Quantum logic operations based on photon-exchange interactions

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Nonlinear interactions between two photons are required for the construction of optical quantum logic gates, but those interactions are normally very weak due to the small magnitude of the electric field associated with a single photon. We note that exchange interactions can have a large effect even when there is no physical interaction between two particles, and we exploit this property for the construction of optical quantum logic gates. We show that the probability of there being two virtually-excited atoms in a medium can be a factor of 2 larger when two nonresonant photons propagate through the same medium as compared to the case in which they propagate through two separate media, in analogy with photon bunching. As a result, the application of one or more laser pulses will produce a nonlinear phase shift that can be used to construct an XOR quantum logic gate. This provides an example of a quantum control process in which one photon can control the state of another photon even when there is no sequence of physical interactions linking the two photons. From a classical point of view, it is not possible to identify a path for the flow of information or a specific cause for the outcome of the control process. [S1050-2947(99)03508-8]

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I. INTRODUCTION

Nonlinear optical effects normally require high-intensity beams of light containing many photons. Roughly speaking, this is because the electric field associated with a single photon is very weak, which causes the physical interaction of a single photon with another particle to be correspondingly small. One way to avoid this difficulty is to confine two photons to a small cavity with a high *Q*-factor, which increases both the magnitude of the electric field and the interaction time. Although nonlinear phase shifts at the two-photon level have been demonstrated in this way [1], the complexity of the required high-*Q* cavities and atomic traps may limit the practical value of these techniques in the construction of full-scale quantum computers [2,3] containing large numbers of qubits.

Even when there is no physical interaction between two identical particles, the requirement that the wave function be symmetric or antisymmetric under their exchange can produce an apparent tendency of the particles to either repel or attract each other, the simplest example being that of photon bunching [4,5]. There can be no real attraction or repulsion in such a case, since there is no actual force between the two particles, but in many respects the net effect is much the same as if there were. Exchange interactions have a major impact in many systems, where relatively strong physical forces would be required to produce any equivalent effect, such as is the case in a neutron star.

The relatively large magnitude of exchange interactions suggests that it may be possible to construct optical quantum logic gates in such a way that the required nonlinear interaction is derived from exchange interactions rather than the relatively weak physical interactions of the photons. As we will show below, the probability of there being two virtually-excited atoms in a medium can be a factor of 2 larger when two off-resonant photons propagate through the same medium as compared to the case in which they propagate through two separate media, in analogy with photon bunching. This difference in the population of the excited atomic states can be exploited by applying a sequence of laser pulses to produce phase shifts in the excited states of the atoms. Since the effects of the laser pulses are dependent on the population of the excited states, a different phase shift is obtained when the two photons are in the same medium than is obtained when they propagate through two separate media, which corresponds to a nonlinear effect. We have also considered several other approaches that appear to be less effective, as described in one of the appendices, including an earlier suggestion that relied on collisions with a buffer gas rather than laser pulses [6].

The difficulty in obtaining nonlinear interactions at the two-photon level by conventional means [7-9] can be seen by considering the probability that an off-resonant photon passing through a medium, such as an atomic vapor cell, will interact with an atom in the medium. This probability can be made to be on the order of unity by simply increasing the number of atoms in the medium, but the probability that two photons will interact with the *same* atom in such a medium will typically be very small. For example, if the medium contains 10^{10} atoms and the total probability of an interaction is on the order of unity, then the probability that two photons will interact with the same atom will be on the order of 10^{-10}. Any nonlinear optical process that requires both photons to interact with the same atom might therefore be expected to be negligibly small. In contrast, the exchange interaction of interest here involves pairs of atoms and does not require both photons to interact with the same atom.

As an example, a conventional process for the production of nonlinear phase shifts (Kerr effect) is illustrated in Fig. 1. Here two photons with frequencies ω_1 and ω_2 interact with a three-level atom. The frequency of photon 2 is relatively close to the transition frequency between atomic levels 2 and 3, so that virtual transitions in which photon 2 is absorbed and then reemitted would produce a phase shift for photons of that frequency. Virtual transitions of that kind can only occur if photon 1 has previously been absorbed by the atom,

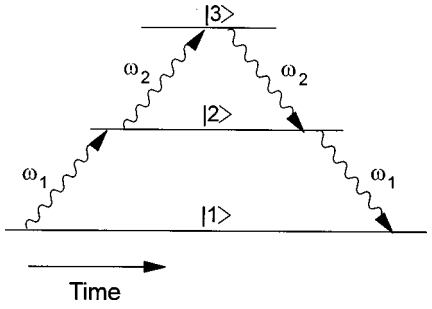

FIG. 1. A conventional mechanism for the production of nonlinear phase shifts (Kerr effect), in which virtual transitions between atomic levels $|2\rangle$ and $|3\rangle$ will produce a phase shift for photons of frequency ω_2, provided that a photon at frequency ω_1 is present to produce a virtual transition from level $|1\rangle$ to level $|2\rangle$. Mechanisms of this kind require both photons to interact with the same atom, which is unlikely to occur at single-photon levels.

since the atom would otherwise be in its ground state and not in level 2 at room temperature. The net result is that the presence or absence of photon 1 can control the phase shift experienced by photon 2. This requires photons 1 and 2 to interact with the same atom, which is very unlikely at single-photon intensities, and conventional mechanisms of this kind are usually not significant at single-photon intensities as a result. To the best of our knowledge, all previous mechanisms [1,7-12] for the production of nonlinear phase shifts at low intensities require two photons to interact with the same atom. Classical arguments based on the flow of information suggest that this must always be the case, as will be discussed in more detail below.

We are concerned, instead, with processes in which two photons interact with two different atoms in a medium [6], such as those labeled *A* and *B* in Fig. 2. In this virtual process, atom *A* absorbs photon 1 and reemits photon 2, while atom *B* absorbs photon 2 and reemits photon 1. This exchange of the two photons will produce an energy shift that can be calculated using perturbation theory, for example, which in turn will produce a shift in the overall phase of the system. If the number N of atoms in the medium is sufficiently high that each photon has a probability on the order of unity of being absorbed by an atom, then the probability amplitude for a process of this kind would also be expected to be on the order of unity. Since the number of pairs of atoms is proportional to N^2, the expected nonlinear phase shift should also be proportional to N^2 in the weak coupling

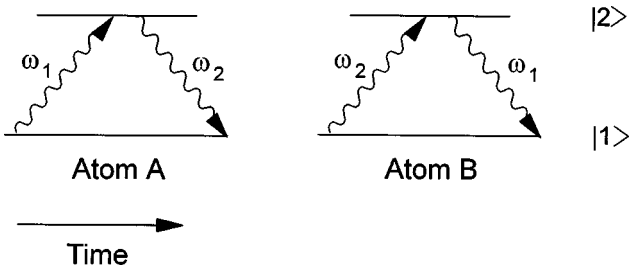

FIG. 2. An exchange interaction in which atom *A* absorbs photon 1 and reemits photon 2, while atom *B* absorbs photon 2 and reemits photon 1. A mechanism of this kind is expected to be relatively strong at single-photon intensities because the two photons are not required to interact with the same atom.

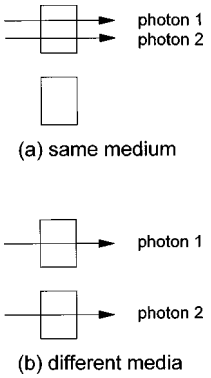

FIG. 3. (a) Two photons passing through an optical medium, such as an atomic vapor cell. (b) The same two photons passing through two separate media. The nonlinear phase shift is equal to the difference in the phases in the two cases, which can be strongly affected by exchange interactions.

limit, whereas conventional mechanisms in which both photons interact with the same atom give a nonlinear phase shift proportional to N. We previously showed [6], however, that the contributions from all Feynman diagrams of this kind cancel out and give no net effect unless the system is perturbed in some way, such as by collisions with a buffer gas. More detailed calculations have subsequently shown that the use of a buffer gas for this purpose is strongly dependent on the nature of the collision process, as is discussed in more detail in Appendix B, and that the random nature of the collisions would also introduce undesirable phase noise.

The difficulties associated with the use of collisions can be avoided by using laser pulses to perturb the excited states of the atoms, and the remainder of this paper will concentrate on that approach. Perhaps the simplest way to understand this mechanism is to consider the probability P_2 that two atoms will be in virtually-excited states at the same time when two non-resonant photons pass through the same medium, as illustrated in Fig. 3(a). We show below that P_2 is a factor of 2 larger when both photons pass through the same medium than is the case when the two photons pass through two separate but otherwise identical media, as illustrated in Fig. 3(b). This increased probability is due to the fact that atom *A* may have been excited by photon 1 while atom *B* was excited by photon 2, or atom A may have been excited by photon 2 while atom *B* was excited by photon 1, as illustrated in Fig. 4. The probability amplitudes for these two processes will constructively interfere provided that

$$\delta\mathbf{k}\cdot\delta\mathbf{r}\ll\frac{\pi}{2}, \tag{1}$$

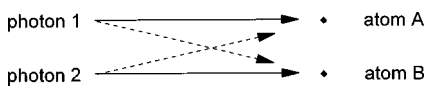

FIG. 4. A virtual state in which atoms *A* and *B* are both excited may have been produced in two ways: Photon 1 may have excited atom *A* while photon 2 excited atom *B*, or photon 1 may have excited atom *B* while photon 2 excited atom *A*. Constructive interference between these two probability amplitudes can produce a factor of 2 enhancement in the probability of there being two excited atoms, in analogy with the Hanbury-Brown and Twiss experiment.

where $\delta\mathbf{k}$ is the difference in the k vectors of the two photons and $\delta\mathbf{r}$ is the difference in the positions of the two atoms. This is the same condition that is required for the observation of the Hanbury-Brown and Twiss effect (photon bunching) and Fig. 4 is analogous to that effect if atoms A and B are viewed as two ''detectors'' placed in front of a well-collimated source.

The factor of 2 difference in P_2 can be exploited by applying a laser pulse to produce a phase shift in the excited states of the atoms in the medium. As mentioned above, the effects of the laser pulse will depend on the population of the excited atomic states and a different phase shift will therefore be produced when the two photons propagate together through the same medium than when they propagate through two different media. This corresponds to a nonlinear phase shift whose origin ultimately derives from the exchange interaction of Fig. 4. A suitable sequence of such laser pulses can give a nonlinear phase shift of π, which can then be used in an interferometer arrangement [13] to produce a Controlled-NOT (XOR) quantum logic gate.

We begin by defining the system of interest and describing the corresponding state vector and Hamiltonian. By neglecting scattering and dispersion, which are both small for large photon detunings, and by making the adiabatic approximation, the quantum state of the entire system can be described by a set of six complex numbers that are taken to be the elements of an effective six-dimensional state vector. The propagation of the photons through the medium and their interaction with the laser pulses can then be determined by solving a six-dimensional eigenvalue problem. After considering the simplest case of a single laser pulse, the choice of an optimal sequence of laser pulses is described. We conclude with a discussion of the nonclassical nature of these results and their inconsistency with classical concepts such as the flow of information and determinism. Appendix A considers the limitations imposed by symmetry considerations while Appendix B discusses a number of less effective approaches, including the use of collisions with a buffer gas or Berry's geometric phase [14]. Appendix C presents a simplified derivation for the case of equal detunings.

II. STATE VECTOR AND HAMILTONIAN

The optical medium will be assumed to be an atomic vapor cell for simplicity, although the basic results should apply equally well to solid-state materials. Equation (1) can be satisfied for a medium with a moderate thickness L if the difference in frequencies of the two incident photons is much less than their average frequency and they propagate in the same direction. For example, $\omega_1-\omega_2$ may be on the order of a few GHz in a typical experiment, which would allow the thickness of the vapor cell to be on the order of 1 cm. In order to minimize reflections from the surface of the medium, it will be assumed that the density of atoms in the medium is slowly varying compared to the wavelength of the photons, as illustrated in Fig. 5(a). The total number N of atoms in the medium will be assumed to be large ($\sim 10^{10}$).

The thickness of the atomic medium can be substantially increased while still satisfying Eq. (1) by using a periodic density of atoms as illustrated in Fig. 5(b), where it is assumed that the photons are propagating in the z direction.

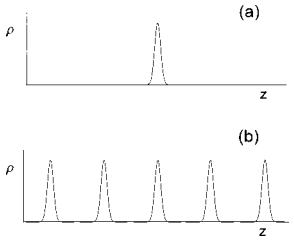

FIG. 5. (a) An atomic medium whose density ρ is a slowly varying function of position z and sufficiently thin that $\delta k\,\delta z \ll \pi/2$, where δz is the thickness. (b) A periodic medium satisfying the condition $\delta k \Delta z = 2p\pi$, where Δz is the periodicity and p is an integer. In either case, there is constructive interference between the probability amplitudes of Fig. 4.

