Cooperative Enhancement of Optical Quantum Gates

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Nonlinear phase shifts at the two-photon level can be greatly enhanced by a cooperative effect involving pairs of atoms, which can give a phase shift proportional to the square of the number of atoms in a medium. This is a nonlocal quantum effect that does not exist for classical states of light. Cooperative effects of this kind may have practical applications in quantum computing. [S0031-9007(97)03199-2]

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High-intensity fields are typically required to produce any significant nonlinear effects [1,2] in optics, whereas the electric field associated with a single photon is normally very weak. This poses a major challenge for the development of an optical approach [3,4] to quantum computing [5–7], which would require nonlinear interactions at the two-photon level. Although relatively large fields can be obtained by confining a single photon to a small cavity, that approach may not be feasible for the construction of large-scale computers due to the size and cost of the high-Q cavities [3] required. This Letter describes a cooperative mechanism involving pairs of atoms that can give a nonlinear phase shift proportional to the square of the number N_A of atoms in a medium. For large values of N_A, the resulting enhancement in the nonlinear phase shift is expected to be of practical use in optical quantum gates. These nonlinear phase shifts are also of fundamental interest, since they are nonlocal quantum effects that do not exist for classical states of light.

Most mechanisms [1–3,8,9] for the production of nonlinear phase shifts involve the interaction of two photons with the same atom, which gives a phase shift proportional to the number of atoms in the medium. In the cooperative mechanism of interest here, two photons interact with each pair of atoms in a medium, such as those labeled A and B in Fig. 1. A typical interaction is illustrated by the Feynman-like diagram of Fig. 2, in which atom A absorbs photon 1 and reemits a photon with frequency ω_2, after which atom B absorbs a photon with frequency ω_2 and reemits photon 1. This interchange of the two photons has no net effect other than to produce a shift in the energy of the system that can be calculated using fourth-order perturbation theory. For simplicity, it will be assumed that the medium is an atomic vapor cell, although the theory applies equally well to a solid or liquid for large detunings. It will be found that there is a large probability that both atoms will be left in the same momentum state that they occupied initially, in which case there are on the order of N_A^2 different Feynman diagrams leading to the same final state, one set for each pair of atoms. Under the appropriate experimental conditions, the contribution from each pair of atoms will have the same phase and the total energy shift will be proportional to N_A^2.

In the dipole approximation [10,11], the interaction Hamiltonian H' is given by

$$H' = -q \sum_i \mathbf{r}_i \cdot \hat{\mathbf{E}}(\mathbf{R}_i), \tag{1}$$

where q is the charge of the electron, $\mathbf{r}_i$ is the location of an electron relative to the center-of-mass coordinate $\mathbf{R}_i$ of atom i, and $\hat{\mathbf{E}}(\mathbf{R}_i)$ is the electric field operator at position $\mathbf{R}_i$. Steady-state perturbation theory [10] gives a fourth-order energy shift of

$$\Delta E = \sum\nolimits'_{lmn} \frac{\langle 0|H'|n\rangle\langle n|H'|m\rangle\langle m|H'|l\rangle\langle l|H'|0\rangle}{(\epsilon_0 - \epsilon_n)(\epsilon_0 - \epsilon_m)(\epsilon_0 - \epsilon_1)} - \sum\nolimits'_{\ln} \frac{\langle 0|H'|n\rangle\langle n|H'|0\rangle\langle 0|H'|l\rangle\langle l|H'|0\rangle}{(\epsilon_0 - \epsilon_n)(\epsilon_0 - \epsilon_1)^2} \tag{2}$$

where $|0\rangle$ is the initial state with energy ϵ_0 while $|l\rangle$, $|m\rangle$, and $|n\rangle$ are a complete set of intermediate eigenstates with energies ϵ_1, ϵ_m, and ϵ_n. The same results [12] can also be obtained using time-dependent perturbation theory in analogy with the usual forward-scattering amplitude approach [10]. The phase shift $\Delta\phi$ can be related to the energy shift ΔE by

$$\Delta\phi = -\int \Delta E(t)dt/\hbar \doteq -\Delta E \Delta t/\hbar, \tag{3}$$

