in Eqs. (38) and (39). If atom-input field interactions are neglected, then the cross terms vanish and

$$I_{N}^{(0)}(t) \equiv \frac{I^{(0)}(t)}{\hbar\omega_{21'}\Gamma_{R}} = \frac{|f(t)|^{2}}{\Gamma_{R}} + \frac{\gamma_{2,1'}'}{\Gamma_{R}^{2}}|w_{R}(t)|^{2}.$$
 (51)

The integrated dimensionless intensity is

$$W_N^{(0)} = \Gamma_R \int_0^\infty I_N^{(0)}(t) dt = 1 + \frac{\gamma'_{2,1'}}{\Gamma_R}.$$
 (52)

To evaluate the effects of atom-input field interactions, we must solve Eq. (43) to next order. Doing so, we find

$$\delta\sigma_{1'2}(t) = ig_1 \frac{\chi(t)}{\delta^2} e^{-i\delta t} \int_0^t \sum_k a_k \chi^*(t') e^{-i(\omega_k - \bar{\omega})t'} dt'.$$
 (53)

Using this result in Eq. (50), we find that the cross terms give an additional contribution, leading to a total dimensionless intensity given by

$$I_{N}(t) = \frac{|f(t)|^{2}}{\Gamma_{R}} + \frac{\gamma'_{2,1'}}{\Gamma_{R}^{2}} |w_{R}(t)|^{2} + 2\frac{\gamma'_{2,1'}}{\Gamma_{R}^{2}} \operatorname{Re}\left[f^{*}(t)w_{R}(t)\int_{0}^{t} f(t')w_{R}^{*}(t')dt'\right].$$
 (54)

The integrated dimensionless intensity is

$$W_N = 1 + \frac{\gamma'_{2,1'}}{\Gamma_R} + \frac{\gamma'_{2,1'}}{\Gamma_R} \left| \int_0^\infty f(t) w_R^*(t) dt \right|^2.$$
(55)

The intensity at time *t* is a function only of the retarded time, as is to be expected. The "interference" term always represents gain, insofar as its time integral is always positive. There is a bump in the dimensionless integrated intensity that is equal to 1/2 the bump height in $g^{(2)}$. As in the first example, the output field intensity does not have the same spatiotemporal shape as the input probe field intensity.

2. State vector approach

The details of the this calculation are given in Appendix A. Here we simply summarize the results. In a state vector approach, the field operators are time-independent Schrödinger operators and all the time dependence is in the state vector. When the state vector is calculated to order $|\chi(t)|/\delta$, the resultant change in field intensity, which is of order $|\chi(t)|^2/\delta^2$, has spurious terms. For example, if the probe pulse does not overlap with the pump pulse, then the intensity calculated in this fashion has a term that corresponds to gain for the probe pulse, which is physically impossible under these conditions. Moreover, the contribution to the change in the integrated intensity in this limit is twice the correct result! It is necessary to go to order $|\chi(t)|^2/\delta^2$ in the *state vector* to get the correct result for the intensity to order $|\chi(t)|^2/\delta^2$. The interference term between the zeroth order and the $|\chi(t)|^2/\delta^2$ contributions to the state vector removes the spurious terms in the field intensity and restores the correct result given in Eq. (54).

IV. PHASE-MATCHED EMISSION

The correlation functions associated with phase-matched emission from an atomic ensemble differ in a qualitative manner from those that have been considered so far. To illustrate the underlying physics, we consider one of the simplest types of phase-matched emission involving only stimulated emission, nondegenerate four-wave mixing. The level scheme is the same as for the single-atom Raman case shown in Fig. 1. At time t = 0, a pump pulse is applied, consisting of two classical field pulses propagating in the $\pm X$ direction that arrive simultaneously in the sample. The fields comprising this pump pulse have propagation vectors \mathbf{k}_{0A} and \mathbf{k}_{0B} and frequencies $\omega_{0A} = k_{0A}c$ and $\omega_{0B} = k_{0B}c$, with

$$(\omega_{0A} - \omega_{0B}) - \omega_{1'1} \approx 0. \tag{56}$$

For an *N*-atom ensemble, the pump pulse prepares the initial atomic state vector,

$$|\psi(0)\rangle_A = \prod_{j=1}^N (\alpha |1\rangle_j + \beta |1'\rangle_j e^{i\mathbf{k}_0 \cdot \mathbf{R}_j}), \tag{57}$$

where the effective propagation vector is

$$\mathbf{k}_0 = \mathbf{k}_{0A} - \mathbf{k}_{0B}.\tag{58}$$

Levels 1 and 1' are two sublevels of the ground state manifold, such that $k_{1'1} = \omega_{1'1}/c$ can be set equal to zero. It is assumed that $|\beta|^2 \ll 1$.

Following excitation, a second pump field arrives on the sample and drives the 1 - 2 transition in each atom. This field has propagation vector $\mathbf{k}_D \approx \mathbf{k}_{0A}$, frequency $\omega_D = k_D c$, and can lead to phase-matched emission on the 1'-2 transition having propagation vector $\mathbf{k}_s = \mathbf{k}_D - \mathbf{k}_0$ and frequency $\omega_s = \omega_D$, provided $\omega_s \approx k_s c$. That is, the requirement for phase matching is $|\mathbf{k}_D - \mathbf{k}_0|c \approx \omega_D$. To separate the Raman emission from the second pump pulse alone, we take $\mathbf{k}_{0A} \approx -\mathbf{k}_{0B}$, such that $\mathbf{k}_0 \approx 2\mathbf{k}_{0A}$ and $\mathbf{k}_s \approx -\mathbf{k}_{0A}$. It is assumed that the pump pulses interact individually with each atom - that is, the atomic density is sufficiently low to neglect any cooperative effects between the atoms.

In addition to the pump pulses, there is a single-photon pulse incident in the \mathbf{k}_s direction having central frequency ω_s . We want to see if this pulse is amplified by the medium and how any amplification is related to the second-order correlation function of the outgoing fields in the phase-matched direction.