Here the thin structure of Fig. 5(a) is repeated at intervals Δz for which $\Delta z\,\delta k = 2p\pi$, where p is an integer. This approach is somewhat similar to the commonly-used technique of quasiphase matching [15] and would allow moderate values of L even for relatively large differences in the frequencies of the two photons. For simplicity, we will assume the geometry of Fig. 5(a) throughout this paper, but the results can be readily extended to the periodic case.

The effects of interest involve two-level atoms, as in Fig. 2, but it will be necessary to vary the energy of the upper atomic levels in a time-dependent manner by applying external fields to the system. This could be done in a variety of ways but, to be specific, we will assume that a laser beam is used to couple the second atomic level to a third atomic state that is of no other interest, as illustrated in Fig. 6. Here photons 1 and 2 are slightly off resonance with the atomic transitions between levels 1 and 2, while the laser beam is sufficiently far from resonance with the atomic transition between levels 2 and 3 that no significant population transfer into level 3 occurs. In that case, the net effect of the laser beam is to shift the energy of level 2 by an amount that can be calculated using perturbation theory or other methods (ac Stark shift). The lack of population in level 3 allows us to use a two-level model for the atoms in which the energy e_A of level 2 is a function of time.

The incident photons are assumed to propagate along the z direction and are represented by multimode Fock states (not merely weak coherent states) corresponding to Gaussian wave packets. The temporal width τ_p of the wave packets is assumed to be much longer than the transit time L/c through the medium, so that the magnitude of the electric field of the

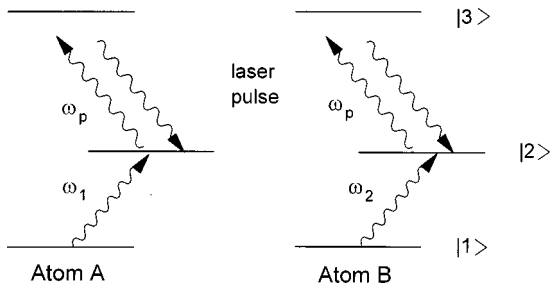

FIG. 6. The application of a laser pulse that is detuned from the transition between levels 2 and 3, which can be used to produce a Stark shift in the energy of level 2 and a corresponding phase shift in that state.

photons is essentially uniform throughout the medium. The effects of interest require a multimode analysis, since the nonlinear phase shift depends on the expectation value of the product of the intensities of the two photons, which would vanish for plane-wave single photons in free space. The incident photons can be represented by two single-photon creation operators, $a_1^\dagger$ and $a_2^\dagger$, defined by

$$a_1^\dagger = \sum_{\mathbf{k}} f_1(\mathbf{k}) a_{\mathbf{k}}^\dagger,$$
$$a_2^\dagger = \sum_{\mathbf{k}} f_2(\mathbf{k}) a_{\mathbf{k}}^\dagger. \tag{2}$$

Here the operator $a_{\mathbf{k}}^\dagger$ creates a plane-wave photon [16] with wave vector $\mathbf{k}$, and $f_1(\mathbf{k})$ and $f_2(\mathbf{k})$ are the Fourier coefficients of the Gaussian wave packets at the initial time t_0. These coefficients are chosen in such a way that the inverse Fourier transforms are given by

$$G_1(z) \equiv \frac{1}{\sqrt{2\pi}} \int e^{i\mathbf{k}\cdot\mathbf{r}} f_1(\mathbf{k}) d^3\mathbf{k} = g e^{i\overline{k_1} z} e^{-(z-z_0)^2/2c^2\tau_p^2},$$
$$G_2(z) \equiv \frac{1}{\sqrt{2\pi}} \int e^{i\mathbf{k}\cdot\mathbf{r}} f_2(\mathbf{k}) d^3\mathbf{k} = g e^{i\overline{k_2} z} e^{-(z-z_0)^2/2c^2\tau_p^2}, \tag{3}$$

where g is a constant and z_0 is the initial location of the center of the wave packets, which is taken to be far from the location of the atoms so that there is initially no interaction. Both wave packets have the same amplitude and width but different values for their central k vectors, $\overline{k_1}$ and $\overline{k_2}$, which are related to the central frequencies of their Fourier spectra by $\overline{\omega_1} = c\overline{k_1}$ and $\overline{\omega_2} = c\overline{k_2}$. Both the medium and the photon wave packets are assumed to have no significant spatial variation in the transverse direction, so that the right-hand side of Eq. (3) involves only the z coordinate. The main results of this paper depend only on the assumption that the modulus of $G_1(z)$ and $G_2(z)$ is a slowly varying function of z and the exact shape of the wave packets is not essential.

The initial state of the field is then given by

$$|\gamma_1, \gamma_2\rangle = a_1^\dagger a_2^\dagger |0\rangle, \tag{4}$$

where $|0\rangle$ is the vacuum. We will also consider the single-photon states defined by

$$|\gamma_1\rangle = a_1^\dagger |0\rangle,$$
$$|\gamma_2\rangle = a_2^\dagger |0\rangle, \tag{5}$$

as well as the following states containing two identical photons:

$$|\gamma_1, \gamma_1\rangle = \frac{1}{\sqrt{2}} (a_1^\dagger)^2 |0\rangle,$$
$$|\gamma_2, \gamma_2\rangle = \frac{1}{\sqrt{2}} (a_2^\dagger)^2 |0\rangle. \tag{6}$$

All of the atoms are assumed to be in their ground state initially, so that the quantum state of the system is initially given by

$$|\Psi(t_0)\rangle = |\gamma_1, \gamma_2\rangle \prod_i |\psi_{1i}\rangle, \tag{7}$$

where $|\psi_{1i}\rangle$ represents atom i in its ground state.

It will be convenient to write the Hamiltonian H as the sum of two parts:

$$H = H_0 + H_{\text{int}}. \tag{8}$$

H_0 represents the energies of the field and the atoms in the absence of any interaction and is given as usual by

$$H_0 = \sum_{\mathbf{k}} (a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + 1/2) \hbar \omega_{\mathbf{k}} + \sum_i 1/2 e_A \sigma_{zi}, \tag{9}$$

where the atoms are labeled with index i, e_A is the energy of the excited state (level 2) of an atom above its ground state, and σ_{zi} is one of the Pauli spin matrices in a two-dimensional Hilbert space consisting of the ground and excited states of atom i. (This does not imply any spin interactions.) The interaction Hamiltonian H_{int} in the Coulomb gauge and in the standard dipole approximation [16] is given by

$$H_{\text{int}} = -q \sum_i \mathbf{r}_i \cdot \mathbf{E}(\mathbf{R}_i). \tag{10}$$

Here q is the charge of the electron and $\mathbf{r}_i$ is the relative coordinate of the electron in atom i, where we are assuming hydrogenlike atomic states. $\mathbf{E}(\mathbf{R}_i)$ is the second-quantized electric field operator [16,17] at the location $\mathbf{R}_i$ of the center of mass of atom i, which is given in the Schrödinger picture [5] and MKSA units by

$$E(\mathbf{R}) = i \sum_{\mathbf{k},j} \left(\frac{\hbar c k}{2\varepsilon_0 V} \right)^{1/2} [\boldsymbol{\lambda}_j e^{i\mathbf{k}\cdot\mathbf{R}} a_{\mathbf{k}} - \boldsymbol{\lambda}_j^* e^{-i\mathbf{k}\cdot\mathbf{R}} a_{\mathbf{k}}^\dagger]. \tag{11}$$

Here ε_0 is the permittivity of free space, V is the volume used for periodic boundary conditions, and λ_j represents the two orthogonal polarization states of a photon. Except for the discussion regarding symmetry considerations in Appendix A, both photons will be assumed to have the same state of polarization and the polarization indices will be dropped, since two photons of orthogonal polarizations cannot undergo an exchange interaction as shown in Figs. 2 and 4 by means of dipole transitions.

III. SCHRÖDINGER'S EQUATION AND THE CHOICE OF BASIS VECTORS

The photon wave packets are not eigenstates of H_0 and they will propagate at the speed of light in the absence of any interactions. As a result, it is much more convenient to work in the interaction picture [5], where the photon state vectors remain constant in the absence of any interaction and the electric field operator becomes time dependent. Schrödinger's equation then involves only the interaction Hamiltonian $H'(t)$:

$$i\hbar \frac{d|\Psi\rangle}{dt} = H'(t)|\Psi\rangle, \tag{12}$$

where $H'(t) = \exp[iH_0(t-t_0)/\hbar] H_{\rm int} \exp[-iH_0(t-t_0)/\hbar]$ as usual. $H'(t)$ will be found to be a slowly-varying function of time after a suitable unitary transformation, which will allow the adiabatic approximation [18] to be used to reduce the solution of the Schrödinger equation to an eigenvalue problem. The eigenvectors can be computed numerically or analytically, but in either case we will need the matrix elements of $H'(t)$ in a suitable basis.

The postulates of quantum mechanics allow us to choose any set of orthonormal basis vectors in Hilbert space (Fock space for the photons). The nonlinear phase shifts of interest correspond to a coherent process in which the photons propagate out of the medium in the same state in which they entered it, aside from an overall phase factor. As a result, it will be convenient to choose a set of basis vectors in Fock space that includes the original state $|\gamma_1,\gamma_2\rangle$ as well as the states $|\gamma_1\rangle$ and $|\gamma_2\rangle$ that can arise as a result of virtual absorption of the photons while they are in the medium. Since we need a complete set of orthonormal basis vectors, we define a set of modified plane-wave creation operators $b_{\mathbf{k}}^\dagger$ that are constructed in such a way that they generate states that are orthogonal to those generated by $a_1^\dagger$ and $a_2^\dagger$:

$$b_k^\dagger = c_n[a_{\mathbf{k}}^\dagger - f_1^*(\mathbf{k})a_1^\dagger - f_2^*(\mathbf{k})a_2^\dagger]. \tag{13}$$

Here c_n is a normalization constant and the last two terms provide the desired orthogonality. Both $[a_1,a_2^\dagger]$ and $\langle\gamma_1|\gamma_2\rangle$ become exponentially small when $|\omega_1-\omega_2|$ is much larger than the frequency spread of the Gaussian wave packets, which will be assumed to be the case, and the set of operators $a_1^\dagger$, $a_2^\dagger$, and $b_{\mathbf{k}}^\dagger$ obey the usual commutation relations [16,19] in that limit. This allows us to choose a set of basis vectors that consists of all of the states that are generated by $a_1^\dagger$, $a_2^\dagger$, and the $b_{\mathbf{k}}^\dagger$ at the initial time t_0. By definition, these basis vectors are independent of time in the interaction picture whereas, in the Schrödinger picture, they correspond to freely-propagating wave packets that do not include the effects of interactions.

The relevant matrix elements can now be calculated in this basis. For example, we will need the matrix elements $\mathcal{E}_1(\mathbf{R}_i,t)$ and $\mathcal{E}_2(\mathbf{R}_i,t)$ of the electric field operator defined by

$$\mathcal{E}_1(\mathbf{R}_i,t) = \langle 0|E(\mathbf{R}_i,t)|\gamma_1\rangle,$$

$$\mathcal{E}_2(\mathbf{R}_i,t) = \langle 0|E(\mathbf{R}_i,t)|\gamma_2\rangle. \tag{14}$$

Making use of Eqs. (2), (5), and (11) allows $\mathcal{E}_1(\mathbf{R}_i,t)$ to be written as

$$\mathcal{E}_1(\mathbf{R}_i,t) = i\left(\frac{\hbar c\bar{k}_1}{2\varepsilon_0 V}\right)^{1/2} \langle 0|e^{iH_0(t-t_0)/\hbar} \times \sum_{\mathbf{k}} [e^{i\mathbf{k}\cdot\mathbf{R}_i}a_{\mathbf{k}} - e^{-i\mathbf{k}\cdot\mathbf{R}_i}a_{\mathbf{k}}^\dagger] e^{-iH_0(t-t_0)/\hbar} \times \sum_{\mathbf{p}} f_1(\mathbf{p})a_{\mathbf{p}}^\dagger|0\rangle. \tag{15}$$

Here we have approximated the term $\hbar ck$ as a constant over the narrow bandwidth of the photons, which allows it to be taken outside of the sum. The commutation relations eliminate all terms except those where $\mathbf{k}=\mathbf{p}$, the remaining sum can be expressed as an integral that is proportional to the density $\rho(\bar{k}_1)$ of photon states, and $H_0/\hbar$ reduces to $\omega_1 = ck_1$ when acting on the eigenstates to the right. Equation (15) then reduces to

$$\mathcal{E}_1(\mathbf{R}_i,t) = i\rho(\bar{k}_1)\left(\frac{\hbar\bar{\omega}_1}{2\varepsilon_0 V}\right)^{1/2} \int e^{i[\mathbf{k}\cdot\mathbf{R}_i - ck(t-t_0)]} f_1(\mathbf{k})d^3\mathbf{k}. \tag{16}$$

Comparison with Eq. (3) shows that, aside from a constant, this expression is equal to the Gaussian function $G_1(Z_i - c(t-t_0))$. For simplicity, we take the center of the medium to be at $z=0$ and evaluate the matrix elements there, since the medium has been assumed to be sufficiently thin that the modulus of the field is essentially uniform over that distance [20]. We also choose z_0 and t_0 in such a way that the photon wave packets are centered on the medium at time $t=0$. In that case, these matrix elements reduce to

$$\mathcal{E}_1(t) = g' e^{-i\bar{\omega}_1 t} e^{-t^2/2\tau_p^2} \equiv \mathcal{E}_0(t) e^{-i\bar{\omega}_1 t},$$
$$\mathcal{E}_2(t) = g' e^{-i\bar{\omega}_2 t} e^{-t^2/2\tau_p^2} \equiv \mathcal{E}_0(t) e^{-i\bar{\omega}_2 t}, \tag{17}$$

where g' is a constant and $\mathcal{E}_0(t)$ is a real function that corresponds to the slowly varying envelope of the Gaussian wave packet at the location of the atoms. It should be noted that the frequency spread of the wave packets is still reflected in the time dependence of $\mathcal{E}_0(t)$ but does not appear in the exponential phase factor, which will be important in considering the effects of subsequent unitary transformations.