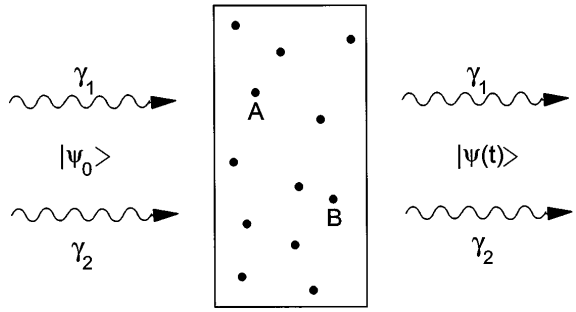

FIG. 1. The geometry of interest, in which two photons are incident upon a medium containing N_A atoms, two of which are labeled A and B.

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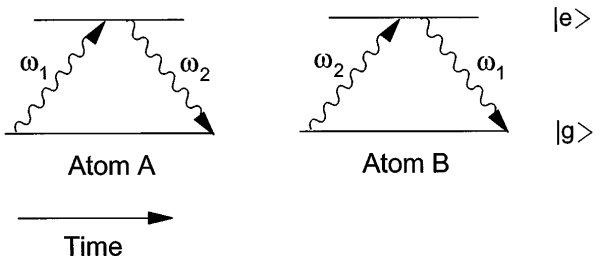

FIG. 2. A Feynman-like diagram in which two atoms exchange two photons, which can give a nonlinear phase shift proportional to the number of pairs of atoms.

where Δt is the interaction time. Equivalent results can be obtained by calculating the change in the dispersion relation giving the wave number $k(\omega)$ as a function of ω.

The center-of-mass contribution to Eq. (2) can be written as a factor f_R given by

$$f_R = \sum_{l_c,m_c,n_c} \langle 0|e^{-i\mathbf{k}_1\cdot\mathbf{R}_B}|n_c\rangle\langle n_c|e^{i\mathbf{k}_2\cdot\mathbf{R}_B}|m_c\rangle \times \langle m_c|e^{-i\mathbf{k}_2\cdot\mathbf{R}_A}|l_c\rangle\langle l_c|e^{i\mathbf{k}_1\cdot\mathbf{R}_A}|0\rangle . \quad (4)$$

Here $|l_c\rangle$, $|m_c\rangle$, and $|n_c\rangle$ represent the atomic center-of-mass intermediate states and it has been assumed that the recoil energy is sufficiently small that it has no significant effect on the total energy of the intermediate states. These eigenstates are a complete set regardless of their form, which allows Eq. (4) to be rewritten as

$$f_R = \langle 0|\cos[(\mathbf{k}_1 - \mathbf{k}_2)\cdot(\mathbf{R}_A - \mathbf{R}_B)]|0\rangle . \quad (5)$$

The fact that the sum over intermediate states includes each pair of atoms (A, B) relabeled in the opposite order (B, A) has also been used. If the medium is sufficiently thin that

$$(\mathbf{k}_1 - \mathbf{k}_2)\cdot(\mathbf{R}_A - \mathbf{R}_B) \ll 1 , \quad (6)$$

then the recoil factor f_R is on the order of unity. The recoil factor f_R is the probability amplitude that the final atomic momentum states will be exactly the same as the initial states, in which case there will be no loss of coherence due to entanglement of the photons with the recoil momentum of the atoms; this is somewhat analogous to the recoilless γ-ray emission of the Mössbauer effect [13]. The remaining events in which there is a change in the atomic momentum states correspond to incoherent scattering events that occur at a rate proportional to N_A and not N_A^2. As a result, the fraction of incoherent events due to atomic recoil is expected to be insignificant in the limit of large N_A.

