Photon Switching by Quantum Interference

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We describe a four-state atomic system that absorbs two photons, but does not absorb one photon. As a switch, in the ideal limit, it operates at an energy cost of one photon per switching event. [S0031-9007(98)07421-3]

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This Letter describes a four-state atomic system that will absorb two photons but will not absorb one photon. This occurs as a result of a quantum interference that prohibits single-photon absorption. The system may function as an optical switch where a pulse of light of one frequency will cause the absorption of light at a second frequency. Because the quantum interference is fragile, the switch is sensitive. In the ideal case, a pulse of energy equal to that of a single photon is sufficient to open and close the second channel.

An energy schematic of the four-state atomic system is shown in Fig. 1. The light pulse which is to be switched on and off is termed as the probe and has angular frequency ω_p. We will take its intensity and pulse length to be sufficiently weak that all of the atoms remain in the ground state $|1\rangle$. The optical field which creates the quantum interference is called the coupling field. It has an angular frequency ω_c and Rabi frequency Ω_c. It is strong, monochromatic, and present for all time. Its intensity determines the bandwidth and speed of the switch but does not affect the switching energy. The frequency of the optical pulse which destroys the quantum interference and thereby causes absorption of probe photons is ω_{24}. For the quantum interference to be sharp, it is required that the $|1\rangle \rightarrow |2\rangle$ transition have a linewidth which is very small as compared to that of the $|1\rangle \rightarrow |3\rangle$ transition.

The suggestion of this Letter is in the class of nonlinear optical processes that utilize electromagnetically induced transparency [1–3]. These processes depend on using quantum interference to cancel the absorption and dispersion of optical resonances to obtain unusually large nonlinear optical susceptibilities [4,5]. In particular, the two-photon nonlinearity which is described here is the absorptive analog of the (reactive) Kerr nonlinearity of Schmidt and Imamoglu *et al.* [6]. It also uses resonances which are of the same nature as those used in the phase conjugating experiments of Hemmer *et al.* [7] and in the nonlinear spectroscopic work of Lukin *et al.* [8]. In related work, Agarwal and Harshawardhan have discussed how interference may be used to enhance or decrease two-photon absorption and to decrease the threshold for optical bistability [9].

Although we are interested in switching one pulsed optical field with another pulsed field, we proceed by first deriving formulas which are strictly valid for monochromatic fields. We then describe conditions for the application of these formulas to the pulsed case. We show that it is the relationship between the power per area which is necessary for switching, and the group velocity delay of the probe pulse, that leads to an invariant switching energy per area equal to that of a single photon per square wavelength.

Working in the interaction representation, the Hamiltonian and dipole moment operators for the four-state system of Fig. 1 are

$$H = -\Delta\omega_c|2\rangle\langle 2| - \Delta\omega_p|3\rangle\langle 3| - \Delta\omega_{24}|4\rangle\langle 4| - \frac{1}{2}\left(\Omega_p|1\rangle\langle 3| + \Omega_c|2\rangle\langle 3| + \Omega_{24}|2\rangle\langle 4| + \text{H.c.}\right), \tag{1a}$$

$$P = \mu_{13}\exp(-j\omega_p t)|1\rangle\langle 3| + \mu_{23}\exp(-j\omega_c t)|2\rangle\langle 3| + \mu_{24}\exp(-j\omega_{24}t)|2\rangle\langle 4| + \text{H.c.} \tag{1b}$$

The detunings from resonance are defined as $\Delta\omega_p = \omega_p - (\omega_3 - \omega_1)$, $\Delta\omega_c = (\omega_p - \omega_c) - (\omega_2 - \omega_1)$, $\Delta\omega_{24} = (\omega_p - \omega_c + \omega_{24}) - (\omega_4 - \omega_1)$, and Ω_c, Ω_p, and Ω_{24} are the respective Rabi frequencies.

To start, we take all fields to be monochromatic and assume that the atoms remain in the ground state for all time.