Using an effective 1D model for the probe field, the Hamiltonian in the interaction representation can be taken as

$$H^{I}(t) = +\hbar \sum_{j=1}^{N} \left[\chi_{D}(t) \sigma_{21}^{(j)}(t) e^{-i\delta t} e^{i\mathbf{k}_{D} \cdot \mathbf{R}_{j}} + \chi_{D}^{*}(t) \sigma_{12}^{(j)}(t) e^{i\delta t} e^{-i\mathbf{k}_{D} \cdot \mathbf{R}_{j}} \right] + \hbar g_{1} \sum_{j=1}^{N} \sum_{k} \sigma_{21'}^{(j)}(t) a_{k}(t) e^{ik\hat{\mathbf{k}}_{s} \cdot \mathbf{R}_{j}} e^{-i(\omega_{k} - \omega_{21'})t} - \hbar g_{1} \sum_{j=1}^{N} \sum_{k} a_{k}^{\dagger}(t) e^{-ik\hat{\mathbf{k}}_{s} \cdot \mathbf{R}_{j}} \sigma_{1'2}^{(j)}(t) e^{i(\omega_{k} - \omega_{21'})t},$$
(59)

where $\chi_D(t)$ is one half the Rabi frequency associated with the classical pump field that drives the 1–2 transition,

$$\delta = \omega_D - \omega_{21},\tag{60}$$

is an atom-field detuning, the $\sigma(t)$'s are raising or lowering interaction representation Heisenberg operators, and g_1 and $\gamma'_{2,1'}$ are defined in Eqs. (35) and (36), respectively. It is assumed that any population in level 2 can be neglected. The only emission on the 1'–2 transition that we consider is linked to the 1–1' coherence created by the pump field that led to the initial state given in Eq. (57).

The corresponding source-field result for this 1D model is

$$E_{+}(t) = E_{+}^{(0)}(t) + E_{+}^{(\text{Source})}(t), \qquad (61)$$

where

$$E_{+}^{(0)}(t) = i \frac{\hbar}{\mu_{21'}} \sqrt{\frac{c}{L} \gamma_{2,1'}'} \sum_{k} a_k e^{-i\omega_k t}, \qquad (62)$$

$$E_{+}^{(\text{Source})}(t) = i \frac{\hbar}{\mu_{21'}} \gamma_{2,1'}' \sum_{j=1}^{N} \sigma_{1'2}^{(j)}(t) e^{-i\omega_{21'}t} e^{-i\mathbf{k}_{21'}\cdot\mathbf{R}_j}, \quad (63)$$

and

$$\mathbf{k}_{21'} = k_{21'} \mathbf{R} / R = \frac{k_{21'}}{k_s} \mathbf{k}_s \approx \mathbf{k}_s,$$

All times are retarded times relative to the center of the sample. The vector $\mathbf{k}_{21'}$ is in the direction of \mathbf{k}_s since the detector is placed in the phase-matched direction.

A. Equations of motion

Perturbation theory is used and only those terms are retained that lead to nonzero values when expectation values are taken with an initial state vector

$$|\psi(0)\rangle = \sum_{k} b_k a_k^{\dagger} |1;0\rangle, \qquad (64)$$

where $|1;0\rangle$ is the eigenket for all the atoms to be in level 1 and the field to be in its vacuum state. The sum over k is restricted to the direction of phase-matched emission and $\omega_k = kc$ is centered at the central frequency ω_s of the phase-matched emission. Moreover, we set

$$\sigma_{11}^{(j)}(t) \approx \sigma_{11}^{(j)}(0^+) \approx 1; \tag{65}$$

$$\sigma_{1'1}^{(j)}(0^+) \approx \sigma_{1'1}^{(j)}(0) + \beta^* e^{-i\mathbf{k}_0 \cdot \mathbf{R}_j},\tag{66}$$

where 0^+ is the time immediately following the initial excitation pulse. In other words, the initial pulse creates a phased, off-diagonal atomic operator.

The classical pump pulse having (half) Rabi frequency $\chi_D(t)$ is taken to be a smooth pulse starting at t = 0 and ending at time *T*, with $\delta T \gg 1$ and $\delta \gg \gamma_2 = \gamma_{2,1} + \gamma_{2,1'}$. The equations of motion for the operators that will be needed are

$$\dot{\sigma}_{1'2}^{(j)}(t) = -i\chi_D(t)e^{i\mathbf{k}_D \cdot \mathbf{R}_j}e^{-i\delta t}\sigma_{1'1}^{(j)}(t);$$

$$\dot{\sigma}_{1'j}^{(j)}(t) = -i\chi_D^*(t)\sigma_{1'j}^{(j)}(t)e^{i\delta t}e^{-i\mathbf{k}_D \cdot \mathbf{R}_j}$$
(67a)

$$+ ig_1 \sum_{j=1}^{N} \sum_{k} \sigma_{21}^{(j)}(t) a_k(t) e^{ik \hat{\mathbf{k}}_s \cdot \mathbf{R}_j} e^{-i(\omega_k - \omega_{21'})t};$$
(67b)

$$\dot{\sigma}_{12}^{(j)}(t) = -i\chi_D(t)e^{-i\delta t}e^{i\mathbf{k}_D\cdot\mathbf{R}_j} -ig_1\sum_k \sigma_{11'}^{(j)}(t)a_k(t)e^{ik\hat{\mathbf{k}}_s\cdot\mathbf{R}_j}e^{-i(\omega_k-\omega_{21'})t}, \quad (67c)$$

along with the adjoints of these equations. Terms involving population operators $\sigma_{22}^{(j)}$ and $\sigma_{1'1'}^{(j)}$ have been suppressed.