The process shown in Fig. 2 can occur with either photon being absorbed first and with either atom making the first transition. This corresponds to four distinct diagrams whose total contribution to the energy shift is given by

$$\Delta E_A = M^4 N_A^2 f_R \left[\frac{n_1(n_2 + 1)}{\delta_1^2(\delta_1 - \delta_2)} + \frac{(n_1 + 1)n_2}{\delta_2^2(\delta_2 - \delta_1)} \right] . \quad (7)$$

Here n_1 and n_2 are the number of photons with frequencies ω_1 and ω_2, respectively, and the detunings δ_1 and δ_2 are defined by

$$\delta_1 = \hbar\omega_1 - E_a \quad \delta_2 = \hbar\omega_2 - E_a , \quad (8)$$

where E_a is the atomic excitation energy. The detunings are assumed to be much larger than the Doppler shift, in which case δ_1 and δ_2 are essentially the same for all of the atoms and the Doppler shifts can be ignored. The matrix element M is the same for all four transitions and is given by

$$M = q\mathcal{E}\langle r\rangle , \quad (9)$$

where $\mathcal{E}$ is the magnitude (variance) of the electric field associated with a single photon while $\langle r\rangle = |\langle e|r|g\rangle|$ is the matrix element of the displacement of the electron. For a single-mode cavity, $\mathcal{E}$ is given by

$$\mathcal{E} = |\langle n = 0|\hat{E}|n = 1\rangle| \sim \sqrt{\frac{\hbar\omega}{2\epsilon V}} , \quad (10)$$

where ϵ is the permittivity of free space and V is the volume of the cavity, while $\mathcal{E}^2 = \langle E^- E^+\rangle$ for the case of localized single-photon wave packets.

An additional set of Feynman-like diagrams in which both photons are absorbed before either is reemitted is illustrated in Fig. 3, while Fig. 4 shows a set of diagrams in which each atom absorbs and reemits the same photon. Including all of the possible sequences of the events, these two sets of diagrams contribute energy shifts given by

$$\Delta E_B = M^4 N_A^2 (1 + f_R) n_1 n_2 \times \left[\frac{1}{\delta_1(\delta_1 + \delta_2)\delta_2} + \frac{1}{\delta_1(\delta_1 + \delta_2)\delta_1} + \frac{1}{\delta_2(\delta_1 + \delta_2)\delta_2} + \frac{1}{\delta_2(\delta_1 + \delta_2)\delta_1} \right] . \quad (11)$$

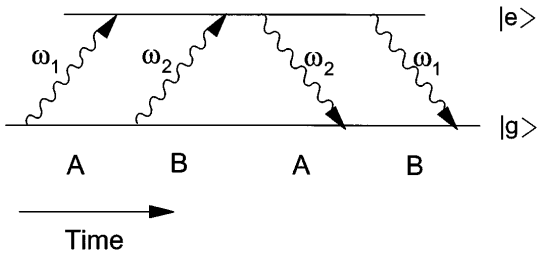

FIG. 3. Additional Feynman-like diagrams in which both photons are absorbed before either is reemitted.

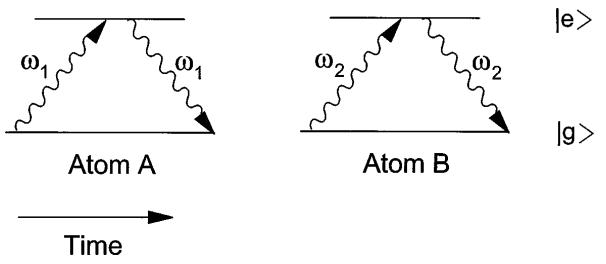

FIG. 4. Additional Feynman-like diagrams in which each atom absorbs and reemits the same photon.

$$\Delta E_C = -M^4 N_A^2 n_1 n_2 \left[\frac{1}{\delta_1^2 \delta_2} + \frac{1}{\delta_2^2 \delta_1} \right]. \quad (12)$$

Higher-order perturbation calculations can be susceptible to various errors, such as the omission of a diagram. As a precaution, the total contribution from all of the diagrams was calculated manually and then verified by a separate calculation that utilized a rule-based symbolic algorithm coded in Mathematica. The results obtained from perturbation theory were also compared with numerical calculations. All three methods gave the same results.