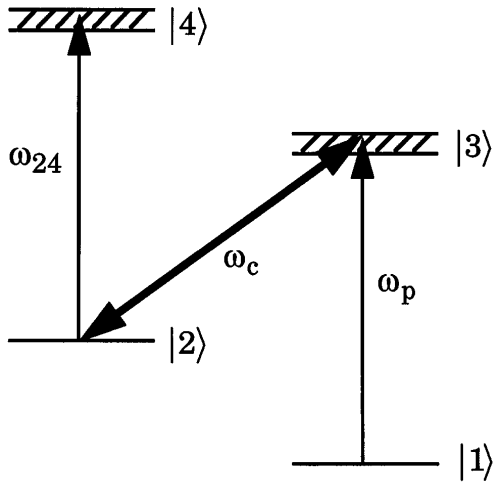

FIG. 1. Energy schematic for a four-state system. The linewidths of states $|3\rangle$ and $|4\rangle$ are greater than all other characteristic frequencies.

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With the probability amplitude $|a_1| = 1$ these assumptions allow a steady-state solution of Schrödinger's equation for the probability amplitudes of states $|2\rangle$ through $|4\rangle$. With these probability amplitudes known, the dipole moment and susceptibility at the probe frequency are calculated. Defining $P(\omega_p) = \epsilon_0 \chi(\omega_p) E(\omega_p)$, the result is

$$\chi^*(\omega_p) = \frac{K(|\Omega_{24}|^2 - 4\Delta\tilde{\omega}_c \Delta\tilde{\omega}_{24})}{4\Delta\tilde{\omega}_p \Delta\tilde{\omega}_c \Delta\tilde{\omega}_{24} - |\Omega_c|^2 \Delta\tilde{\omega}_{24} - |\Omega_{24}|^2 \Delta\tilde{\omega}_p}, \tag{2}$$

where the constant $K = N|\mu_{13}|^2/\hbar\epsilon_0$. By defining the complex detunings $\Delta\tilde{\omega}_p = \Delta\omega_p + j\gamma_{13}$, $\Delta\tilde{\omega}_c = \Delta\omega_c + j\gamma_{12}$, and $\Delta\tilde{\omega}_{24} = \Delta\omega_{24} + j\gamma_{24}$, we introduce (Lorentzian) dephasing linewidths for the respective transitions. The susceptibility of Eq. (2) describes both the resistive nonlinearity of this work, and also, in the appropriate limit, the associated Kerr nonlinearity of Imamoglu [6].

To specialize to the pure resistive nonlinearity we take the linewidths of states $|3\rangle$ and $|4\rangle$ to be large as compared to all other characteristic frequencies, including Ω_c. This assumption causes $\gamma_{23} = \gamma_{13}$ and allows the square of the Rabi frequencies Ω_p, Ω_c, and Ω_{24} to be grouped with the linewidths and replaced by the golden rule transition rates

$$W_p = \frac{\Omega_p^2}{2\gamma_{13}}, \qquad W_c = \frac{\Omega_c^2}{2\gamma_{23}}, \qquad W_{24} = \frac{\Omega_{24}^2}{2\gamma_{24}}. \tag{3}$$

We neglect $\Delta\omega_p$ and $\Delta\omega_{24}$ as compared to their respective linewidths and, because the coupling laser is monochromatic, then let $\Delta\omega_c = \Delta\omega_p$. By combining the susceptibility of Eq. (2) with Maxwell's equations, we obtain expressions for the power loss $2\alpha L$, the phase shift βL, and the group velocity delay time for the probe pulse of frequency ω_p. These are

$$\begin{aligned}
2\alpha L &= NL\sigma_{13}\left[\frac{(W_{24} + 2\gamma_{12})(W_c + W_{24} + 2\gamma_{12}) + 4\Delta\omega_p^2}{(W_c + W_{24} + 2\gamma_{12})^2 + 4\Delta\omega_p^2}\right], \\
\beta L &= NL\sigma_{13}\left[\frac{\Delta\omega_p W_c}{(W_c + W_{24} + 2\gamma_{12})^2 + 4\Delta\omega_p^2}\right], \\
T_D &= \frac{\partial(\beta L)}{\partial\omega} = NL\sigma_{13}\left\{\frac{W_c(W_c + W_{24} + 2\gamma_{12})^2 - 4W_c\Delta\omega_p^2}{[(W_c + W_{24} + 2\gamma_{12})^2 + 4\Delta\omega_p^2]^2}\right\}.
\end{aligned} \tag{4}$$

The group delay time of the probe pulse T_D is relative to the speed of light in vacuum. In Eq. (4) and in the following equations, we write the matrix elements of the $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |4\rangle$ transitions in terms of the cross sections for power absorption σ_{ij} of the respective transitions. These cross sections, in terms of the matrix elements, are $\sigma_{ij} = \omega_{ij}|\mu_{ij}|^2/\epsilon_0 c\hbar\gamma_{ij}$.