B. Second-order correlation function

First, we neglect all atom-input field interactions and retain only terms linear in $|\chi_D|$. In that approximation,

$$\sigma_{1'2}^{(j)}(t) \approx \frac{\chi_D(t)e^{-i\delta t}}{\delta} e^{i\mathbf{k}_D \cdot \mathbf{R}_j} \sigma_{1'1}^{(j)}(t);$$
(68a)

$$\sigma_{l'1}^{(j)}(t) \approx \sigma_{l'1}^{(j)}(0) \approx \sigma_{l'1}^{(j)}(0) + \beta^* e^{-i\mathbf{k}_0 \cdot \mathbf{R}_j}.$$
 (68b)

By combining Eqs. (63) and (68), we see that

$$E_{+}^{(\text{Source})}(t) = i \frac{\hbar}{\mu_{21'}} \gamma_{2,1'}' \sum_{j=1}^{N} \frac{\chi_D(t) e^{-i\delta t}}{\delta} e^{-i\omega_{21'}t} e^{i\mathbf{k}_D \cdot \mathbf{R}_j} e^{-i\mathbf{k}_{21'} \cdot \mathbf{R}_j} \\ \times \left[\sigma_{1'1}^{(j)}(0) + \beta^* e^{-i\mathbf{k}_0 \cdot \mathbf{R}_j}\right].$$
(69)

The term proportional to $\sigma_{1'1}^{(j)}(0)$ is similar to the one we encountered for Raman emission from a single atom. Since $\mathbf{k}_D - \mathbf{k}_{21'} \approx \mathbf{k}_0$, this term is not phase-matched. The second term *is* phase-matched and leads to a contribution that is of order $N|\beta|^2$ times larger than the $\sigma_{1'1}^{(j)}(0)$ term. We assume that

$$N|\beta|^2 \gg 1,\tag{70}$$

so that the phase-matched term is dominant. For perfect phase matching,

$$E_{+}^{(\text{Source})}(t) \approx i \frac{\hbar N}{\mu_{21'}} \gamma_{2,1'}' \frac{\chi_1(t) e^{-i\delta t}}{\delta} e^{-i\omega_{21'} t} \beta^*.$$
(71)

Then, following the same procedure used in the previous cases we find

$$g^{(2)}(t_1, t_2) = \frac{\gamma'_{2,1'}}{\Gamma_{\rm pm}} \frac{|f(t_1)|^2 |w_{\rm pm}(t_2)|^2 + |f(t_2)|^2 |w_{\rm pm}(t_1)|^2 + 2\operatorname{Re}[f^*(t_1)w_{\rm pm}(t_1)f(t_2)w_{\rm pm}^*(t_2)]}{|f(t_1)|^2 |f(t_2)|^2},\tag{72}$$

where

$$w_{\rm pm}(t) = N \sqrt{\Gamma_{\rm pm}} \frac{\chi_D(t)}{\delta} \beta^* \Theta(t), \tag{73}$$

$$\Gamma_{\rm pm}^{-1} = N^2 |\beta|^2 \int_0^\infty dt \; \frac{|\chi^D(t)|^2}{\delta^2},\tag{74}$$

f(t) is defined in Eq. (15), and

$$\int_{-\infty}^{\infty} dt \left| w_{\rm pm}(t) \right|^2 = 1.$$
(75)

Note that $g^{(2)}(t_1, t_2)$ is proportional to N^2 . The time-integrated correlation function is

$$g^{(2)} = 2\frac{\gamma'_{2,1'}}{\Gamma_{\rm pm}} \left(1 + \left| \int_0^\infty dt \, f(t) w_{\rm pm}^*(t) \right|^2 \right).$$
(76)

As in the Raman case, if you match the temporal profile of the input pulse with that of $w_{pm}(t)$, then you will get twice the coincidence counts when the pulses overlap compared to the case when they do not overlap. The increase in coincidence counts that can be attributed to HBT interference.

C. Field intensity

If we neglect interactions between the probe and the atoms and neglect absorption of the probe field, then the dimensionless field intensity is simply the sum of the contributions from the atoms and the probe field,

$$I_{N}^{(0)}(t) = \frac{I^{(0)}(t)}{\hbar\omega_{21'}\Gamma_{\rm pm}} = \frac{|f(t)|^2}{\Gamma_{\rm pm}} + \frac{\gamma_{2,1'}'}{\Gamma_{\rm pm}^2} |w_{\rm pm}(t)|^2.$$
(77)

033824-8

The integrated dimensionless intensity is

$$W_N^{(0)} = \Gamma_{\rm pm} \int_0^\infty I_N^{(0)}(t) dt = 1 + \frac{\gamma'_{2,1'}}{\Gamma_{\rm pm}}.$$
 (78)

Atom-input field interactions modify this result primarily in two ways. First there is the same type of stimulated emission that we encountered in the Raman problem. If the inequality in Eq. (70) is satisfied, then this contribution, proportional to N, can be neglected. In addition there can be some decrease in probe intensity resulting from Rayleigh scattering, also, proportional to N. This contribution can be neglected relative to the phase-matched contribution if

$$\frac{\gamma'_{2,1'}\gamma_{2,1'}T}{N\int_0^T dt \,|\chi_D(t)|^2} \ll 1.$$
(79)

In this limit, any bump in the dimensionless integrated intensity that occurs when the probe and pump pulses overlap is now negligibly small compared with the bump in $g^{(2)}$. In this respect, the situation is very close to HBT interference where stimulated emission plays no role.

V. CONCLUSIONS AND DISCUSSION

We have examined a number of problems in which a singlephoton pulse is incident on an atom or an ensemble of atoms. In each case, there can be a field radiated by the atom(s) in the absence of the input pulse. As such there is always the possibility that, when an input pulse is applied, there can be a nonvanishing value of the second-order correlation function of the output field. The output field consists of the input field, the field radiated by the atom(s) and interference terms which correspond to the modification of the input field produced by its interaction with the atom(s). In the weak coupling approximation adopted in this work, such interference terms have a negligible effect on the intensity of the input field. Moreover, to lowest order, they do not affect the number of coincidence counts measured by detectors that monitor the output field. Nevertheless, in all the cases studied, there can be an increase in coincidence counts when the input field overlaps with the field radiated by the atom(s). We have argued that such an increase is nothing more that constructive HBT or HOM interference.