The total energy shift from all of the above diagrams can be reduced to

$$\Delta E = M^4 N_A^2 f_R \frac{n_1 \delta_2^2 - n_2 \delta_1^2}{\delta_1^2 \delta_2^2 (\delta_1 - \delta_2)}. \quad (13)$$

This result may seem somewhat surprising, since it does not contain any nonlinear terms proportional to $n_1 n_2$. All of the individual diagrams contain such terms but they all cancel completely.

This cancellation between the diagrams can be avoided by adding a buffer gas to increase the rate of collisions, which affects the different Feynman diagrams by different amounts. To see why this is the case, suppose that $|\delta_1 - \delta_2| \ll |\delta|$, where $\delta = \delta_1 \sim \delta_2$, and consider the amount of time Δt_{virt} that the system spends in the various virtual (intermediate) states. From the Heisenberg uncertainty relation

$$\Delta t_{\text{virt}} \sim \frac{\hbar}{\Delta E_{\text{virt}}}, \quad (14)$$

where ΔE_{virt} is the lack of energy conservation in a particular virtual state. As a result, the system spends a relatively large amount of time in the second intermediate state of Fig. 2, where $\Delta E_{\text{virt}} = |\delta_1 - \delta_2| \ll |\delta|$, and is much more likely to undergo a collision during that time than is the case for the diagrams of Fig. 3.

The effects of collisions can be estimated by assuming that each of the virtual states has a finite lifetime due to collisions, which is equivalent to replacing the energy ϵ_m of each intermediate state with a complex energy $\epsilon_m - iw$, where w is equal to the spectral linewidth due to collisions. Making this substitution in Eqs. (7), (11), and (12) gives a total energy shift of

$$\Delta E = -\frac{2M^4 N_A^2 n_1 n_2 f_R}{\delta^3} \frac{w^2}{(\delta_1 - \delta_2)^2} \quad (15)$$

to lowest order [14] in $w/(\delta_1 - \delta_2)$, where the nonlinear term proportional to $n_1 n_2$ is the only one that has been retained. It can be seen that the nonlinear phase shift is proportional to a collision factor f_C defined by

$$f_C = \frac{w^2}{(\delta_1 - \delta_2)^2}. \quad (16)$$

Experiments can be designed in such a way that $f_C \sim 1$, in which case the collisions effectively eliminate the contribution from one set of diagrams while having little effect on the others. It should be noted that large values of the detuning itself ($|\delta| \gg w$) are still possible under these conditions and that a very wide range of collision times can be obtained by an appropriate choice of solids, liquids, or gases.

The effects of collisions can be rigorously calculated by including the buffer gas atoms and their scattering Hamiltonian as part of the system and doing perturbation theory to higher (sixth) order [15]. The results obtained have the same form as those of Eq. (15), where the value of the parameter w depends on the details of the collision process and the detuning. Higher-order perturbation theory shows that there can be a large contribution from virtual collision processes that do not conserve energy and are temporary in the sense that the system quickly returns to its original state. It should be noted that these virtual collision processes cannot be included in a master-equation approach in which the buffer gas is considered to be part of the "environment" whose effects are assumed to be irreversible and represented by various decay rates for the density matrix of the "system."

This dependence on collisions is somewhat similar to the collision-induced resonances observed in four-wave mixing, where the probability amplitudes for certain processes are zero in the absence of collisions but give rise to extremely narrow (subnatural) linewidths at high pressures [16]. Collisions need not introduce large amounts of decoherence or random phase fluctuations. For example, consider the usual linear phase shift (index of refraction) of visible light in air or a transparent solid such as glass. Although there are a large number of collisions taking place, there is also a large probability amplitude for the virtual processes responsible for the change in the index of refraction to occur without a collision process. The remaining processes are incoherent and correspond to a relatively small scattering rate, since the incoherent processes have a stronger dependence on the detuning than does the coherent phase shift. Phase fluctuations due to the random nature of the collision process are not significant since the observed phase is an average over a macroscopically large number of atoms.