We now allow W_{24} and W_p, but not W_c, to be functions of time and ask for the conditions for which Eq. (4) remains valid. There is both a single atom (adiabaticity) and a macroscopic (group velocity delay) condition. To obtain the adiabatic condition, we use the Hamiltonian of Eq. (1) with the decay rates as defined above to write the equations for the probability amplitudes of states $|1\rangle$ through $|4\rangle$. We take the derivatives of the probability amplitudes of states $|3\rangle$ and $|4\rangle$ to vary slowly as compared to the linewidths of these states and drop these derivatives. These equations are

$$\begin{aligned}
\frac{\partial a_1}{\partial t} &= j\frac{\Omega_p}{2} a_3, \\
\frac{\partial a_2}{\partial t} + \gamma_{12} a_2 &= j\frac{\Omega_c}{2} a_3 + j\frac{\Omega_{24}}{2} a_4, \\
a_3 &= \frac{j}{2\gamma_3}[\Omega_p^*(t) a_1 + \Omega_c^* a_2], \\
a_4 &= j\frac{\Omega_{24}^*(t)}{2\gamma_{24}} a_2,
\end{aligned} \tag{5}$$

and, with the previous definitions, combine to

$$\frac{\partial a_2}{\partial t} + \left(\gamma_{12} + \frac{W_c}{2} + \frac{W_{24}}{2}\right) a_2 = -\frac{W_c}{2}\left(\frac{\Omega_p^*}{\Omega_c^*}\right) a_1. \tag{6}$$

The first terms of a series solution of Eq. (6) are

$$\begin{aligned}
a_2(t) = &-\frac{\Omega_p^*(t)}{\Omega_c^*} a_1 + \frac{(2\gamma_{12} + W_{24})}{W_c}\frac{\Omega_p^*(t)}{\Omega_c^*} a_1 \\
&+ \frac{2}{W_c \Omega_c^*}\frac{\partial\Omega_p^*}{\partial t} a_1 + \cdots + \ldots.
\end{aligned} \tag{7}$$

The first term is the solution with monochromatic fields and no dephasing of the $|1\rangle \rightarrow |2\rangle$ transition. The second is the (steady-state) switching term. The last results from the time variation of the probe pulse and leads to the group delay discussed below. For the steady-state solution to apply, this term must be small as compared to the first, i.e.,

$$\frac{1}{\Omega_p^*}\frac{\partial\Omega_p^*}{\partial t} \ll \frac{1}{W_c}. \tag{8}$$

The maximum rate of variation of $\Omega_p(t)$ and therefore the switch bandwidth is set by the (adjustable) golden rule transition rate W_c [10].

There is a more important constraint: The pulses of frequencies ω_{24} and ω_p must not slip apart as they travel through the medium. In a medium where W_c is less

than the Einstein A coefficient, the group velocity of the probe pulse may be many orders of magnitude slower than the velocity of light [11]. Because of the small probe assumption, the switching pulse ω_{24} has a group velocity which is very close to c. For large W_c, the group delay time, relative to the speed of light T_D, of Eq. (4) is

$$T_D = \frac{NL\sigma_{13}}{W_c}. \tag{9}$$

Note that at unity absorption depth, i.e., $\sigma_{13}NL = 1$, the delay time is equal to the minimum pulse width $1/W_c$ and, to be observable, requires that the optical thickness be greater than unity. With cool atoms, where W_c may be at least a factor of 10 smaller than the Einstein A coefficient and with $\sigma_{13}NL = 10$ in a length of 1 mm, a group velocity slower than about $10^{-5}c$ is expected.

We next solve the first of Eqs. (4) for the transition rate $(W_{24})_{\text{crit}}$ and power density $(P_{24}/A)_{\text{crit}}$ at which the power loss at the probe frequency is equal to unity, i.e., $(2\alpha L) = 1$. For W_c large compared to W_{24} and γ_{12}, but small compared to γ_{13}, $(W_{24})_{\text{crit}} = (W_c/NL\sigma_{13})$ and the critical power density is

$$\left(\frac{1}{\hbar\omega_{24}}\right)\left(\frac{P_{24}}{A}\right)_{\text{crit}} = \left(\frac{1}{\sigma_{13}\sigma_{24}}\right)\left(\frac{1}{NL}\right)W_c. \tag{10}$$