The matrix elements involving the modified plane-wave states can be evaluated in the same way, with the result that

$$|\langle 0|E(0,t)|\mathbf{k}\rangle| \ll |\langle 0|E(0,t)|\gamma_1\rangle| \tag{18}$$

for times at which the wave packets overlap the location of the medium. Here $|\mathbf{k}\rangle = b_k^\dagger|0\rangle$ is one of the modified plane-wave basis states. This reflects the fact that the fields of photon 1 and photon 2 are concentrated in that region, whereas the plane-wave states are not localized. We therefore make the approximation that the matrix elements in-

volving the plane-wave states can be neglected and that there is no coupling into those modes. This corresponds to the neglect of scattering and dispersion, which are expected to be small in the limit of large detunings, and it also neglects small radiative corrections such as the Lamb shift [19]. The validity of this approximation when using a sequence of laser pulses will be discussed in Sec. VI.

The photon detunings, defined as $\delta_1=\hbar\overline{\omega_1}-e_A$ and $\delta_2=\hbar\overline{\omega_2}-e_A$, will be assumed to be much smaller in magnitude than $\overline{\omega_1}$ or $\overline{\omega_2}$. The rotating wave approximation (energy conservation) then ensures that the absorption of a photon must be accompanied by the excitation of an atom [17]. Having neglected any coupling into the plane-wave states, the only states of the field that can occur when both photons propagate in the same medium, as in Fig. 3(a), are linear combinations of the states $|\gamma_1,\gamma_2\rangle$, $|\gamma_1\rangle$, $|\gamma_2\rangle$, $|0\rangle$, $|\gamma_1,\gamma_1\rangle$, and $|\gamma_2,\gamma_2\rangle$. These six basis vectors span the occupied region of Fock space for this system and the state of the field can be specified by its probability amplitudes in this basis, which form the elements of a six-dimensional state vector.

If the photons are linearly polarized along the x direction, for example, then the atomic part of the relevant matrix element is given by

$$\begin{aligned}\langle\varphi_{2i}|H'(t)|\varphi_{1i}\rangle&=-q\langle\varphi_{2i}|e^{iH_0(t-t_0)/\hbar}x_i e^{-iH_0(t-t_0)/\hbar}|\varphi_{1i}\rangle\\&=d_0e^{ie_A(t-t_0)/\hbar},\end{aligned}\tag{19}$$

where d_0 is the magnitude of the dipole moment between these two states. For the case of degenerate hydrogenlike atomic states, $|\varphi_{2i}\rangle$ corresponds to the linear combination of states that is excited by the absorption of a linearly polarized photon, as discussed in more detail in Appendix A.

Since there are only two incident photons, there can be at most two excited atoms, which will be labeled i and j with $i>j$ to avoid counting the same state twice. The total number of atomic states is thus on the order of N^2. The basis vectors for the combined system of electromagnetic field and atoms consist of the tensor products of the various atomic states with the six field states described above. In that basis, we define $c(\gamma_1,\gamma_2)$ to be the probability amplitude to have both of the original photons and no excited atoms. The probability amplitude for atom i to be excited with photon 1 remaining will be denoted by $c(\gamma_1,i)$, while $c(\gamma_2,i)$ will denote the probability amplitude for atom i to be excited with photon 2 remaining. The probability amplitude for atoms i and j to be excited with no photons remaining will be denoted $c(0,i,j)$, with $i>j$. The probability amplitudes to have two identical photons and no excited atoms will be denoted by $c(\gamma_1,\gamma_1)$ and $c(\gamma_2,\gamma_2)$.

The time dependence of these probability amplitudes can be found from the Schrödinger equation, Eq. (12), and the corresponding matrix elements of $H'(t)$:

$$\begin{aligned}i\hbar\frac{d}{dt}c(\gamma_1,\gamma_2)&=\sum_i Me^{i\delta_2t/\hbar}c(\gamma_1,i)\\&\quad+\sum_i Me^{i\delta_1t/\hbar}c(\gamma_2,i),\end{aligned}$$

$$\begin{aligned}i\hbar\frac{d}{dt}c(\gamma_1,i)&=Me^{-i\delta_2t/\hbar}c(\gamma_1,\gamma_2)+\sum_{j<i}Me^{i\delta_1t/\hbar}c(0,i,j)\\&\quad+\sum_{j>i}Me^{i\delta_1t/\hbar}c(0,j,i)\\&\quad+\sqrt{2}Me^{-i\delta_1t/\hbar}c(\gamma_1,\gamma_1),\end{aligned}$$

$$\begin{aligned}i\hbar\frac{d}{dt}c(\gamma_2,i)&=Me^{-i\delta_1t/\hbar}c(\gamma_1,\gamma_2)+\sum_{j<i}Me^{i\delta_2t/\hbar}c(0,i,j)\\&\quad+\sum_{j>i}Me^{i\delta_2t/\hbar}c(0,j,i)\\&\quad+\sqrt{2}Me^{-i\delta_2t/\hbar}c(\gamma_2,\gamma_2),\end{aligned}$$

$$\begin{aligned}i\hbar\frac{d}{dt}c(0,i,j)&=Me^{-i\delta_1t/\hbar}c(\gamma_1,i)+Me^{-i\delta_2t/\hbar}c(\gamma_2,i)\\&\quad+Me^{-i\delta_1t/\hbar}c(\gamma_1,j)+Me^{-i\delta_2t/\hbar}c(\gamma_2,j),\end{aligned}\tag{20}$$

$$i\hbar\frac{d}{dt}c(\gamma_1,\gamma_1)=\sum_i\sqrt{2}Me^{i\delta_1t/\hbar}c(\gamma_1,i),$$

$$i\hbar\frac{d}{dt}c(\gamma_2,\gamma_2)=\sum_i\sqrt{2}Me^{i\delta_2t/\hbar}c(\gamma_2,i).$$

Here M is a brief notation for the basic matrix element defined by

$$M(t)=d_0\mathcal{E}_0(t)\tag{21}$$

and it will be a real number for an appropriate choice of the relative phase between the two atomic states. The factors of $\sqrt{2}$ that appear in these equations are due to stimulated emission into states containing two photons or absorption from those states.

Because the atoms are all subjected to the same field, the following probability amplitudes will all be equal:

$$\begin{aligned}c(\gamma_1,i)&=c(\gamma_1,i'),\\c(\gamma_2,i)&=c(\gamma_2,i'),\\c(0,i,j)&=c(0,i',j')\end{aligned}\tag{22}$$

for all values of i, i', j, and j'. This allows Eq. (20) to be simplified by introducing a new set of variables:

$$\begin{aligned}c(\gamma_1)&\equiv\sqrt{N}c(\gamma_1,i),\\c(\gamma_2)&\equiv\sqrt{N}c(\gamma_2,i),\\c(0)&\equiv\sqrt{N(N-1)/2}c(0,i,j).\end{aligned}\tag{23}$$

These new variables have been chosen in such a way that the squared modulus of $c(\gamma_1)$ gives the total probability that photon 1 is present and photon 2 has been absorbed, regard-

less of which atom is excited, with a similar role for $c(\gamma_2)$. The squared modulus of $c(0)$ similarly gives the total probability of there being two excited atoms and no photons. With this change of variables, Eq. (20) becomes

$$i\hbar\frac{d}{dt}c(\gamma_1,\gamma_2)=\sqrt{N}Me^{i\delta_2 t/\hbar}c(\gamma_1)+\sqrt{N}Me^{i\delta_1 t/\hbar}c(\gamma_2),$$

$$\begin{aligned}i\hbar\frac{d}{dt}c(\gamma_1)&=\sqrt{N}Me^{-i\delta_2 t/\hbar}c(\gamma_1,\gamma_2)\\&\quad+\sqrt{2(N-1)}Me^{i\delta_1 t/\hbar}c(0)\\&\quad+\sqrt{2N}Me^{-i\delta_1 t/\hbar}c(\gamma_1,\gamma_1),\\ i\hbar\frac{d}{dt}c(\gamma_2)&=\sqrt{N}Me^{-i\delta_1 t/\hbar}c(\gamma_1,\gamma_2)\\&\quad+\sqrt{2(N-1)}Me^{i\delta_2 t/\hbar}c(0)\\&\quad+\sqrt{2N}Me^{-i\delta_2 t/\hbar}c(\gamma_2,\gamma_2),\end{aligned}\tag{24}$$

$$\begin{aligned}i\hbar\frac{d}{dt}c(0)&=\sqrt{2(N-1)}Me^{-i\delta_1 t/\hbar}c(\gamma_1)\\&\quad+\sqrt{2(N-1)}Me^{-i\delta_2 t/\hbar}c(\gamma_2),\end{aligned}$$

$$i\hbar\frac{d}{dt}c(\gamma_1,\gamma_1)=\sqrt{2N}Me^{i\delta_1 t/\hbar}c(\gamma_1),$$

$$i\hbar\frac{d}{dt}c(\gamma_2,\gamma_2)=\sqrt{2N}Me^{i\delta_2 t/\hbar}c(\gamma_2),$$

which involves a total of six complex variables.

An inspection of Eq. (24) reveals that it is equivalent to Schrödinger's equation for a six-dimensional vector whose components are taken to be

$$|\psi\rangle_{\rm eff}\equiv\begin{pmatrix}c(\gamma_1,\gamma_2)\\c(\gamma_1)\\c(\gamma_2)\\c(0)\\c(\gamma_1,\gamma_1)\\c(\gamma_2,\gamma_2)\end{pmatrix},\tag{25}$$

provided that the Hamiltonian is chosen to be

$$H_{\rm eff}=M\begin{bmatrix}0&\sqrt{N}e^{i\delta_2 t}&\sqrt{N}e^{i\delta_1 t}&0&0&0\\ \sqrt{N}e^{-i\delta_2 t}&0&0&\sqrt{2(N-1)}e^{i\delta_1 t}&\sqrt{2N}e^{-i\delta_1 t}&0\\ \sqrt{N}e^{-i\delta_1 t}&0&0&\sqrt{2(N-1)}e^{i\delta_2 t}&0&\sqrt{2N}e^{-i\delta_2 t}\\ 0&\sqrt{2(N-1)}e^{-i\delta_1 t}&\sqrt{2(N-1)}e^{-i\delta_2 t}&0&0&0\\ 0&\sqrt{2N}e^{i\delta_1 t}&0&0&0&0\\ 0&0&\sqrt{2N}e^{i\delta_2 t}&0&0&0\end{bmatrix},\tag{26}$$

where $\hbar$ has been omitted to make the notation more compact. Since the six components of $|\psi\rangle_{\rm eff}$ completely determine the state of the system and their squared moduli give the total probabilities of the various photon states, we refer to $|\psi\rangle_{\rm eff}$ as the effective state vector for the system.

The physical meaning of the effective state vector can be understood by considering the state of the full system that corresponds to each of its elements. For example, the second component of $|\psi\rangle_{\rm eff}$ corresponds to the state

$$|\Psi(\gamma_1)\rangle=\frac{1}{\sqrt{N}}\sum_i|\gamma_1,i\rangle,\tag{27}$$

where each atom has an equal probability amplitude to be excited. Although there are $\sim N$ other linear combinations of the states $|\gamma_1,i\rangle$, the Hamiltonian couples the initial state only to the particular linear combination shown in Eq. (27) and all of the other linear combinations are unexcited or ''dark'' and can be ignored. Similar comments can be made with regard to the other components of $|\psi\rangle_{\rm eff}$, which correspond to the probability amplitudes of the only linear combinations of states that can evolve from the initial state under the action of the Hamiltonian. The Hamiltonian of Eq. (26) could be derived in a somewhat simpler way by arbitrarily defining these six state vectors and showing that the Hamiltonian does not couple them to any other states, after which the matrix elements in Eq. (26) could be written down by inspection.