A similar situation is expected for the nonlinear phase shifts of interest here in the limit of large N_A and large detunings.

The magnitude $\Delta\phi_{\rm non}$ of the nonlinear phase shift can be put in perspective by comparing it to the magnitude $\Delta\phi_{\rm lin}$ of the usual linear phase shift, which can be calculated [10] from the second-order energy shift:

$$\Delta E^{(2)} = \sum_l \frac{\langle 0|H'|l\rangle\langle l|H'|0\rangle}{\epsilon_0 - \epsilon_1} = M^2\frac{N_A}{\delta}. \qquad (17)$$

Consider the ratio R defined by

$$R = \left| \frac{\Delta\phi_{\rm non}}{(\Delta\phi_{\rm lin})^2} \right| = 2\frac{\hbar}{\delta\Delta t} f_R f_C = \frac{2}{\delta\omega\Delta t} f_R f_C, \qquad (18)$$

where $\delta\omega$ is the detuning expressed as an angular frequency. Experiments can be designed [15] in such a way that $f_R \sim 1$, $f_C \sim 1$, and $\delta\omega\Delta t \sim 1$, in which case

$$\Delta\phi_{\rm non} \sim \Delta\phi_{\rm lin}^2. \qquad (19)$$

If N_A is sufficiently large that $\Delta\phi_{\rm lin}$ is equal to 1 rad, then $\Delta\phi_{\rm non}$ will also be on the order of 1 rad, which is sufficient for the implementation of quantum logic gates. An approximate calculation suggests that a nonlinear phase shift of π rad can be obtained from a 1 cm path length through a typical atomic vapor cell with a detuning of 10 GHz; much larger detunings are possible for crystalline materials.

An interesting feature of this effect is the fact that energy is not conserved at the location of atom A nor at atom B but it is conserved globally, which requires nonlocal correlations between the effects of the two atoms. It can be shown that nonlinear phase shifts of this kind cannot be derived from the commonly used assumption [1,2] that the polarization of the medium is proportional to a set of nonlinear susceptibility coefficients ($\chi^{(3)}$); the local nature of the polarizations induced in this way is inconsistent with the nonlocal nature of the effect. It can also be shown that nonlinear phase shifts of this kind can be produced only by photon number (Fock) states, while high-intensity coherent states will produce a small amount of incoherent scattering instead.

Some of the potential advantages of an optical approach for quantum computing based on this effect have been described previously [4]. The increased magnitude of the phase shift can be used to relax the requirements on the system and may eliminate the need for high-Q cavities [3], atomic beams, etc., none of which appear to be well suited for the construction of a full-scale quantum computer. More importantly, it should be possible to use large detunings to avoid the decoherence associated with scattering [17] and absorption, which is one of the main concerns in quantum computing. This approach may eventually allow the construction of large numbers of quantum gates on a single substrate using optical waveguides and microfabrication techniques.

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[12] The correct form of the second term in Eq. (2) will not be obtained from time-dependent perturbation theory unless the interaction is slowly turned on and then off again.
[13] For example, see H. Frauenfelder, *The Mossbauer Effect* (Benjamin, New York, 1962).
[14] The expansion of Eq. (15) can be replaced with a lengthy but exact expression when $w/(\delta_1 - \delta_2)$ is no longer small.
[15] A detailed discussion of the higher-order collision calculations and the design of an appropriate experiment are too lengthy for inclusion here and will be submitted for publication elsewhere.
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[17] The Kramers-Kronig dispersion relations require that any increase in the phase shift must be associated with an increase in the total scattering rate at other frequencies. The fourth-order diagrams discussed here do give an increase in the scattering rate, as required, but those losses can be made insignificant compared to the phase shift by using large detunings, just as in the linear case.