The energy per area at frequency ω_{24} which is necessary for switching is the product of the critical power density and the delay time of Eq. (9)

$$\frac{\text{Energy}}{\text{Area}} = \left(\frac{P_{24}}{A}\right)_{\text{crit}} T_D = \frac{\hbar\omega_{24}}{\sigma_{24}}. \tag{11}$$

The cross section of an atom which is naturally broadened and spontaneously decays into a single polarization is $\sigma = 3\lambda^2/(2\pi)$ and, in this case,

$$\frac{\text{Energy}}{\text{Area}} = \left(\frac{2\pi}{3}\right)\frac{\hbar\omega_{24}}{\lambda_{24}^2}. \tag{12}$$

Therefore, in the limit of ideal assumptions, a pulse at frequency ω_{24} with the energy of a single photon and focused to a spot size of a half a wavelength is sufficient to make the medium opaque to a photon of frequency ω_p. In this limit, the duration of the pulse of frequency ω_{24} must be equal to the group delay time T_D. We note that a switch which works by saturation of a two-state atom also has an ideal switching energy of a single photon per square wavelength [12]. But because it works by sequential absorption and decay, it has a characteristic time constant equal to the Einstein A coefficient. Here, the switch may open or close with a speed equal to the golden rule transition rate W_c.

Figures 2(a) and 2(b) show the power transmission $\exp(-2\alpha L)$ and phase shift βL as a function of the detuning $\Delta\omega_p$. The figures are plotted using Eq. (4). Both assume an optical depth $NL\sigma_{13} = 10$. Both take $W_c = 1$ and $\gamma_{12} = 10^{-4}$. In each case, the switch is shown open ($W_{24} = 0$) and closed ($W_{24} = W_{\text{crit}} = 0.1$), such that the transmission is approximately $\exp(-1)$.

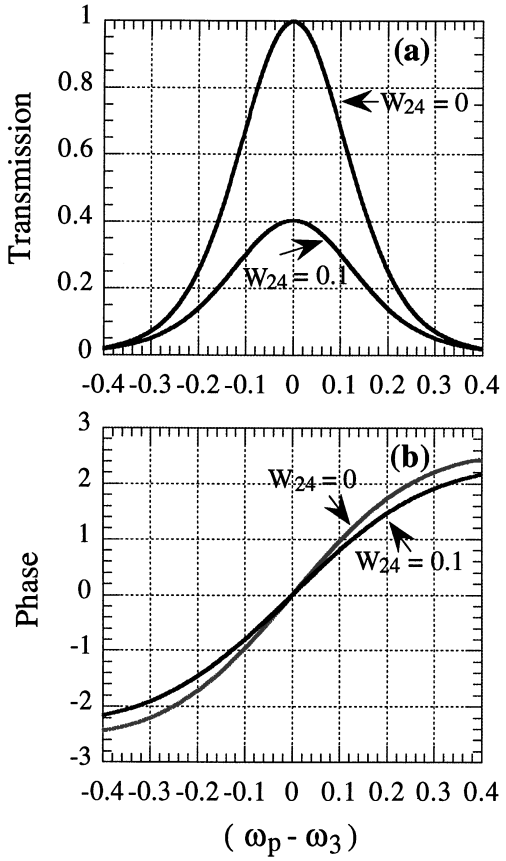

FIG. 2. Power *transmission* and *phase* shift as a function of the detuning $\Delta\omega_p = \Delta\omega_c$. The switch is shown open ($W_{24} = 0$) and closed [$W_{24} = (W_{24})_{\text{crit}} = 0.1$]. The golden rule transition and dephasing rates are $W_c = 1$ and $\gamma_{12} = 10^{-4}$, respectively.

We next examine the reciprocal question: How do photons of frequency ω_p affect the transmission of photons at frequency ω_{24}. Retaining the assumption that W_c is sufficiently strong that $|a_1|^2 = 1$, we calculate the dipole moment and absorption at frequency ω_{24}. Proceeding as above, one finds that the critical power density at frequency ω_p which results in $\alpha L = 1$ at frequency ω_{24} is

$$\left(\frac{1}{\hbar\omega_p}\right)\left(\frac{P_p}{A}\right)_{\text{crit}} = \left(\frac{1}{\sigma_{13}\sigma_{24}}\right)\left(\frac{1}{NL}\right)W_c. \tag{13}$$

Note that the right-hand side is the same as that of Eq. (10). There is exact reciprocity: The number of photons per second which will close one channel is the same as the number which will close the other. In the ideal limit of a single photon in each channel, both are absorbed, while a single photon in either channel alone is not.