The exponential factors in Equation (26) are rapidly varying functions of time. This time variation can be eliminated by making a unitary transformation given by

$$|\psi'\rangle_{\rm eff}=e^{-ih_0 t}|\psi\rangle_{\rm eff},\tag{28}$$

where the matrix h_0 is taken to be

$$h_0=\begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -\delta_2 & 0 & 0 & 0 & 0 \\ 0 & 0 & -\delta_1 & 0 & 0 & 0 \\ 0 & 0 & 0 & -(\delta_1+\delta_2) & 0 & 0 \\ 0 & 0 & 0 & 0 & (\delta_1-\delta_2) & 0 \\ 0 & 0 & 0 & 0 & 0 & (\delta_2-\delta_1) \end{bmatrix}. \tag{29}$$

After this transformation, the effective state vector obeys the equation

$$ih\frac{d}{dt}|\psi'\rangle_{\text{eff}}=H'_{\text{eff}}|\psi'\rangle_{\text{eff}}, \tag{30}$$

where the effective Hamiltonian now has the form

$$H'_{\text{eff}}=\begin{bmatrix} 0 & \sqrt{N}M & \sqrt{N}M & 0 & 0 & 0 \\ \sqrt{N}M & -\delta_2 & 0 & \sqrt{2(N-1)}M & \sqrt{2N}M & 0 \\ \sqrt{N}M & 0 & -\delta_1 & \sqrt{2(N-1)}M & 0 & \sqrt{2N}M \\ 0 & \sqrt{2(N-1)}M & \sqrt{2(N-1)}M & -(\delta_1+\delta_2) & 0 & 0 \\ 0 & \sqrt{2N}M & 0 & 0 & (\delta_1-\delta_2) & 0 \\ 0 & 0 & \sqrt{2N}M & 0 & 0 & (\delta_2-\delta_1) \end{bmatrix}. \tag{31}$$

Equations (30) and (31) determine the time evolution of the system for the case in which both photons propagate in the same medium, as in Fig. 3(a), and they will form the basis for most of the remaining analysis. For simplicity, the primes in Eqs. (30) and (31) will be omitted below.

For comparison purposes, we will also need to calculate the properties of the system when each photon propagates in a separate medium, as in Fig. 3(b). In this case, we have two independent systems whose time evolution can be calculated separately, after which the state vector for the overall system will be equal to the tensor product of the two individual state vectors. For the case in which only photon 1 is incident on a medium, an analysis similar to that presented above gives an effective state vector with the following components:

$$|\psi_1\rangle_{\text{eff}}\equiv\begin{pmatrix} c'(\gamma_1) \\ c'(0) \\ c'(\gamma_2) \end{pmatrix}. \tag{32}$$

Here $c'(\gamma_1)$ is the total probability amplitude that photon 1 remains with no excited atoms, $c'(0)$ represents the probability amplitude that the incident photon has been absorbed and there is one excited atom, and $c'(\gamma_2)$ represents the probability amplitude that photon 1 has been absorbed and another photon of frequency $\overline{\omega}_2$ has been reemitted. The effective Hamiltonian for this system is

$$H_{1\text{ eff}}=\begin{bmatrix} 0 & \sqrt{N}M & 0 \\ \sqrt{N}M & -\delta_1 & \sqrt{N}M \\ 0 & \sqrt{N}M & (\delta_2-\delta_1) \end{bmatrix}. \tag{33}$$

For the case in which only photon 2 is incident on a medium, the corresponding quantities are

$$|\psi_2\rangle_{\text{eff}}\equiv\begin{pmatrix} c''(\gamma_2) \\ c''(0) \\ c''(\gamma_1) \end{pmatrix}, \tag{34}$$

$$H_{2\text{ eff}}=\begin{bmatrix} 0 & \sqrt{N}M & 0 \\ \sqrt{N}M & -\delta_2 & \sqrt{N}M \\ 0 & \sqrt{N}M & (\delta_1-\delta_2) \end{bmatrix}. \tag{35}$$

IV. TWO-PHOTON DRESSED STATES

The photon wave packets have been assumed to be far from the medium at the initial time t_0 so that their interaction with the atoms will be exponentially small at that time. The initial state vector $|\psi_0\rangle$ is then given in the above basis by

$$|\psi_0\rangle=\begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}, \tag{36}$$

which is an eigenstate of $H_{\rm eff}$ with $M(t)=0$. It will be assumed that the Gaussian wave packets are sufficiently broad that $\mathcal{E}_0(t)$ is slowly varying on the time scale set by the diagonal terms of $H_{\rm eff}$. In that case, the adiabatic approximation [18] is valid and the state vector will slowly evolve into the corresponding instantaneous eigenstate of $H_{\rm eff}$. If no laser pulses are applied to the medium, the state vector will evolve back into $|\psi_0\rangle$ as the wave packets propagate away from the medium, since we have neglected scattering and dispersion.

We will primarily be interested in values of $\sqrt{N}M/\delta$ that are large enough to produce a perturbed state vector $|\psi(t)\rangle$ that is substantially different from $|\psi_0\rangle$ but not so large as to approach any level crossings, which would be nonadiabatic. Values of $\sqrt{N}M/\delta$ of this magnitude can be readily achieved in an atomic vapor cell under conditions in which the scattering and absorption are relatively small. This strongly perturbed eigenstate can be thought of as a two-photon dressed state [21].

Since the nonlinear interaction between two single photons is usually negligible, it might be expected that two photons would propagate independently through a medium and that the two-photon dressed state may be nothing more than the tensor product of two single-photon dressed states. That is not the case due to exchange interactions of the kind that are illustrated in Figs. 2 and 4. In particular, Fig. 4 suggests that there should be a factor of 2 increase in the probability of there being two virtually-excited atoms when two photons propagate in the same medium as compared to the case in which they propagate through two separate media. In order to quantitatively investigate this possibility, we define P_{1S} as the probability that there is exactly one excited atom in the medium for the case in which both photons propagate in the same medium, as in Fig. 3(a), and we define P_{2S} as the probability that there are two excited atoms under the same conditions. We also define P_{1D} and P_{2D} as the corresponding probabilities when the two photons propagate in different media, as in Fig. 3(b). In terms of the effective probability amplitudes defined above, these probabilities are given by

$$P_{1S}=|c(\gamma_1)|^2+|c(\gamma_2)|^2, \tag{37}$$

$$P_{2S}=|c(0)|^2, \tag{38}$$

$$P_{1D}=|c'(0)|^2(1-|c''(0)|^2)+|c''(0)|^2(1-|c'(0)|^2), \tag{39}$$

$$P_{2D}=|c'(0)|^2|c''(0)|^2. \tag{40}$$

Constructive interference between the two processes shown in Fig. 4 then suggests that

$$\frac{P_{2S}}{P_{2D}}=2 \tag{41}$$

should hold, at least to lowest order in perturbation theory.

From the adiabatic approximation, these probability amplitudes can be found by calculating the instantaneous eigenvectors of $H_{\rm eff}$, $H_{1\,\rm eff}$, and $H_{2\,\rm eff}$ that correspond to the perturbed form of the initial state vectors. For moderate values of $\sqrt{N}M/\delta$, the appropriate eigenvector in each case will be the one whose energy is nearest the initial value of zero. The results of a numerical calculation of the relevant eigenvectors in the limit of large N are summarized in Table I for the case in which $\delta_1=-2$, $\delta_2=3$, and $\sqrt{N}M=1/2$. (We will specify times in units of nanoseconds and energies in units of $\hbar$ divided by 1 ns, which puts the various plots and numerical results on a scale that is typical of experiments of this kind.) The results of Table I were calculated numerically to an accuracy of 40 significant digits but only the first 20 digits are shown in the table. Rather than simply taking the limit of large N by replacing $\sqrt{N-1}$ by $\sqrt{N}$ in the Hamiltonians, the numerical calculations were performed using a characteristically large value for N and a correspondingly small value of M. The specific results shown in Table I were obtained using $N=10^{12}$ and $M=0.5\times10^{-6}$, but equivalent results were obtained for other values of these parameters. The advantage of this approach is that it includes the usual mechanisms for nonlinear optics in which both photons interact with the same atom, the magnitude of which will be seen to be a factor of $1/N$ smaller than the exchange interactions of interest here.

TABLE I. Numerical results of an eigenvector calculation of the two-photon dressed state for the case in which $\delta_1=-2$, $\delta_2=3$, $N=10^{12}$, and $\sqrt{N}M=1/2$.

$c(\gamma_1,\gamma_2)$	0.959 141 163 220 186 939 59
$c(\gamma_1)$	0.154 292 855 116 767 135 53
$c(\gamma_2)$	−0.227 657 784 572 637 029 94
$c(0)$	−0.053 939 771 936 222 158 01
$c(\gamma_1,\gamma_1)$	0.021 988 495 337 845 905 76
$c(\gamma_2,\gamma_2)$	0.031 951 276 598 403 394 29
E_S	−0.038 245 115 666 580 845 33
$c'(\gamma_1)$	0.971 837 992 302 328 512 77
$c'(0)$	−0.234 534 582 685 425 689 06
$c'(\gamma_2)$	0.022 900 791 304 068 227 03
E_1	−0.120 665 473 331 518 234 61
$c''(\gamma_2)$	0.986 558 239 421 311 819 62
$c''(0)$	0.162 624 965 900 796 428 55
$c''(\gamma_1)$	0.015 998 771 693 051 438 80
E_2	0.082 420 357 664 940 187 49
P_{1S}	0.075 634 352 016 604 905 31
P_{1D}	0.078 543 851 013 127 825 91
P_{2S}	0.002 909 498 996 531 659 49
P_{2D}	0.001 454 749 498 267 077 32
P_{2S}/P_{2D}	1.999 999 999 998 284 816 99
$\langle N_e\rangle_S$	0.081 453 350 009 668 224 29
$\langle N_e\rangle_D$	0.081 453 350 009 661 980 56
E_1+E_2	−0.038 245 115 666 578 047 11

It can be seen from Table I that P_{2S}/P_{2D} is equal to 2, as expected, to a precision of 12 significant digits. The discrepancy in the twelfth decimal place is approximately equal to $1/N$, which reflects the contribution from conventional effects in which both photons do interact with the same atom. For example, the absorption of photon 1 will depopulate the ground state of one of the atoms, which in turn will prevent the virtual absorption and reemission of photon 2 by that

atom and thus give a conventional nonlinear phase shift (Kerr effect). These effects are of no interest here but they do illustrate the relatively small magnitude of conventional nonlinear interactions between individual photons. The eigenvalue calculations also showed that P_{2S} is proportional to N^2 in the weak coupling limit ($\sqrt{N}M/\delta \ll 1$), as expected.

The fact that $P_{2S}/P_{2D}=2$ to such a high degree of accuracy may be somewhat surprising, since the interference of probability amplitudes illustrated in Fig. 4 corresponds only to the lowest-order Feynman diagram and it might therefore have been expected that Eq. (41) may only hold to lowest order in perturbation theory. Our numerical results suggest that Eq. (41) is satisfied exactly for all values of $\sqrt{N}M/\delta$ in the limit of large N, at least up to the first level crossing. The fact that $P_{2S} \neq P_{2D}$ shows that there is an effective interaction between the two photons and that they do not propagate independently through the medium.

An examination of Table I also shows that there are some properties of the system that are the same whether the two photons propagate in the same medium or two different media. Let E_S be the energy of the two-photon dressed state that occurs when both photons propagate through the same medium, and let E_1 and E_2 be the energies of the single-photon dressed states that occur when each photon propagates in a different medium. The numerical eigenvalue calculations show that $E_S=E_1+E_2$ to within a precision of $1/N$, which demonstrates that the eigenstates of the total system have the same energy in both cases in the limit of large N. If that were not the case, there would be a nonlinear phase shift even in the absence of any laser pulses or other perturbations, which is not allowed by the symmetry arguments of Appendix A. The higher-energy eigenstates (those that do not correspond to $|\psi_0\rangle$) also have the same energies in both cases.

A similar situation can also be observed if we define $\langle N_e\rangle_S$ and $\langle N_e\rangle_D$ to be the mean number of excited atoms for the case in which the photons propagate in the same or different media:

$$\langle N_e\rangle_S \equiv P_{1S}+2P_{2S},$$

$$\langle N_e\rangle_D \equiv P_{1D}+2P_{2D}. \tag{42}$$

The numerical results of Table I show that

$$\langle N_e\rangle_S = \langle N_e\rangle_D \tag{43}$$

in the limit of large N, which demonstrates that the mean number of excited states is the same in the two cases; this is also required by the symmetry arguments of Appendix A. Equations (41)–(43) can be combined to obtain the difference in the probabilities of having exactly one excited atom:

$$P_{1S}-P_{1D}=-P_{2S}, \tag{44}$$

which shows that the probability of a single-excited-atom state is reduced when both photons propagate through the same medium. This result can be understood (in the context of perturbation theory) from the fact that the generation of a two-excited-atom state must come at the expense of depleting the probability amplitudes for the one-excited-atom states.