Our results call into question some approaches used to analyze coincidence counts measured in certain down-conversion experiments [9]. The effective Hamiltonian in an interaction representation that describes down conversion is often taken as

$$H^{I}(t) = \hbar \sum_{k_{1},k_{2}} (B_{k_{1}k_{2}}(t)e^{-i(\omega_{L}-\omega_{k_{1}}-\omega_{k_{2}})t}a_{k_{1}}^{\dagger}a_{k_{2}}^{\dagger} + \text{adjoint}),$$
(80)

where $a_{k_1}^{\dagger}$ and $a_{k_2}^{\dagger}$ are creation operators for the signal and idler modes, respectively, ω_L is the frequency of the pump field, and *B* is a function that represents the entanglement between the signal and idler modes. Given the Hamiltonian Eq. (80), the source-field and state vector approaches developed in this paper can be used to evaluate both the signal and idler field intensities for an initial single-photon state that is incident in the direction of the signal photon probe pulse. As in the Raman case, the signal field intensity calculated to order B^2 using the state vector approach has spurious terms and does not agree with the correct result to order B^2 , which can be obtained using a source-field approach. However, the *idler* field intensity calculated to order B^2 using the lowest-order state vector *does* agree.

The increase in the idler signal intensity when the input probe field overlaps with the pump field is sometimes taken to be a *definitive* signature of stimulated emission [11]. It is our opinion that this assignment is not consistent with conventional definitions of stimulated emission. Consider the two level scheme shown in Fig. 2(a), in which a two photon pulsed pump field having central frequency $\omega_L \approx (\omega_{21} + \delta)/2$ drives the 1-2 transition. The field is off-resonant, but leads to Raman-like scattering on the 2-1' transition (level 1' now has parity opposite to that of level 1). A single-photon probe field having central frequency ω_L is also incident on the atom and can be time-delayed from the pump field. Clearly, there will be increased Raman emission when the probe field overlaps with the pump field. In effect, the probe field provides an additional channel for Raman scattering. In analogy with the logic followed in the analysis of down conversion experiments, the increase in Raman emission can be attributed to stimulated emission produced by the probe. However, in this case, the intensity of the probe field is actually reduced by its interaction with the atom, even if the transition rate from level 1 to 1' is increased. That is, you can call this stimulated emission if you wish, owing to the increased rate, but it is not the conventional use of the term, which we claim always

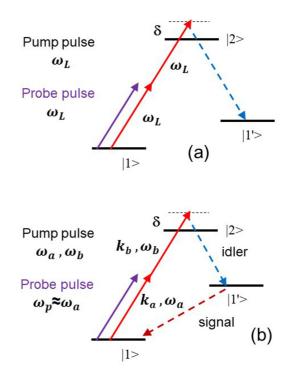


FIG. 2. Level schemes increased field intensity on an undriven transition. (a) Raman-like scheme (b) phase-matched emission at both the "signal" and "idler" frequencies.

involves the constructive interference of an incident field with the field scattered from a medium.

Now consider the same level scheme but with a two photon pump field that drives the 1–2 transition in an atomic ensemble—Fig. 2(b). In this case the pump field consists of two fields having propagation vectors \mathbf{k}_a and \mathbf{k}_b and frequencies $\omega_a = k_a c \approx \omega_{1'1}$ and $\omega_b = k_b c \approx \omega_{21'} + \delta$. In addition, there is a single-photon probe field that is incident on the ensemble having propagation vectors $\mathbf{k}_p \approx \mathbf{k}_a$ and frequency $\omega_p = k_p c \approx \omega_{1'1}$ that can be delayed relative to the pump field. In the absence of the probe field there is nonphased matched Raman emission at frequency $\omega_a + \omega_b - \omega_{1'1}$ in all directions. In addition there is phase-matched emission of correlated two-photon states, with one photon having wave vector approximately equal to $\mathbf{k}_1 \approx \mathbf{k}_b$ (idler) and the other approximately equal to $\mathbf{k}_2 \approx \mathbf{k}_b$ (signal) [12]. When the probe field does not overlap with the pump field, emission on the idler transition is unaffected by the presence of the probe field; however, when it does overlap, the phase-matched emission on the idler transition is increased. Again it is possible to say that the increased rate for phase-matched emission on the idler transition is a result of stimulated emission produced by the probe pulse (even though the probe intensity actually decreases), but we feel this corresponds to an unconventional definition.

Returning to down conversion, the increase in phase matched idler intensity is analogous to that for the level scheme shown in Fig. 2(b). It is not clear in the down-conversion experiments whether or not the probe field is amplified or absorbed by the medium. The relatively large increase in the idler intensity results from a corresponding increase in the rate for phase-matched emission, which we

have argued is not the conventional definition of stimulated emission. In Appendix B, we try to relate the approach taken in this paper with that followed in some of the analyses of down-conversion experiments.

ACKNOWLEDGMENTS

P.R.B. thanks P. Milonni and D. Steel for helpful discussions. This research was supported by the ARL Center for Distributed Quantum Information, AFOSR, and the National Science Foundation.

APPENDIX A: STATE VECTOR CALCULATION OF THE RAMAN FIELD INTENSITY AND $g^{(2)}$

In this Appendix, we repeat the field intensity calculation for the Raman problem using a state vector approach. That is we calculate $|\psi(t)\rangle$ using perturbation theory with the Hamiltonian Eq. (34) and then evaluate

$$I(t) = 2\epsilon_0 c A \langle \psi(t) | E_-(X_B) E_+(X_B) | \psi(t) \rangle, \qquad (A1)$$

where $E_+(X_B)$ is now the *time-independent* Schrödinger operator

$$E_{+}(X_{B}) = i \frac{\hbar}{\mu_{21'}} \sqrt{\frac{c}{L}} \gamma_{2,1'}' \sum_{k} a_{k} e^{ikX_{B}}$$
(A2)

and t is the nonretarded time (in this Appendix, t always refers to the nonretarded time and t_r to the retarded time). The perturbation theory result is

$$|\psi(t)\rangle = e^{-iH_0 t/\hbar} |\psi^I(t)\rangle, \tag{A3}$$

where

$$|\psi^{I}(t)\rangle = \begin{bmatrix} 1 - \frac{i}{\hbar} \int_{0}^{t} H^{I}(t') dt' \\ -\frac{1}{\hbar^{2}} \int_{0}^{t} H^{I}(t') dt' \int_{0}^{t'} H^{I}(t'') dt'' + \dots \end{bmatrix} |\psi(0)\rangle,$$
(A4)

$$H_0 = \hbar \omega_{21} |2\rangle \langle 2| + \hbar \omega_{1'1} |1'\rangle \langle 1'| + \sum_k \hbar \omega_k a_k^{\dagger} a_k, \qquad (A5)$$

and

$$H^{I}(t) = \hbar[\chi(t)\sigma_{21}e^{-i\delta t} + \chi^{*}(t)\sigma_{12}e^{i\delta t}] + \hbar g_{1} \sum_{k} \sigma_{21'}a_{k}e^{-i(\omega_{k}-\omega_{21'})t} - a_{k}^{\dagger}\sigma_{1'2}e^{i(\omega_{k}-\omega_{21'})t}.$$
(A6)

The operators in this equation are *time-independent* Schrödinger operators.