For the validity of the assumption $|a_1|^2 = 1$, the product of the total transition rate from state $|1\rangle$ and the time T_D during which the switch is open, must be less than unity. The transition rate from state $|1\rangle$ is

$$W = -\frac{\partial}{\partial t}|a_1|^2 = \left(\frac{2\gamma_{12} + W_{24}}{W_c}\right)\frac{\sigma_{13}\frac{P}{A}}{\hbar\omega_{13}}. \tag{14}$$

For a small dephasing rate γ_{12}, this condition may be

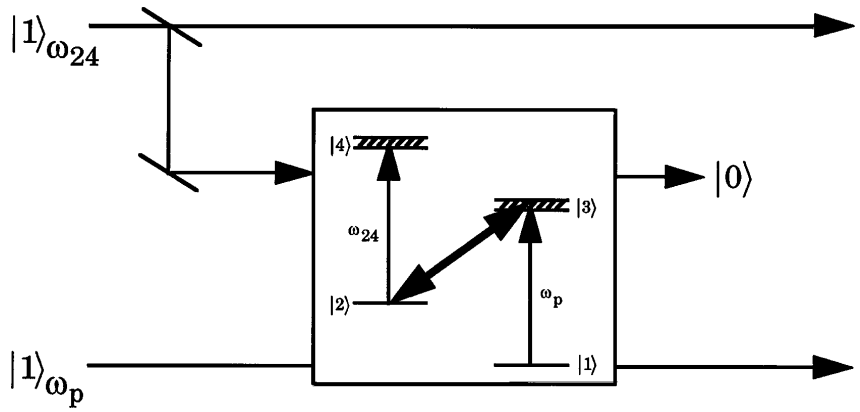

FIG. 3. Formation of a threefold entangled state. The measurables are the presence or absence of photons at frequencies ω_p and ω_{24} and the presence or absence of fluorescence from state $|4\rangle$.

written as

$$W_{24}W_p(N\sigma_{13}L) < W_c^2. \quad (15)$$

Since the two-photon absorption depends on the simultaneous arrival of two photons, it may have application to the processing of quantum entangled states. Suppose that a parametric down-converter is tuned to generate a twin photon pair $|1\rangle_{\omega_p}|1\rangle_{\omega_{24}}$ and one photon, say $|1\rangle_{\omega_{24}}$, is split into the two arms of a 50%-50% beam splitter. If one part of this entangled state interacts with $|1\rangle_{\omega_p}$ in an ideal two-photon absorber (Fig. 3), the simultaneous presence and absence of two-photon absorption generates a threefold state [13].

$$\frac{1}{\sqrt{2}}(|10\rangle + |01\rangle)_{\omega_{24}} \otimes |1\rangle_{\omega_p} \otimes |1\rangle_{\text{atom}}$$

$$\rightarrow \frac{1}{\sqrt{2}}(|00\rangle_{\omega_{24}}|0\rangle_{\omega_p}|4\rangle_{\text{atom}} + |01\rangle_{\omega_{24}}|1\rangle_{\omega_p}|1\rangle_{\text{atom}}). \quad (16)$$

Here $|1\rangle_{\text{atom}}$ denotes the condition where all of the atoms are in the ground state, and $|4\rangle_{\text{atom}}$ denotes a single atom in an excited state and all others in the ground state. The measurables are the presence or absence of a photon in each channel and the presence of absence of fluorescence of state $|4\rangle$ atoms.

The ability to construct this type of interference switch is dependent on a very high ratio of allowed to nonallowed transition linewidths. It is therefore of interest to note that by using a buffer gas to eliminate time of flight broadening, linewidths of hyperfine transitions in Cs vapor as low as 42 Hz have recently been reported [14].

In summary, we have described an optical system which, in the ideal limit, may absorb two photons but not a single photon. It may function as an absorptive optical switch with an adjustable bandwidth and with an invariant switching energy of one photon per atomic cross section. Because it operates by quantum interference, almost all of the atoms remain in the ground state and, except when switched, beams propagate as in vacuum. Since its action depends on the simultaneous arrival of photons, it may also have application to the processing of quantum superposition states.

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