Although the numerical calculations leave little doubt that $P_{2S}/P_{2D}=2$ in the limit of large N, we have also obtained analytic solutions for the eigenvectors and related probabilities using Mathematica [22]. The resulting expressions are sufficiently complex and lengthy that they are of little practical use and have not been included here. These analytic solutions also satisfy Eqs. (41) and (43) in the limit of large N, at least to the extent that those results are obtained when any set of numerical values of the parameters are inserted into the analytic expressions; we have not yet succeeded in simplifying the analytic expressions to obtain Eqs. (41) and (43) directly. The factor of 2 can be easily derived for the case of equal detunings ($\delta_1=\delta_2$), as described in Appendix C.

V. INDIVIDUAL LASER PULSES

The factor of 2 increase in the probability of there being two virtually-excited atoms can be exploited in a variety of ways to produce a nonlinear phase shift. This section describes the effects of a single laser pulse, which is the simplest approach, while the next section considers the use of sequences of laser pulses to produce nonlinear phase shifts of arbitrary magnitude with minimal loss. A number of less effective approaches are described in Appendix B, including collisions with a buffer gas [6], Berry's geometric phase [14], and avoided level crossings [21].

It will be assumed for now that a single laser pulse is applied to the medium when the photon wave packets are centered on it. The electric field of the laser pulse will produce a change in the energy of level 2 of any excited atoms via the ac Stark shift [23] while giving a negligible transfer of population into level 3, as illustrated in Fig. 6. The duration of the laser pulse is assumed to be much shorter than the relevant time scales over which the population of the quantum states of the system can change in accordance with the Schrödinger equation and $H_{\rm eff}$. In that case, the net effect of the laser pulse will be to produce an impulsive phase shift $\Delta\varphi_e$ in the excited states of the atoms that is given by

$$\Delta\varphi_e = -\int \Delta E(t)dt/\hbar, \tag{45}$$

where $\Delta E(t)$ is the change in the energy of the excited states that is produced by the application of the field. The intensity and duration of the laser pulse can be adjusted to give any desired value of $\Delta\varphi_e$, which will be chosen to be $\pi/2$ for now.

If the state vector $|\psi\rangle$ had the form shown in Eq. (25) immediately before the laser pulse, then immediately after the laser pulse the system will be in a new state $|\psi'\rangle$ given by

$$|\psi'\rangle = \begin{pmatrix} c(\gamma_1,\gamma_2) \\ e^{i\pi/2}c(\gamma_1) \\ e^{i\pi/2}c(\gamma_2) \\ e^{i\pi}c(0) \\ c(\gamma_1,\gamma_1) \\ c(\gamma_2,\gamma_2) \end{pmatrix}. \tag{46}$$

The second and third components of the state vector correspond to a single excited atom and are subjected to a $\pi/2$ phase shift, while the fourth component corresponds to two excited atoms and experiences a total phase shift of π.

The new state vector $|\psi'\rangle$ can be conveniently expressed as a linear combination of the original state vector and another vector $|\psi_\perp\rangle$ that is orthogonal to $|\psi\rangle$:

$$|\psi'\rangle = re^{i\varphi}|\psi\rangle + |\psi_\perp\rangle, \tag{47}$$

where r and φ are both real. The coefficient of the $|\psi\rangle$ term is given by the projection of $|\psi'\rangle$ onto $|\psi\rangle$:

$$re^{i\varphi} = \langle\psi|\psi'\rangle = r' + i(|c(\gamma_1)|^2 + |c(\gamma_2)|^2). \tag{48}$$

Here

$$r' = |c(\gamma_1,\gamma_2)|^2 - |c(0)|^2 + |c(\gamma_1,\gamma_1)|^2 + |c(\gamma_2,\gamma_2)|^2 \tag{49}$$

is a real number that includes the contributions from the terms that were unaffected by the laser pulse as well as the contribution from the two-excited-atom state. For simplicity, we will consider the weak coupling limit in which r' is approximately equal to unity and the other terms, including φ, are much smaller in comparison. Expanding the left-hand side of Eq. (48) to first order [24] in φ then gives

$$\varphi = |c(\gamma_1)|^2 + |c(\gamma_2)|^2 = P_1. \tag{50}$$

Equation (50) applies to the case in which both photons are propagating in the same medium, while a similar result applies when the photons are propagating in two different media, so that

$$\varphi_S = P_{1S},$$

$$\varphi_D = P_{1D}, \tag{51}$$

where φ_S and φ_D are the overall phase shifts in the two cases. The nonlinear phase shift $\Delta\varphi_{\rm non}$ is then equal to the difference of the two

$$\Delta\varphi_{\rm non} = P_{1S} - P_{1D} \tag{52}$$

and Eq. (44) gives

$$\Delta\varphi_{\rm non} = -P_{2S} \tag{53}$$

in the limit of weak coupling.

Equation (53) shows that the nonlinear phase shift is directly proportional to the probability that two atoms will be virtually excited at the same time, which is a factor of 2 larger when the two photons propagate in the same medium due to the exchange interaction shown in Fig. 4. Based on the eigenvalue calculations of the preceding section, the nonlinear phase shift is thus expected to be proportional to N^2 in the weak coupling limit, which makes it much larger than the phase shift from conventional mechanisms such as that shown in Fig. 1. The nonlinearity depends on the fact that the two-excited-atom states undergo a phase shift of π, which is equivalent to a minus sign, and contribute to r' rather than to the phase shift as a result. No nonlinear phase shift would be obtained if the contribution to φ from the two-excited-atom states was simply twice the contribution from a single excited atom, as is the case in the limit of weak laser pulses ($\Delta\varphi_e \ll 1$); the net phase shift would then depend only on the mean number of excited atoms, which is the same in both cases from Eq. (43). This dependence on the overall phase of the two-excited-atom state can be viewed as a nonlocal property of the system, as is discussed in more detail in Sec. VII.

VI. SEQUENCES OF LASER PULSES

The construction of quantum logic gates will require a nonlinear phase shift of π, which cannot be produced by a single laser pulse from Eq. (53). The probability of generating the orthogonal state $|\psi_\perp\rangle$ in Eq. (47) must also be minimized, since it corresponds to a loss mechanism in which the system makes a transition out of the basis of states that represent the qubits in a quantum computer. For a single laser pulse, the probability of such a transition is on the order of P_1 and is larger than $\Delta\varphi_{\rm non}$. Both of these difficulties can be avoided by using an appropriate sequence of laser pulses designed to give a phase shift of π with $|\psi_\perp\rangle = 0$. The optimal design of pulse sequences of this kind is a nonlinear optimization problem that is still under investigation. Here we will describe two different approaches, one based on a sequence of short laser pulses as described above and a second, more efficient approach that makes use of longer pulses with narrow bandwidths.

In the first approach, a sequence of n_p short pulses is applied at times t_j with amplitudes a_j. The time intervals between the pulses were assumed to be sufficiently small compared to τ_p that $\mathcal{E}_0(t)$ was approximately constant throughout the pulse sequence. The value of n_p was chosen to be sufficiently large (~ 10) that there were more than enough degrees of freedom to cancel all of the components of $|\psi_\perp\rangle$. A Monte Carlo approach was used in which a set of initial values for the t_j and a_j were chosen at random and used as the starting point for a numerical algorithm that varied the t_j and a_j to minimize the ratio of the loss (squared modulus of $|\psi_\perp\rangle$) divided by the net nonlinear phase shift, where the time evolution of the Schrödinger equation was calculated numerically. A random set of initial values may only lead to a local minimum, but the process was repeated many times until the optimal solution was obtained. Most of the randomly-chosen starting points lead to a solution with $|\psi_\perp\rangle = 0$ but the corresponding values of $\Delta\varphi_{\rm non}$ varied considerably.

During a laser pulse, the system will be excited into a virtual state in which the atom is in level 3 and one or both of the photons have been absorbed, as illustrated in Fig. 6. The detuning of this virtual state will be different for states $|\gamma_1\rangle$ and $|\gamma_2\rangle$, which causes the phase shift for $|\gamma_2\rangle$ to differ from that of $|\gamma_1\rangle$ by a factor f that could be controlled by adjusting the frequency of the laser pulse. We included this possibility in the analysis by taking the phase shifts to be given by

$$\varphi_1 = \Delta\varphi_e,$$

$$\varphi_2 = f\Delta\varphi_e, \tag{54}$$

$$\varphi_0 = (1+f)\Delta\varphi_e,$$

TABLE II. Optimal results obtained from a sequence of 10 short laser pulses as a function of the parameter f. The condition $|\psi_\perp\rangle = 0$ was satisfied in all cases.

f	$\Delta\varphi_{\text{non}}$
2.0	0.318
1.1	0.125
1.01	0.022
1.00	no solution

which generalizes Eq. (45). Here φ_1, φ_2, and φ_0 are the phase shifts in the states $|\gamma_1\rangle$, $|\gamma_2\rangle$, and $|0\rangle$, and the value of $\Delta\varphi_e$ depends on the amplitude of the laser pulses (all the pulses were assumed to have the same duration).

The optimal results obtained for a sequence of 10 laser pulses are summarized as a function of f in Table II, where the squared modulus of $|\psi_\perp\rangle$ after the sequence of pulses was zero in all cases. It can be seen that the optimal value of the nonlinear phase shift decreases as f approaches 1, and no solution could be found with $|\psi_\perp\rangle = 0$ for the case of $f = 1$. This suggests that an asymmetry between the effects of photon 1 and photon 2 is required in order to obtain nonlinear phase shifts with no loss. (A similar asymmetry is also required for the case in which collisions with a buffer gas are used instead of laser pulses, as is discussed in Appendix B.) The magnitude of $\Delta\varphi_{\text{non}}$ from a sequence of short pulses is relatively small in any case and this approach is straightforward but may be of limited practical value.

We have therefore investigated a more complex but much more efficient five-pulse approach that uses longer laser pulses with narrow bandwidths to produce transitions between specific states of the system. In this method, the two incident photons are assumed to be closer to resonance with level 3 than level 2, as illustrated in Fig. 7, but the detuning is still sufficiently large that the population in level 3 is relatively small. The frequency of the laser pulses can then be tuned to produce resonant transitions into level 2, where one frequency will produce a resonant absorption of photon 1 and another frequency will produce a resonant absorption of photon 2. Since level 3 is again a virtual state, the net effect can be represented as an effective matrix element for photon absorption into level 2.

The frequency and amplitude of the first laser pulse are chosen to produce a resonant absorption of photon 2 (a Rabi oscillation [23,25] of π), so that the system makes a complete transition from state $|\gamma_1, \gamma_2\rangle$ to state $|\gamma_1\rangle$ as illustrated in Fig. 8. A comparison of the matrix elements of H_{eff} in Eq. (31) with $H_{2\,\text{eff}}$ of Eq. (35) shows that the Rabi frequency for this transition is the same whether the two photons are in the same medium or in two different media, so that this transition occurs in either case.

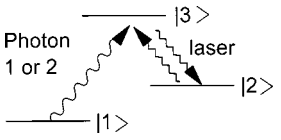

FIG. 7. A laser-induced transition, in which photon 1 or 2 is off-resonance from level 3, but the application of a laser pulse allows a resonant transition into level 2.

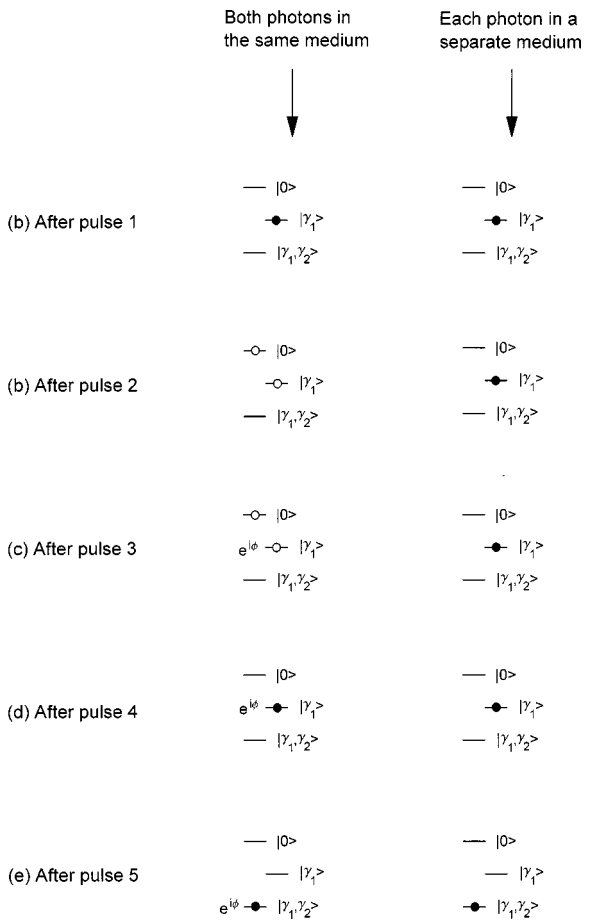

FIG. 8. A five-pulse sequence producing a nonlinear phase shift of π. (a) Pulse 1 produces a transition from the initial state $|\gamma_1, \gamma_2\rangle$ to the state $|\gamma_1\rangle$ in which only photon 1 is present. (b) Pulse 2 has no net effect when the photons are in two different media but produces a superposition of states $|\gamma_1\rangle$ and $|0\rangle$ when both photons are in the same medium. (c) Pulse 3 produces a phase shift in the state $|\gamma_1\rangle$ when both photons are in the same medium. (d) Pulse 4 returns the system to state $|\gamma_1\rangle$. (e) The last pulse returns the system to its initial state aside from a relative phase shift of π.