Let us define $|\psi_j^I(t)\rangle$ as the *j*th-order perturbation theory result. Thus,

$$\psi_0^I(t) \rangle = |\psi(0)\rangle = \sum_k b_k a_k^{\dagger} |1;0\rangle, \tag{A7}$$

$$\begin{aligned} \left|\psi_{1}^{I}(t)\right\rangle &= -\frac{i}{\hbar} \int_{0}^{t} H^{I}(t') dt' \left|\psi(0)\right\rangle \\ &= -i\hbar \int_{0}^{t} dt' \chi(t') \sigma_{21} e^{-i\delta t'} \left|\psi(0)\right\rangle \\ &\approx \frac{\chi(t)}{\delta} e^{-i\delta t} \sum_{k} b_{k} a_{k}^{\dagger} \left|2;0\right\rangle, \end{aligned} \tag{A8}$$

and

$$\begin{split} \left| \psi_{2}^{I}(t) \right\rangle &= -\frac{1}{\hbar^{2}} \int_{0}^{t} H^{I}(t') dt' \int_{0}^{t'} H^{I}(t'') dt'' \left| \psi(0) \right\rangle \\ &= -i \frac{g_{1}}{\delta} \sum_{k,k'} b_{k} \int_{0}^{t} dt' \, e^{i \left(\omega_{k'} - \bar{\omega} \right) t'} \chi(t') a_{k}^{\dagger} a_{k'}^{\dagger} |1'; 0\rangle. \end{split}$$

$$(A9)$$

We are interested in times t > T. Since the excitation adiabatically follows the field, the contribution given in Eq. (A8) vanishes. To this order of perturbation theory, the state vector consists of the original single photon state vector plus a two-photon state vector. In theories of down conversion, this two-photon component is said to constitute stimulated emission when the pulses overlap. To see if this is actually the case in the Raman problem, we need to evaluate this term.

From Eqs. (A1), (A3), (A7), and (A9), we find that there are two contributions to the signal,

$$I_{0}(t) = 2\epsilon_{0}cA \langle \psi_{0}^{I}(t) | e^{iH_{0}t/\hbar} E_{-}(X_{B}) E_{+}(X_{B}) e^{-iH_{0}t/\hbar} | \psi_{0}^{I}(t) \rangle$$
(A10)

and

$$I_{2}(t) = 2\epsilon_{0}cA \langle \psi_{2}^{I}(t) | e^{iH_{0}t/\hbar} E_{-}(X_{B}) E_{+}(X_{B}) e^{-iH_{0}t/\hbar} | \psi_{2}^{I}(t) \rangle.$$
(A11)

It is not overly difficult to show that

$$I_0(t) = \hbar \omega_{21'} |f(t_r)|^2,$$
 (A12)

in agreement with the first term in Eq. (51).

The second term is only a bit more difficult to calculate. It is equal to

$$I_{2}(t) = \frac{\hbar\omega_{21'}}{\delta^{2}} \left(\frac{c}{L}\right)^{2} \sum_{k_{1},k_{2},k,k',k'_{1},k'_{2}} \exp\left[i\left(\omega_{k'_{1}} + \omega_{k'_{2}} - \omega_{k_{1}} - \omega_{k_{2}}\right)t\right] \int_{0}^{t} dt'' \chi^{*}(t'') e^{-i(\omega_{k'_{1}} - \bar{\omega})t''} b_{k'_{2}}^{*} \int_{0}^{t} dt' \chi(t') e^{i(\omega_{k_{1}} - \bar{\omega})t'} b_{k_{2}} \\ \times e^{-ik'X_{B}} e^{ikX_{B}} \langle 0|a_{k'_{1}}a_{k'_{2}}a_{k'_{1}}^{\dagger}a_{k}a_{k_{1}}^{\dagger}a_{k_{2}}^{\dagger}|0\rangle.$$
(A13)

By applying the commutation relations for the creation and annihilation operators, we find that there are four terms that contribute,

A: $k = k_1; k' = k'_1; k_2 = k'_2;$

B:
$$k = k_2; k' = k'_1; k_1 = k'_2;$$
 (A15)

C:
$$k = k_1; k' = k'_2; k_2 = k'_1;$$
 (A16)

$$D: k = k_2; k' = k'_2; k_1 = k'_1.$$
(A17)

(A14)

Term A is evaluated as

$$I_{2A}(t) = \hbar \omega_{21'} \frac{\gamma'_{2,1'}}{\Gamma_R} |w_R(t_r)|^2,$$
(A18)

in agreement with the second term in Eq. (54). Terms B and C lead to

$$I_{2B}(t) + I_{2C}(t) = \hbar \omega_{21'} \frac{\gamma'_{2,1'}}{\Gamma_R} f^*(t_r) w_R(t_r) \int_0^t f(t') w_R^*(t') dt' + \text{c.c.}$$
(A19)

This equation *almost* agrees with the third term in Eq. (54), but the upper limit of the integral is t instead of t_r [recall that the t in Eq. (54) actually the retarded time t_r]. Moreover, there is an additional term,

$$I_{2D}(t) = \hbar \omega_{21'} \frac{\gamma'_{2,1'}}{\Gamma_R} |f(t_r)|^2 \int_0^t |w_R(t')|^2 dt', \qquad (A20)$$

not found in Eq. (54).

For arbitrary probe pulse characteristics and arbitrary retardation, both the change in intensity given by Eqs. (A18)-(A20) and the change in integrated intensity differ signifi*cantly* from the correct results given in Eqs. (54) and (55). For example, if the probe pulse does not overlap with the pump pulse, the $I_{2D}(t)$ term still corresponds to gain for the probe pulse, which is physically impossible. Moreover, the contribution to the change in the integrated intensity from the $I_{2D}(t)$ term leads to a result which is twice the correct result!

The differences between the source-field and state vector approaches can be resolved if we calculate the *fourth*-order contribution to $|\psi^{I}(t)\rangle$. The fourth-order contribution contains a term which can interfere with the zeroth-order contribution. There are two chains in the perturbation chain that contribute to this fourth-order contribution,

$$A: |1;1_{k}\rangle \rightarrow |2;1_{k}\rangle \rightarrow |1';1_{k},1_{k'}\rangle \rightarrow |2;1_{k'}\rangle \rightarrow |1;1_{k'}\rangle;$$
(A21)

$$B: |1; 1_k\rangle \to |2; 1_k\rangle \to |1'; 1_k, 1_{k'}\rangle \to |2; 1_k\rangle \to |1; 1_k\rangle,$$
(A22)

where $|1_k\rangle = a_k^{\dagger}|0\rangle$ is a field state with a photon in mode k and $|1_k, 1_{k'}\rangle = a_k^{\dagger} a_{k'}^{\dagger} |0\rangle$ is a field state with one photon in mode k and another in mode k'. In evaluating the last two terms in the perturbation chains, we encounter factors of the form

$$-\int_0^t dt' \chi^*(t') e^{i\delta t'} \int_0^{t'} dt'' G(t''),$$

where G(t'') is some function. We switch the order of integration to obtain

$$-i\int_0^t dt'' G(t'')\int_{t''}^t dt' \chi^*(t')e^{i\delta t'} \approx \int_0^t dt'' \chi^*(t'')e^{i\delta t''} G(t'')/\delta,$$

where we have again used the adiabatic following approximation. In this manner we find

$$\psi_{4A}^{I}(t) \rangle = -\frac{|g_{1}|^{2}}{\delta^{2}} \sum_{k_{1},k_{2}} \int_{0}^{t} dt' \chi^{*}(t') e^{-i(\omega_{k_{1}}-\bar{\omega})t'} \\ \times \int_{0}^{t'} dt'' e^{i(\omega_{k_{2}}-\bar{\omega})t''} \chi(t'') b_{k_{1}} |1;1_{k_{2}}\rangle; \quad (A23)$$

$$\begin{split} \left| \psi_{4B}^{I}(t) \right\rangle &= -\frac{|g_{1}|^{2}}{\delta^{2}} \sum_{k_{1},k_{2}} \int_{0}^{t} dt' \chi^{*}(t') e^{-i(\omega_{k_{2}}-\bar{\omega})t'} \\ &\times \int_{0}^{t'} dt'' \, e^{i(\omega_{k_{2}}-\bar{\omega})t''} \chi(t'') b_{k_{1}}|1;1_{k_{1}}\rangle. \end{split}$$
(A24)

In forming I(t) there are now cross terms of the type

$$I_{c}(t) = 2\epsilon_{0}cA \langle \psi_{0}^{I}(t) | e^{iH_{0}t/\hbar} E_{-}(X_{B}) E_{+}(X_{B}) e^{-iH_{0}t/\hbar} | \psi_{4}^{I}(t) \rangle + \text{c.c.}, \qquad (A25)$$

with

$$\left\langle \psi_{0}^{I}(t) \right| = \sum_{k_{1}'} b_{k_{1}'}^{*} \left\langle 1; 1_{k_{1}'} \right|.$$
 (A26)

In evaluating this expression for the A contribution to $|\psi_4^I(t)\rangle$, we find that the sum over k_2 gives rise to the delta function $\delta(t'' - t + X_B/c)$, which implies that the integral over t'' contributes only if $t' > t - X_B/c$. Using this result and Eq. (15), we obtain

$$I_{cA}(t) = -\hbar\omega_{21'} \frac{\gamma'_{2,1'}}{\Gamma_R} f^*(t_r) w_R(t_r) \int_{t-X_B/c}^t f(t') w_R^*(t') dt' + \text{c.c.},$$
(A27)

which exactly cancels the nonretarded part of Eq. (A19). Similarly in evaluating the expression for the *B* contribution, we find that the sum over k_1 gives rise to the δ function $\delta(t'' - t')$ leading to

$$I_{cB}(t) = -\frac{1}{2}\hbar\omega_{21'}\frac{\gamma'_{2,1'}}{\Gamma_R}|f(t_r)|^2 \int_0^t |w_R(t')|^2 dt' + \text{c.c.}, \quad (A28)$$

which exactly cancels the D contribution in Eq. (A20) [the factor of 1/2 arises when we set $\int_0^{t'} dt'' \delta(t'' - t') G(t'') =$ G(t')/2]. Thus the source-field and state vector approaches now agree.

It is also possible to calculate $g^{(2)}$ using the state vector approach if the field operators are evaluated at the same time, but different positions, that is

$$g^{(2)}(t, X_1, X_2) = \frac{\langle E_-(X_1, t)E_-(X_2, t)E_+(X_2, t)E_+(X_1, t)\rangle}{\langle E_-(X_1, t)E_+(X_1, t)\rangle\langle E_-(X_2, t)E_+(X_2, t)\rangle}.$$
(A29)

With this definition all times are the same so we can replace this expression by one in which all the field operators are time-independent Schrödinger operators and the expectation value is evaluated using the time-dependent ket $|\psi^{I}(t)\rangle$. Using Eq. (A2), we are then able to show that we can reproduce Eq. (45) with t_1 replaced by $t - X_1/c$ and t_2 by $t - X_2/c$. Although the results are the same, it is much easier to interpret the results using the source-field approach since the incident input field and source-field separate in a natural fashion. With the state vector approach, the two contributions are intermixed so it is not as easy to conclude that the value of $g^{(2)}$ that is obtained arises as if the input field and atom do not interact.