The frequency of the second pulse is then chosen to be on resonance for photon 1 transitions and its amplitude is adjusted to produce a complete (2π) Rabi oscillation back into the initial state $|\gamma_1\rangle$ for the case in which the two photons are in separate media. A comparison of H_{eff} with $H_{1\,\text{eff}}$ now shows that the relevant matrix element is a factor of $\sqrt{2}$ larger for the case in which the two photons are in the same media due to the quantum interference of Fig. 4. As a result, the probability amplitude for state $|0\rangle$ oscillates through zero and the system is left in a superposition of $|\gamma_1\rangle$ and $|0\rangle$ in the latter case, as illustrated in Fig. 9(a). These results were obtained by numerical integration of Schrödinger's equation for a gaussian laser pulse with a width (standard deviation) of 30 ns.

The fact that the system is now in a superposition of states if and only if the photons are propagating in the same medium allows a third pulse to produce an arbitrary phase shift in that case. The frequency of pulse 3 is chosen to be slightly off-resonance from a photon 1 transition and its amplitude is chosen, once again, to return the system to state $|\gamma_1\rangle$ for the case in which the two photons are in different media (another

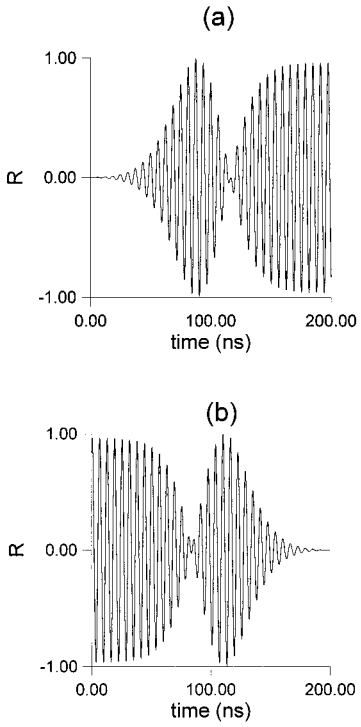

FIG. 9. Plots of the real part R of the probability amplitude of state $|0\rangle$ as a function of time for the case in which both photons propagate in the same medium, illustrating the effects of a sequence of five laser pulses. (a) Effects of pulse 2, which produces a superposition of states $|\gamma_1\rangle$ and $|0\rangle$. (b) Effects of pulse 4, which reverses the effects of pulse 2 and returns the system to state $|\gamma_1\rangle$, aside from a phase shift that can be controlled by pulse 3.

2π Rabi oscillation). The effects of this pulse on the probability amplitude of state $|\gamma_1\rangle$ for the case in which both photons propagate in the same medium can be seen from Fig. 10, where the radius of the dashed circle is equal to the modulus of the probability amplitude of this state just before the pulse. The vector labeled **a** in the figure represents the contribution from the probability amplitude of state $|\gamma_1\rangle$ before the pulse, which is reduced in magnitude by its coupling into state $|0\rangle$ during the pulse. The vector labeled **b** represents the contribution from the probability amplitude of state $|0\rangle$ before the pulse, which is coupled back into state $|\gamma_1\rangle$ during the pulse. The magnitude of vector **b** can be adjusted by varying the detuning of the pulse, while the phase of the pulse can be used to ensure that the sum of the two vectors lies on the dashed circle. This allows the modulus of the amplitude of state $|\gamma_1\rangle$ to be maintained at its original value while an arbitrary phase shift can be introduced by moving the resultant vector to an arbitrary point on the dashed circle.

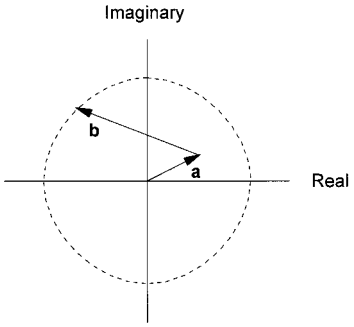

FIG. 10. Real and imaginary parts of the probability amplitude of state $|\gamma_1\rangle$ as a result of pulse 3, for the case in which both photons propagate in the same medium. The radius of the dashed circle represents the magnitude of the probability amplitude of state $|\gamma_1\rangle$ before the pulse, while vector **a** represents the contribution from that probability amplitude alone after the pulse has partially coupled it into state $|0\rangle$. Vector **b** represents the contribution from the initial probability amplitude of state $|0\rangle$, which is partially coupled into $|\gamma_1\rangle$ by the pulse. The phase and detuning of the pulse can be adjusted to make the resultant vector lie anywhere on the dashed circle, which gives an arbitrary phase shift.

The reason for maintaining the modulus of $|\gamma_1\rangle$ during pulse 3 is that it allows a fourth pulse to act as the inverse of pulse 2, since the state of the system is now the same as it was after pulse 2 except for a phase shift. The amplitude and frequency of pulse 4 are therefore chosen to be the same as pulse 2, which applies a 2π Rabi oscillation and leaves the system in $|\gamma_1\rangle$ once again for the case in which the two photons are in separate media. At the same time, the phase of this pulse can be adjusted to eliminate the $|0\rangle$ component and leave the system entirely in $|\gamma_1\rangle$ for the case in which the two photons are in the same medium, as shown in Fig. 9(b).

A fifth pulse that is identical to pulse 1 is then applied to produce a π Rabi oscillation and transfer the system back to the original state $|\gamma_1, \gamma_2\rangle$, aside from the phase shift that was generated during pulse 3. Once again, the matrix elements and the Rabi frequency are the same for this transition whether the photons travel in the same or different media.

A numerical algorithm was used to determine the correct parameters for the five-pulse sequence described above. The net effect of the resulting pulse sequence was to produce a phase shift of π for the case in which both photons travel through the same medium relative to the case in which they travel through two different media. Alternatively, an arbitrary nonlinear phase shift could be produced using a different choice of the frequency, amplitude, and phase of pulse 3. This approach does not produce any losses (in the form of an orthogonal state vector $|\psi_\perp\rangle$) to within the approximations that were described above.

The above analysis assumed once again that the only relevant modes of the electromagnetic field are those generated by $a_1^\dagger$ and $a_2^\dagger$. It is expected that this condition can be satisfied experimentally by using a thick medium and appropriate phase matching conditions, in which case conservation of energy and momentum can greatly suppress the emission of photons into other modes. More detailed numerical calculations that include the effects of scattering and dispersion are planned.

VII. NONCLASSICAL NATURE OF THE RESULTS

One question that naturally arises is whether or not these nonlinear phase shifts can be understood classically or if they are inherently quantum-mechanical in nature. In this section, we consider the possibility of describing the photons as either classical particles or as classical waves, and conclude that neither description is consistent with the observed effects. We then show that nonlinear phase shifts of this kind cannot be produced by a local polarizability of the medium

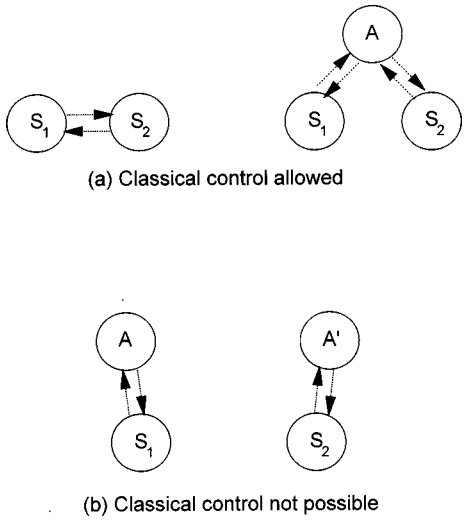

FIG. 11. (a) Two classical systems, S_1 and S_2, that are connected by a sequence of physical interactions that may involve one or more auxiliary systems labeled *A*. (b) Two classical systems that are not connected by a sequence of physical interactions. There is no path for the flow of information in the latter case and a classical control process cannot occur.

and suggest that the only correct interpretation must involve nonlocal correlations between fluctuations in the polarization of the medium at two distant locations.

Any consistent classical interpretation would have to include the particlelike nature of light and the fact that a measurement could, at least in principle, be performed to determine which photon interacted with which atom. If we assume that the photons are classical particles, then the probability that both photons will interact with the same atom is negligible for a typical medium, as discussed above. This places a fundamental limitation on the ability of one photon to control the state of the other in any classical theory since, in general, the control of one classical system by another is only possible if there is some physical interaction (force) connecting the two systems, either directly or through a chain of interacting systems as illustrated in Fig. 11(a). No control is possible if each system interacts only within two disconnected sets of systems, as illustrated in Fig. 11(b). The sequence of physical interactions connecting the two system in Fig. 11(a) provides a path for the flow of information from one system to the other and is consistent with the assumption that there must be a specific cause for every effect (determinism). In contrast, the quantum-mechanical exchange interaction of Fig. 4 has the same form as the two disconnected systems in Fig. 11(b), which suggests that one photon can control the state of another photon even when there is no sequence of physical interactions connecting the two particles [26].

Although the exchange interaction of Fig. 4 has the same form as the two disconnected systems in Fig. 11(b), we do not know which photon will interact with which atom. From a classical point of view, that uncertainty is irrelevant: if the photons never interact with the same atom, there is no path for the flow of information, regardless of which photon interacted with which atom. In quantum mechanics, the interference of these probability amplitudes gives us the possibility of a control process even though there is no classical interaction between the two photons. In the quantum-mechanical exchange interaction illustrated in Fig. 2, atom *A* absorbs photon 1 and reemits photon 2, while atom *B* absorbs photon 2 and re-emits photon 1, which suggests that both photons must interact with both atoms in some sense in a quantum-mechanical description. This is once again irrelevant from a classical point of view, since the photons never have a causal effect on the same atom and no path for the flow of classical information can be identified as a result.

If we were to simply ignore the particlelike nature of light and represent the photons by classical waves, the intensity of the light beams would be so low that they would have negligible effect on the properties of the atoms and they could not produce a significant change in the index of refraction of the medium. For example, suppose that an absorptive filter is placed in front of the two incident beams of light in order to reduce their intensities by a large factor f_a. In a classical theory in which the nonlinear effects are proportional to the product of the intensities, the nonlinear phase shift would be reduced by a factor of f_a. In contrast, the nonlinear phase shifts of interest here would be unaffected by such an attenuation process, provided that we only accept events in which both photons are transmitted through the attenuator and actually detected. The persistence of nonlinear effects at arbitrarily low intensities is a hallmark of nonclassical behavior [27,28].

More generally, it is possible to give a simple proof that the predicted phase shifts are inconsistent with any classical theory in which the medium responds locally to an applied field. To show this, we make the usual assumption [7–9] that the nonlinear response of the medium can be described by a series of nonlinear susceptibility coefficients. Since there are four electric fields involved here (two incoming and two outgoing), the relevant dipole moment $\mathbf{P}(\mathbf{r},t)$ induced at location $\mathbf{r}$ and time t is given [7–9] by

$$\mathbf{P}(\mathbf{r},t)=\boldsymbol{\chi}^{(3)}\cdot\mathbf{E}(\mathbf{r},t)^3, \tag{55}$$

where $\mathbf{E}(\mathbf{r},t)$ represents the classical field and $\boldsymbol{\chi}^{(3)}$ is the third-order susceptibility coefficient. The change $\delta\mathbf{E}(\mathbf{r}',t')$ in the electric field produced in the forward direction can then be found by integrating over the volume of the medium

$$\delta\mathbf{E}(\mathbf{r}',t')=\int G(\mathbf{r}',t';\mathbf{r},t)\mathbf{P}(\mathbf{r},t)d^3\mathbf{r}\,dt, \tag{56}$$

where $G(\mathbf{r}',t';\mathbf{r},t)$ is the appropriate Green's function. $\mathbf{E}(\mathbf{r},t)$ in Eq. (55) can be replaced by the incident field $\mathbf{E}_0(\mathbf{r},t)$ in the limit of weak fields, which gives

$$\delta\mathbf{E}(\mathbf{r}',t')=\int G(\mathbf{r}',t';\mathbf{r},t)\boldsymbol{\chi}^{(3)}\cdot\mathbf{E}_0(\mathbf{r},t)^3d^3\mathbf{r}\,dt. \tag{57}$$

All of the volume elements contribute with the same phase in the forward direction, in which case the integral of Eq. (57) is proportional to the volume of the medium. Since the induced phase shift is proportional to $\delta\mathbf{E}$, this gives a nonlinear phase shift proportional to N, not N^2, which shows that the local nature of the induced dipole moment precludes any description of these effects in terms of nonlinear susceptibilities.