(A24)

APPENDIX B: SINGLE-MODE CALCULATION OF THE RAMAN FIELD INTENSITY AND $g^{(2)}$

In this Appendix, we calculate the correlation function and the field intensity for the Raman problem using the types of formalisms often employed in theories of down conversion

1. Operator approach

In theories of down conversion and in "beam splitter" theories of attenuators or amplifiers [8,9,13], an input-output approach is taken, based in part on a "single-mode" approach. Quotation marks are used since the theories are often used for pulsed fields in which the field annihilation and creation operators actually correspond to localized operators of a pulsed field. Consider first the case when the probe and pump fields overlap, that is the probe field has the same temporal shape as the pump field,

$$\chi(t) = \eta \chi_0 f(t), \tag{B1}$$

where η is constant having units of \sqrt{t} . Since the fields have the same spatiotemporal profile, it is assumed in such approaches that the field emitted by the atoms in the absence of the probe field is emitted into the same mode as the probe field.

With this assumption, the output annihilation operator is written as

$$a_{\rm out} = Ta_{\rm in} + Rb^{\dagger}, \tag{B2}$$

where

$$R = \frac{\gamma'_{2,1'}}{\delta^2} \int_0^\infty dt \, |\chi(t)|^2$$
(B3)

and $b^{\dagger} = \sigma_{1'1}$ is an atomic raising operator satisfying $[b, b^{\dagger}] = \sigma_{11} - \sigma_{1'1'} \approx 1$. To maintain the commutation relation $[a_{\text{out}}, a_{\text{out}}^{\dagger}] = 1$, it is then necessary that

$$T = \sqrt{1 + R^2} \approx 1 + R^2/2.$$
 (B4)

The initial state is one in which there is a single photon in the input mode and the atom is in state $|1\rangle$. The b^{\dagger} operator leads to atomic excitation, that is $b^{\dagger}|1\rangle = |1'\rangle$. In this limit, to lowest order in R^2 , it follows that the dimensionless integrated field intensity W_N and the time-integrated second-order correlation function $g^{(2)}$ are

$$W_N = \langle a_{\text{out}}^{\dagger} a_{\text{out}} \rangle \sim (1 + R^2) + R^2 = 1 + 2R^2;$$
 (B5a)

$$g^{(2)} = \frac{\langle a_{\text{out}}^{\dagger} a_{\text{out}}^{\dagger} a_{\text{out}} \rangle}{\langle a_{\text{out}}^{\dagger} a_{\text{out}} \rangle^2} \sim 4R^2.$$
(B5b)

However, if the probe pulse arrives at the atom after the pump field is no longer present, then it is assumed that the atomic emission and the probe field emission are in distinct field modes $[a_{out}(probe) = a_{in}(probe)]$ and

$$W_N' \sim 1 + R^2, \tag{B6a}$$

$$g^{(2)'} = 2R^2.$$
 (B6b)

In this picture, the increase in W_N resulting from stimulated emission $[W_N - W'_N = R^2]$ is correlated with the increase in the coincidence counts $[g^{(2)} - g^{(2)\prime} = 2R^2]$. Although this approach produces the correct limits for overlapping and nonoverlapping pulses, it does not properly account for the time-dependence of the operators.

To see why this is the case and to make connection between the "single-mode" and exact formulations, we follow Loudon [13] and define a time-dependent annihilation operator by

$$a_{\text{out}}(t) = \sqrt{\frac{c}{L}} \sum_{\omega} a_{\omega}(t) e^{-i\omega t} \to \frac{1}{\sqrt{2\pi}} \int d\omega \, a(\omega, t) e^{-i\omega t}.$$
(B7)

The continuous field operator $a(\omega, t)$ is a function of t owing to the atom-field interactions. As a consequence, $[a_{out}(t), a_{out}^{\dagger}(t')] \neq \delta(t - t')$, as is the case for continuous *free-field* operators. In perturbation theory, it then follows from Eqs. (B7), (38), (39), and (43) that

$$a_{\text{out}}(t) = a_0(t) + \gamma'_{2,1'} \frac{\chi(t)}{\delta^2} e^{-i\tilde{\omega}t} \int_0^t a_{\text{in}}(t') \chi^*(t') dt' + \sqrt{\gamma'_{2,1'}} \frac{\chi(t)}{\delta} e^{-i\tilde{\omega}t} \sigma_{1'1}(0),$$
(B8)

where the continuous free-field operator is defined by

$$a_0(t) = \sqrt{\frac{c}{L}} \sum_{\omega} a_{\omega} e^{-i\omega t} \to \frac{1}{\sqrt{2\pi}} \int d\omega \, a(\omega) e^{-i\omega t}.$$
 (B9)

Given the fact that the initial state of the field is the singlephoton state

$$|1_f\rangle = a_f^{\dagger}|0\rangle, \tag{B10}$$

where

$$a_f^{\dagger} = \int dt f(t) e^{-i\bar{\omega}t} a_0^{\dagger}(t), \qquad (B11)$$

and that

$$a_0(t)|1_f\rangle = f(t)e^{-i\bar{\omega}t}|0\rangle, \tag{B12}$$

it is straightforward to show that Eq. (B8) leads to the correct retarded field intensity given in Eq. (54) (recall that *t* is the retarded time). Equation (B8) differs from the input-output form given in Eq. (B2); that is, the first term of output field operator $a_{out}(t)$ depends in a nonlocal way on the input field operator $a_{in}(t)$.

2. State vector approach

In some theories of down-conversion, one also finds a type of hybrid Schrödinger-Heisenberg single-mode approach using a state vector theory in lowest-order perturbation theory. To mirror the down conversion calculations, we must consider two limits, overlapping and nonoverlapping pulses.

When the probe and pump field pulses overlap, the effective Hamiltonian for the Raman problem can be taken as

$$H(t) = \hbar G(t)(a_1^{\dagger}b^{\dagger} + a_1b),$$
 (B13)

where a_1^{\dagger} is a creation operator for the output field mode, b^{\dagger} acting on the atomic ket $|1\rangle$ converts it to $|1'\rangle$, and

$$G(t) = \left|\frac{\eta \chi_0 f(t)}{\delta}\right|^2 \tag{B14}$$

accounts for the atom-field coupling. To lowest order, the state vector is then given by

$$|\psi(T)\rangle \approx |\psi(0)\rangle - i \int_0^T dt G(t) a_1^{\dagger} b^{\dagger} |\psi(0)\rangle.$$
 (B15)

If we take $|\psi(0)\rangle = a_1^{\dagger}|0\rangle$, then

$$\begin{split} |\psi(T)\rangle &\approx a_{1}^{\dagger}|0;1\rangle - i\int_{0}^{T}dtG(t)a_{1}^{\dagger}a_{1}^{\dagger}b^{\dagger}|0;1\rangle \\ &= |1_{1};0\rangle - i\sqrt{2}R|2_{1},1'\rangle, \end{split} \tag{B16}$$

where $|n_F; n_A\rangle$ is the eigenket of an *n* photon state in mode 1 and the atom in state n_A . If $R^2 \ll 1$, then

$$W_N = \langle \psi(T) | a_1^{\dagger} a_1 | \psi(T) \rangle = 1 + 2R^2; \qquad (B17a)$$

$$g^{(2)} \approx \langle \psi(T) | a_1^{\dagger} a_1^{\dagger} a_1 a_1 | \psi(T) \rangle = 4R^2, \qquad (B17b)$$

just as in the operator approach.

However, if the probe and pump pulses do not overlap, then the initial state vector is $|\psi(0)\rangle = a_2^{\dagger}|0\rangle$ (the probe pulse is in PHYSICAL REVIEW A 101, 033824 (2020)

a mode distinct from the one produced by the atom-vacuum field interaction), leading to

$$\begin{aligned} |\psi(T)\rangle &\approx a_{2}^{\dagger}|0;1\rangle - i\int_{0}^{T}dt G(t)a_{2}^{\dagger}a_{1}^{\dagger}b^{\dagger}|0;1\rangle \\ &= |1_{2};0\rangle - iR|1_{1},1_{2};1'\rangle, \end{aligned} \tag{B18}$$

$$W'_N = \langle \psi(T) | (a_1^{\dagger} + a_2^{\dagger}) (a_1 + a_2) | \psi(T) \rangle = 1 + R^2,$$
 (B19)

and

$$g^{(2)\prime} \sim \frac{\langle \psi(T) | (a_1^{\dagger} + a_2^{\dagger}) (a_1^{\dagger} + a_2^{\dagger}) (a_2 + a_1) (a_2 + a_1) | \psi(T) \rangle}{\langle \psi(T) | (a_1^{\dagger} + a_2^{\dagger}) (a_2 + a_1) | \psi(T) \rangle^2} \approx 2R^2.$$
(B20)

The factor of $\sqrt{2}$ in Eq. (B16), resulting from stimulated emission can then be viewed as the origin of the increase in coincidence counts. This is the type of reasoning used in the explanation of the down conversion experiments [9], but there seems to be no formal justification for this "single-mode"– type approach. Although this approach gives the correct answer in the limits of overlapping and nonoverlapping pulses, it is no substitute for the more rigorous calculation given in Appendix A that fully accounts for retardation. Moreover, as we have already seen, a state vector approach carried out in lowest order leads to an incorrect result for the field intensity.

[1] A. Einstein, Phys. Z. 18, 121 (1917).

- [2] See, for example, B. E. A. Saleh and M. C. Teich, *Fundamentals of Photonics*, 2nd ed. (John Wiley and Sons, New York, 2013), pp. 501–510.
- [3] E. T. Jaynes and F. W. Cummings, Proc. IEEE 51, 89 (1963).
- [4] See, for example, D. Valente, Y. Li, J. P. Poizat, J. M. Gérard, L. C. Kwek, M. F. Santos, and A. Auffèves, New J. Phys. 14, 083029 (2012); K. A. Fischer, OSA Continuum 1, 772 (2018); A. H. Kiilerich and K. Molmer, Phys. Rev. Lett. 123, 123604 (2019).
- [5] J. Lampen, A. Duspayev, H. Nguyen, H. Tamura, P. R. Berman, and A. Kuzmich, Phys. Rev. Lett. **123**, 203603 (2019).
- [6] R. Hanbury Brown and R. Q. Twiss, Nature 177, 27 (1956).
- [7] C. K. Hong and Z. Y. Ou, and and L. Mandel. Phys. Rev Lett. 59, 2044 (1987).
- [8] See, for example, H. Fearn and R. Louden, J. Opt. Soc. Amer. B 6, 917 (1988); L. Mandel and E. Wolf, *Optical Coherence* and Quantum Optics (Cambridge University Press, New York, 1995), Chaps. 14 and 22; R. Loudon, *The Quantum Theory of Light*, 3rd ed. (Oxford University Press, New York, 2000), Sec. 6.8; T. Legero, T. Wilk, A. Kuhn, and G. Rempe, in Advances

in Atomic, Molecular, and Optical Physics, edited by P. R. Berman, C. C. Lin, and E. Arimondo (Academic Press, New York, 2006), pp. 253–289; Z. Y. J. Ou, *Muti-Photon Quantum Interference* (Springer, New York, 2007), Part 1, and references therein.

- [9] See, for example, A. Lamas-Linares, C. Simon, J. C. Howell, and D. Bouwmeester, Science 296, 712 (2002); F. W. Sun, B. H. Liu, Y. X. Gong, Y. F. Huang, Z. Y. Ou, and G. C. Guo, Phys. Rev. Lett. 99, 043601 (2007); S. Dong, X. Yao, W. Zhang, S. Chen, W. Zhang, L. You, Z. Wang, and Y. F. Huang, ACS Photonics 4, 746 (2017).
- [10] See, for example, P. R. Berman and V. S. Malinovsky, *Principles of Laser Spectroscopy and Quantum Optics* (Princeton University Press, Princeton, NJ, 2011), Secs. 19.3 and 19.4.
- [11] Z. Y. J. Ou, *Muti-Photon Quantum Interference* (Springer, New York, 2007), Sec. 7.2.2.
- [12] T. Chanelière, D. N. Matsukevich, S. D. Jenkins, T. A. B. Kennedy, M. S. Chapman, and A. Kuzmich, Phys. Rev. Lett. 96, 093604 (2006).
- [13] R. Loudon, *The Quantum Theory of Light, Third Edition* (Oxford University Press, New York, 2000), Chap. 6.