Equation (57) shows that the nonlinear phase shift cannot be due to a local polarization of the medium, which suggests that the correct interpretation must involve nonlocal correlations between the polarizations induced at two different locations in the medium. This is consistent with the factor of 2 increase in the probability of there being two excited atoms, each of which has a dipole moment. Since the classical phase associated with a single photon is totally random, these induced dipole moments have zero mean, while the factor of 2 indicates a nonlocal correlation between the two.

These nonlinear phase shifts are due to the interference of quantum-mechanical probability amplitudes, which reflects the fact that we do not know which photon interacted with which atom. This dependence on quantum interference provides an interesting example of complementarity in quantum mechanics: In principle, a measurement could be performed to determine which photon interacted with which atom, and such a measurement would always show a negligible probability for both photons to have interacted with the same atom. On the other hand, any such measurement would also destroy the quantum interference that is responsible for the nonlinear phase shift. In that case, can we really say that the phase shift is due to photons that never interacted with the same atom? What we can say is that there is no classical interaction between the two photons, as in Fig. 11(b), and that the effect is not due to a sequence of interaction terms in the quantum-mechanical Hamiltonian, just as is the case for the usual exchange interactions.

The random nature of the quantum theory is clearly incompatible with the classical assumption that every effect must have a specific cause (determinism). This is especially true of the nonlocal correlations between the random results of measurements made on pairs of distant particles, which are inconsistent with any deterministic interpretation in which information does not travel faster than the speed of light [29]. Our results show that the inconsistency between quantum mechanics and classical determinism is not limited to random events; a quantum control process of this kind has a definite result even though, from a classical point of view, it is not possible to identify a path for the flow of information or a specific cause for the outcome of the process.

Finally, it can be seen from Eq. (46) that these effects are inherently dependent on the overall phase of a two-photon state, which is a nonlocal property of the system. As a result, there are some analogies between this effect and the two-photon interferometer [30,31], which does violate Bell's inequality. Although this system does not violate Bell's inequality and the two photons have uncertain positions in overlapping beams, it seems apparent that these effects are inherently nonlocal as well as nonclassical.

VIII. DISCUSSION AND CONCLUSIONS

In this paper, we have shown that exchange interactions can be used to obtain nonlinear phase shifts at the two-photon level. The origin of the nonlinear phase shifts can be understood from the fact that quantum interference effects can produce a factor of 2 increase in the probability of there being two virtually-excited atoms when two nonresonant photons propagate through the same medium, in analogy with the Hanbury-Brown and Twiss effect (photon bunching). The application of one or more laser pulses will then produce a nonlinear phase shift, since the effects of the pulses are dependent on the number of excited atoms. Nonlinear optical effects from conventional mechanisms are normally very weak at the two-photon level because of the weak electric field associated with the photons and the requirement that both photons interact with the same atom. The use of exchange interactions avoids this difficulty, since the two photons can interact with two different atoms, which is much more likely to occur.

The question naturally arises as to whether or not these effects are similar to any of the nonlinear mechanisms that have been discussed by earlier authors who may not have emphasized the role of exchange interactions. For example, a four-wave mixing experiment [23] at low intensities is similar to the extent that there are two incoming and two outgoing beams, and a nonlinear phase shift proportional to N should be expected from coherent forward scattering via conventional mechanisms such as that illustrated in Fig. 1. But the effects of interest here are proportional to N^2 whereas Eq. (57) shows that phase shifts of that kind cannot be obtained from conventional approaches [7–9] based on the use of nonlinear susceptibility coefficients. In addition, the results of Appendixes A and B suggest that exchange interactions cannot give rise to any nonlinear phase shifts unless the system is non-adiabatically perturbed by an additional field, which is not the case in most conventional nonlinear optics experiments. Finally, the results obtained here require that the two incoming beams of light be sufficiently well collimated that they satisfy Eq. (1), as in the Hanbury-Brown and Twiss effect [4], which is certainly not a feature of most other effects in nonlinear optics. For these reasons, the use of exchange interactions to produce nonlinear phase shifts appears to be qualitatively different from earlier mechanisms.

Our approach involves pairs of atoms whereas, to the best of our knowledge, all of the previous mechanisms [1,10–12] for the production of nonlinear phase shifts at single-photon intensities have involved the interaction of two photons with the same atom. For example, the nonlinear phase shifts observed by Turchette *et al.* [1] were obtained using a high-Q cavity and a ''V'' system in which one photon could depopulate the ground state of an atom, thereby preventing any interaction with that atom by a second photon. Their experiments were performed under conditions in which there was typically only one atom in the cavity at any given time, so that both photons must have interacted with the same atom. Schmidt and Imamoglu [11] have proposed a mechanism in which electromagnetically induced transparency (EIT) [32] could be used to greatly reduce absorption near an atomic resonance, which would allow an incident beam of light to undergo a large phase shift by being tuned very near resonance; the phase shift could then be modulated (Kerr effect) using a control beam that produces a ''shelving'' transition into a fourth atomic state. The effect can be further enhanced by confining the photons to a resonant cavity [12]. Their approach clearly requires that at least one photon from the control beam and a second photon from the probe must both interact with the same four-level atom, while the probability of such an event is greatly enhanced by the small detuning and the use of a cavity. Harris and Yamamoto [33] have described a similar optical switch using EIT and the same

four-level scheme but with controlled absorption rather than a phase shift, while other mechanisms involving coherent population trapping and other forms of quantum interference have also been suggested [34,35]. Harris and Yamamoto [33] showed that, in their approach at least, the minimum photon flux in the control beam corresponds to one photon per cross-sectional area, which is sufficient to ensure that there will always be an interaction between a control photon and every atom in the medium. In contrast, our approach eliminates the need for both photons to interact with the same atom, which allows the use of a single control photon even when there are a large number of atoms in the medium.

Cooperative effects involving two or more atoms can play an important role in a number of other phenomena in quantum optics, such as superradiance [36] and two-photon absorption [37–40]. Superradiance can be viewed as an extreme case of photon bunching and is thus somewhat analogous to the effects discussed here, although it does not produce nonlinear phase shifts nor does it involve only two photons. Teich and Wolga [37] showed that the rate of two-photon absorption by a single atom (or any pointlike absorber) is twice as large for chaotic light as it is for coherent light, which is analogous to both the Hanbury-Brown and Twiss effect and to Eq. (41). Cooperative effects in two-photon absorption have been considered in detail [38–41], but most discussions either assumed a single mode of the field [38,40–41], in which case there are no exchange interactions as shown in Figs. 2 and 4, or Eq. (1) did not hold [39]. Multiatom effects can also be produced by long-range dipole-dipole interactions [39,40] or local field effects [42], but these are physical interactions rather than exchange interactions.

Nonlinear phase shifts of this kind can be used to implement quantum logic gates [13] that may be of practical use in quantum computing. One of the advantages of an optical approach is the fact that the logic gates would be physically independent from each other and could be connected as desired using optical fibers, whereas the difficulty in providing a logical connection between arbitrary devices is a major drawback in other approaches being considered [43]. One potential difficulty in any optical approach to quantum computing is the need to produce large numbers of single-photon input states on demand. We have previously suggested [44] that large numbers of single-photon states could be efficiently produced by starting with a weak coherent state containing an average of one photon in each of a large number of optical fibers. The optical XOR quantum logic gates could then be used to perform a quantum nondemolition measurement of the number of photons, after which a series of electro-optic switches could select the output of those fibers containing one and only one photon. Optical fiber loops could be used as memory storage devices; although the intrinsic storage time would be at most a few microseconds, the switching time of the logic gates would be many orders of magnitude faster, which may allow the use of quantum error correction techniques. Highly efficient methods for single-photon detection would also be required in order to read out the results at the end of a calculation; this could also be accomplished by using the XOR gates to perform quantum nondemolition measurements of the photon number, which could be repeated to obtain very high overall single-photon detection efficiencies. The practical issues involved in the implementation of a quantum computer have been described in more detail elsewhere [44], where the conclusion appears to be that all of the necessary functions could be performed provided only that the XOR gates themselves are sufficiently efficient. The practical issues involved in the implementation of a quantum computer are beyond the intended scope of this paper, which is primarily concerned with the fundamental physical implications of the use of exchange interactions in a quantum control process.

There are a number of areas of physical importance that still need to be investigated in more detail, including the potential effects of dispersion and scattering, which are believed to be relatively small but have been neglected here. The use of phase-matching techniques to suppress the emission of photons into other modes of the field appears promising but will require further analysis. A more detailed numerical investigation of these and other issues is planned.

In our opinion, the most significant conclusion from this work is that it is possible to implement a quantum control process even when there is no physical interaction between two systems, as in Fig. 11(b). This shows that the discrepancy between quantum mechanics and classical determinism is not merely a question of randomness, nor is it limited to random events: a quantum control process of this kind has a definite outcome, even though, from a classical point of view, it is not possible to identify a path for the flow of information or a specific cause for the outcome of the event. Although these nonlinear phase shifts do not violate Bell's inequality, they can be viewed as being due to nonlocal correlations between fluctuations in the polarization of the medium at two different locations. Our most fundamental understanding of the nature of control and ''cause-and-effect'' may have to be revised to include nonclassical control processes of this kind.

ACKNOWLEDGMENTS

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APPENDIX A: SYMMETRY CONSIDERATIONS

There are a number of other Feynman-like diagrams involving pairs of atoms in addition to the exchange interaction shown in Fig. 2. For example, the various events can occur in a different time order or each atom may absorb and reemit the same photon. We showed in an earlier paper [6] that the contributions from all of these processes cancel out, at least to lowest order in perturbation theory, and that there will be no net nonlinear phase shift unless the system is perturbed in a way that eliminates the cancellation. The reason for this cancellation can be understood from the following symmetry considerations.

It will be assumed that the atomic transitions of interest are between a ground state with magnetic quantum number $m=0$ and three excited states, corresponding to $m=1$, 0, or -1, that are degenerate in the absence of any external field. The absorption of a photon that is linearly polarized along the x axis and incident along the z axis will induce a transi-

tion to an atomic state $|\varphi_x\rangle$ that is a linear combination of states with $m=\pm 1$; in the forward direction, such a state can only reemit an x-polarized photon. In a similar manner, y-polarized photons propagating along the z direction are only coupled to an orthogonal atomic state $|\varphi_y\rangle$. As a result, the exchange interaction of Fig. 2 can only occur if the two photons have the same polarization and there will be no nonlinear phase shift of this kind for two photons of different polarizations.

Consider the case in which photon 1 is linearly polarized along the x direction while photon 2 is linearly polarized along a direction that is midway between the x and y axes, so that the initial state of the field can be written as

$$|\psi_0\rangle=|x_1\rangle(|x_2\rangle+|y_2\rangle)/\sqrt{2}. \tag{A1}$$

The nonlinear phase shift due to exchange interactions will be assumed to have a magnitude of π, so that, in the absence of any external fields, the state of the photons after passing through the medium will be

$$|\psi\rangle=|x_1\rangle(e^{i\pi}|x_2\rangle+|y_2\rangle)/\sqrt{2}, \tag{A2}$$

where the nonlinear phase shift has been applied only to the term in which both photons have the same polarization. It can be seen from Eq. (A2) that the polarization of photon 1 remains unchanged while that of photon 2 has been rotated through an angle of 90°.

Now consider a new coordinate frame x',y' that has been rotated by 45° with respect to the original x,y coordinate frame. The initial state of the system can now be written as

$$|\psi_0\rangle=(|x_1'\rangle+|y_1'\rangle)|y_2'\rangle/\sqrt{2} \tag{A3}$$

and the final state becomes

$$|\psi\rangle=(|x_1'\rangle+e^{i\pi}|y_1'\rangle)|y_2'\rangle/\sqrt{2}. \tag{A4}$$

In this case, the polarization of photon 2 remains unchanged while the polarization of photon 1 is rotated by 90°. Equations (A2) and (A4) are inconsistent with each other, which shows that a nonlinear phase shift of this kind is not possible for a medium with rotational symmetry and degenerate atomic states. This explains why the Feynman diagrams all cancel in the absence of an external perturbation and it also shows that the mean number of excited atoms must be the same whether the photons propagate in the same or two different media, as in Eq. (43), since a nonzero energy difference and nonlinear phase shift would be obtained if that were not the case.

APPENDIX B: ALTERNATIVE APPROACHES

Perhaps the simplest way to eliminate the rotational symmetry of the system would be to apply a linearly polarized laser beam as illustrated in Fig. 6, where the laser intensity is assumed to be constant in time rather than pulsed. Since the population in level 3 may play a significant role here, we explicitly included those states in the system rather than approximating their effects with an effective coupling Hamiltonian and energy shift, as was done for the pulse sequences described in the text. This gives an 11-dimensional effective state vector and Hamiltonian, which were derived in the same way as the six-dimensional state vector used for the two-level system. The nonlinear phase shift was once again determined numerically by calculating the corresponding eigenvalues, also in analogy with the six-state calculations. The results of this analysis showed that the application of a continuous laser beam does not produce any nonlinear phase shifts proportional to N^2, despite the breaking of the rotational symmetry. Conventional linear phase shifts proportional to N were obtained, however, and it was apparent that the application of the laser beam could be used to ''control'' the state of the photons by producing a rotation of the polarization of the photons, as was independently observed recently by Wielandy and Gaeta [45]. We also considered a number of other configurations, such as the commonly-used ''V'' and ''Λ,'' but were unable to obtain any nonlinear phase shifts that would be significant at the two-photon level. The conclusion appears to be that steady-state perturbations of the system are not sufficient to eliminate the cancellation of the Feynman diagrams discussed above.

Since time-independent perturbations do not suffice, we also considered slowly-varying perturbations where the time evolution of the system would be given by the adiabatic approximation and the associated Berry phase might produce a nonlinear phase shift. Berry [14] considered the case in which the Hamiltonian $H(\mathbf{R}(t))$ depends on two parameters that can be considered to be the components of a two-dimensional, time-dependent vector $\mathbf{R}(t)$. If the system is slowly propagated in one of the eigenstates $|n(\mathbf{R})\rangle$ of the Hamiltonian around a closed path C in this two-dimensional parameter space, the system will undergo a geometric phase shift $\Delta\varphi_g$ that is given by the surface integral

$$\Delta\varphi_g=-\int\int_C d\mathbf{S}\cdot\mathbf{V}_n(\mathbf{R}), \tag{B1}$$

where the vector $\mathbf{V_n(R)}$ is defined as

$$\mathbf{V_n}(\mathbf{R})=\mathrm{Im}\sum_{m\neq n}\frac{\langle n(\mathbf{R})|\nabla_{\mathbf{R}}H(\mathbf{R})|m(\mathbf{R})\rangle\times\langle m(\mathbf{R})|\nabla_{\mathbf{R}}H(\mathbf{R})|n(\mathbf{R})\rangle}{[E_m(\mathbf{R})-E_n(\mathbf{R})]^2}. \tag{B2}$$

Here $E_n(\mathbf{R})$ is the energy of eigenstate $|n(\mathbf{R})\rangle$. We considered a set of possible parameters that included the time-dependent amplitude and phase of the external laser beam as well as the photon gaussian envelope function $\mathcal{E}_0(t)$. The vector $\mathbf{V}_n(\mathbf{R})$ was found to be zero for all combinations of two of these parameters. Although it is possible that we did not consider some relevant set of parameters, the conclusion appears to be that a Berry phase cannot be used to produce nonlinear phase shifts of this kind. We also investigated the possibility of adiabatically varying the parameters in such a way as to pass through a region of an avoided level crossing, and we found that this also did not produce a nonlinear phase shift proportional to N^2.

The above results suggest that the perturbation that is required to break the rotational symmetry of the system must also be nonadiabatic, as it is for the two types of laser sequences discussed in the text. The remaining possibility that we have considered in some detail is the use of a static field to break the symmetry in combination with collisions with a buffer gas to produce random phase shifts in the excited states of the atoms. This analysis was also based on the six-dimensional effective state vector described in the text, but here a density matrix calculation was required since we were no longer dealing with pure states. The simplest assumption that one can make with regard to the effects of the collisions is that they are instantaneous events, that they randomize the phase of the excited states of the atoms, and that they occur at random times at a rate that is independent of the detuning (energy unbalance) of the virtual atomic states. Given those assumptions, it can be shown that the off-diagonal terms of the density matrix involving states $|\gamma_1\rangle$, $|\gamma_2\rangle$, and $|0\rangle$ decay at rates β_1, β_2, and β_0 that are related by

$$\beta_1=\beta_2=\tfrac{1}{2}\beta_0. \tag{B3}$$

The decay rate β_0 is twice as large as the other two because it corresponds to a state with two excited atoms, which doubles the probability that a collision event will occur in a small time interval.

A numerical calculation of the time-evolution of the density matrix gave the result that no nonlinear phase shift proportional to N^2 will be produced by atomic collisions if Eq. (B3) is satisfied. The assumption that the collision rate is independent of the detuning of these virtual atomic states does not seem plausible, however, for an inelastic collision process with a final state energy exchange that is resonant with the energy of state $|\gamma_1\rangle$ but not in resonance with the energy of $|\gamma_2\rangle$, for example, which would enhance the rate of collisions for $|\gamma_1\rangle$. The same can be said for a collision process that is elastic but has a virtual state whose energy difference is close to resonance with $|\gamma_1\rangle$ or $|\gamma_2\rangle$. We therefore performed additional density matrix calculations in which $\beta_1\neq\beta_2$, where it was assumed that

$$\beta_0=\beta_1+\beta_2. \tag{B4}$$

The results of a typical calculation of this kind are shown in Fig. 12, which is a plot of the nonlinear phase shift as a function of the average detuning, $\delta=(\delta_1+\delta_2)/2$. The average detuning was varied while the difference in detunings, $\delta_2-\delta_1$, was held fixed at a constant value of 5 GHz, which may be typical of experiments of this kind. The results

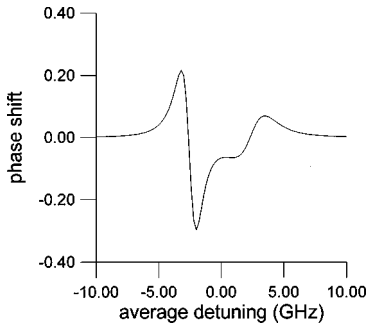

FIG. 12. Nonlinear phase shift as a function of the average photon detuning for the case of collisions with a buffer gas. A nonzero phase shift is only obtained if $\beta_1\neq\beta_2$; here $\beta_1=2\beta_2$.

shown in the figure correspond to $\beta_1=2\times10^9\,\mathrm{s}^{-1}$, $\beta_2=1\times10^9\,\mathrm{s}^{-1}$, and $\beta_0=3\times10^9\,\mathrm{s}^{-1}$. The magnitude of the nonlinear phase shift as a function of the number of atoms, for a fixed average detuning of $\delta=-1/2$, is shown in Fig. 13; these results are consistent with a nonlinear phase shift proportional to N^2.

The calculations described in Ref. [6] had assumed that the details of the collision process were not crucial and had simply taken equal damping rates for all of the virtual states, which is inconsistent with Eq. (B3). On the other hand, similar results are obtained provided that $\beta_1\neq\beta_2$. This condition may occur, for example, if a magnetic field produces a Zeeman shift in the atomic energy levels whose magnitude is comparable to the difference in the detunings of the two photons, as was the case in the preliminary experiments described in Ref. [44]. In that case, an inelastic collision process that transfers angular momentum to the atoms may be closer to resonance with $|\gamma_1\rangle$ than $|\gamma_2\rangle$, which would be expected to give $\beta_1\neq\beta_2$. In any event, it is now clear that the details of the collision process do play an important role and that the effects of a laser pulse are much easier to analyze and control, which is why we are now concentrating on the effects of sequences of laser pulses.

Finally, we note that all of the mechanisms that we have investigated and found to produce N^2 nonlinear phase shifts (without substantial loss) involve nonadiabatic perturbations that are asymmetric with respect to the role of the two photons. The impulsive phase-shift approach requires that the laser pulses be detuned an unequal distance from $|\gamma_1\rangle$ and $|\gamma_2\rangle$, the longer laser-pulse approach allows the selective absorption of one photon or the other, and the buffer-gas approach requires that $\beta_1\neq\beta_2$. The need for this kind of

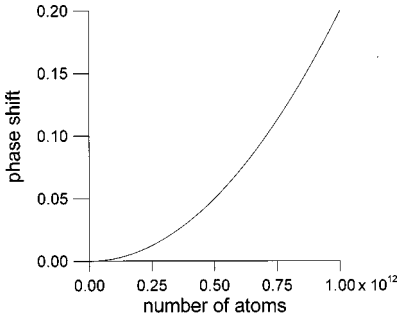

FIG. 13. Nonlinear phase shift as a function of the number of atoms in the medium for the case of collisions with a buffer gas.

asymmetry appears to be related to the fact that the contribution from the exchange process shown in Fig. 2 will have the opposite sign if photon 2 is absorbed first instead of photon 1; in a perturbation theory treatment, the lack of energy conservation in the second intermediate state is equal in magnitude but opposite in sign, which causes the contributions from these two diagrams to cancel in the limit of large detunings [17,41]. It is possible to give examples of various ''gedanken experiments'' in which there would be an apparent violation of causality if this kind of cancellation did not occur for two distant atoms.

APPENDIX C: EQUAL DETUNINGS

It was suggested during the review of this paper that the crucial factor of 2 in Eq. (41) could be derived in a very simple way if $\delta_1=\delta_2$. In that case, there are two linear combinations of states that are not coupled to the initial state by the Hamiltonian and are therefore unexcited or ''dark.'' To see this, we introduce a new set of basis states defined by

$$
\begin{aligned}
|1\rangle &= |\gamma_1,\gamma_2\rangle,\\
|2\rangle &= (|\gamma_1\rangle+|\gamma_2\rangle)/\sqrt{2},\\
|3\rangle &= |0\rangle,\\
|4\rangle &= (|\gamma_1,\gamma_1\rangle+|\gamma_2,\gamma_2\rangle)/\sqrt{2},\\
|5\rangle &= (|\gamma_1\rangle-|\gamma_2\rangle)/\sqrt{2},\\
|6\rangle &= (|\gamma_1,\gamma_1\rangle-|\gamma_2,\gamma_2\rangle)/\sqrt{2}.
\end{aligned}
\tag{C1}
$$

States $|5\rangle$ and $|6\rangle$ are then uncoupled from the other four states and can be neglected:

$$
\langle i|H_{\rm eff}|j\rangle = 0 \quad (\text{for } i>4,\ j\leq 4). \tag{C2}
$$

This gives a four-dimensional Hamiltonian in this basis for the remaining states, which is given by

$$
H_{\rm eff}=\begin{bmatrix} 0 & \sqrt{2}M' & 0 & 0\\ \sqrt{2}M' & -\delta & 2M' & \sqrt{2}M'\\ 0 & 2M' & -2\delta & 0\\ 0 & \sqrt{2}M' & 0 & 0 \end{bmatrix} \tag{C3}
$$

in the limit of large N, where $M'=\sqrt{N}M$. The coupling between these four states is illustrated in Fig. 14.

To lowest order in perturbation theory, the probability amplitude of state $|3\rangle$, in which there are two excited atoms, is given by

$$
A_2=\frac{\langle 3|H_{\rm eff}|2\rangle\langle 2|H_{\rm eff}|1\rangle}{(e_1-e_3)(e_1-e_2)}, \tag{C4}
$$

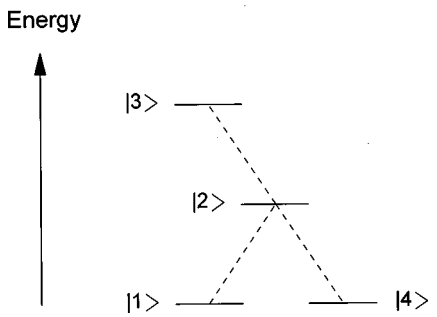

FIG. 14. Four-state system for the case of equal detunings ($\delta_1=\delta_2$). The dashed lines represent the coupling of the states by the Hamiltonian. The remaining two states are not coupled to the initial state and can be ignored.

where e_1, e_2, and e_3 are the unperturbed energies of the states. Using the matrix elements from Eq. (C3) gives the probability of there being two excited atoms when both photons pass through the same medium:

$$
P_{2S}=A_2^2=2\frac{M'^4}{\delta^4}. \tag{C5}
$$

In contrast, perturbation theory applied to the Hamiltonians of Eqs. (33) and (55) gives

$$
P_{2D}=\frac{M'^4}{\delta^4} \tag{C6}
$$

for the probability of there being two excited atoms when the two photons pass through two different media. A comparison of Eqs. (C5) and (C6) gives the factor of 2 in Eq. (41). Since the characteristic equation associated with this eigenvalue problem corresponds to a fourth-order polynomial, explicit expressions for the exact eigenvalues and eigenvectors exist but are sufficiently lengthy that they have not been included here.

For the more general case in which $\delta_1\neq\delta_2$, there is no linear combination of states that are uncoupled in this way and the dimension of the Hamiltonian cannot be reduced as a result. This can be shown by considering a sequence of state vectors in which each successive vector is generated by letting the Hamiltonian act on the previous state in the sequence, with the first state taken to be the initial state $|\gamma_1,\gamma_2\rangle$, and then subtracting off the projection of that state onto each of the preceding states. For $\delta_1=\delta_2$, the fifth vector in the sequence is zero after subtracting its projection onto the previous four states, which shows that two states are uncoupled; the fifth and sixth vectors in the sequence are nonzero for $\delta_1\neq\delta_2$.

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