Giant Kerr nonlinearities using refractive-index enhancement

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By utilizing refractive-index enhancement with vanishing absorption, a scheme is suggested that achieves giant Kerr nonlinearities between two weak laser beams. One application of this scheme is discussed and an all-optical distributed Bragg reflector is proposed that works at very low light levels.

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Over the past decade, there has been a growing interest in techniques that achieve significant nonlinear interactions at the single-photon level [1]. These schemes are exciting and important because of both practical and fundamental reasons. Key practical applications include all-optical switches that can operate at an energy cost of a single photon [2] and two-qubit quantum gates between two single photons [3]. From a more basic science perspective, these schemes significantly modify our understanding of light-matter interactions. We now know and are comfortable accepting that light beams can be slowed down to m/s group velocities [4,5], can be brought to a complete stop [6–8], or can even be made to travel backward inside an atomic medium [9].

Perhaps the most promising approach for achieving significant nonlinear interactions at the single-photon level is the technique of electromagnetically induced transparency (EIT) [10]. Since the pioneering work of Imamoglu and colleagues [11], various schemes to achieve single-photon switches and gates using EIT have been proposed [12–14]. EIT relies on quantum interference to create a narrow transmission window in an otherwise highly absorbing and opaque medium. At the point of vanishing absorption, there is a steep variation of the refractive index. This steep dispersion of the index, and therefore the phase accumulation, lies at the heart of all EIT-based proposals. Zhu and colleagues demonstrated EIT-based switches that work at an energy cost of less than one photon per atomic cross section [15]. Recently, significant nonlinear interactions with only several hundreds of photons have been demonstrated by using ultracold atomic clouds inside a hollow-core fiber [16].

In this paper, we suggest an alternative approach to achieve significant nonlinear interactions between single photons. Our approach uses refractive-index enhancement with vanishing absorption to achieve a giant Kerr nonlinearity between two weak laser beams. Before proceeding with a detailed description, we would like to summarize the key achievement of this paper. Let's first consider a laser beam interacting with an ensemble of two level atoms. For this simple case, the third-order nonlinear susceptibility of the medium is proportional to $\chi^{(3)} \sim \frac{1}{|\Delta+j\Gamma|^2(\Delta+j\Gamma)}$, where Γ is the (amplitude) decay rate of the excited state and Δ is the detuning of the laser beam from the transition. This third-order susceptibility is responsible for well-known effects such as intensity-dependent refractive index and optical self-focusing. Now consider two off-resonant beams that are very largely detuned from the excited electronic state interacting with a three-level atomic system in a Λ configuration. If the frequency difference of the two beams is close to the frequency of the Raman transition, the third-order susceptibility is enhanced and is proportional to $\chi^{(3)} \sim \frac{1}{|\Delta+j\Gamma|^2(\delta\omega+j\gamma)}$, where γ is the decay rate of the Raman transition and $\delta\omega$ is the two photon detuning. Typically, to reduce nonlinear absorption, one chooses $\delta\omega \gg \gamma$ such that the imaginary part of the susceptibility is much smaller than the real part: $\mathrm{Im}(\chi^{(3)}) \ll \mathrm{Re}(\chi^{(3)})$. The key achievement of our scheme is to obtain a purely real third-order nonlinear susceptibility of $\chi^{(3)} \sim \frac{1}{|\Delta+j\Gamma|^2\gamma}$ due to destructive interference in the imaginary part. If the decay rate of the Raman transition is negligible ($\gamma \approx 0$), one can therefore obtain an arbitrarily large value for $\chi^{(3)}$ and obtain significant nonlinear interactions between weak beams. The key advantage of our scheme over EIT is that it does not require a strong-coupling laser beam. The key disadvantage is that single-photon linear absorption of the beams is not eliminated. As we will see, this drawback limits the minimum energy required to be on the order of tens of photons per atomic cross section.

Figure 1 shows the energy level diagram of our suggestion. The scheme relies on recently suggested refractive-index enhancement with vanishing absorption that utilizes the interference of two Raman transitions [17,18]. Although the scheme is general, for concreteness, we focus on a real system and consider a ^{87}Rb D_1 line transition with the level structure shown in Fig. 1. We choose the ground state of the system to be $|g\rangle \rightarrow |F=2, m_F=0\rangle$ and the two excited Raman states to be $|1\rangle \rightarrow |F=2, m_F=-2\rangle$ and $|2\rangle \rightarrow |F=2, m_F=2\rangle$. Two off-resonant beams, termed the probe and control beams, couple the ground state $|g\rangle$ to excited Raman states $|1\rangle$ and $|2\rangle$. The two beams have opposite circular polarizations (σ^+ for E_p and σ^- for E_c). The quantities $\delta\omega_1$ and $\delta\omega_2$ are two photon detunings of the laser beams from each Raman transition, respectively, and they are defined as $\delta\omega_1 = (\omega_1 - \omega_g) - (\omega_p - \omega_c)$ and $\delta\omega_2 = (\omega_2 - \omega_g) - (\omega_c - \omega_p)$. The refractive-index enhancement scheme as originally suggested requires two separate control laser beams whose frequencies can be tuned to control the position of the two resonances independently. In the scheme of Fig. 1, however, there is only one control laser. The position of the Raman resonances can be controlled by shifting the hyperfine levels through a combination of a Stark shift and Zeeman shift. The Zeeman shift can simply be provided with a magnetic field pointing along the propagation direction of the laser beams. The Stark shift can either be provided with a dc electric field or a separate detuned laser beam. We define the quantity $\delta \equiv \delta\omega_1 + \delta\omega_2$ to represent the separation of the two Raman resonances as the probe (or the control) laser frequency is scanned.

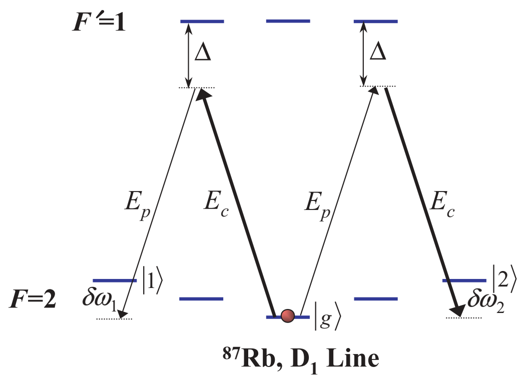

FIG. 1. (Color online) The proposed scheme. For concreteness, we focus on a real atomic system and consider two off-resonant laser beams, E_p and E_c, interacting with ^{87}Rb atoms through the D_1 line. The two laser beams are opposite circularly polarized and couple the ground state $|F=2, m_F=0\rangle$ to two excited Raman states $|F=2, m_F=-2\rangle$ and $|F=2, m_F=2\rangle$. The quantities $\delta\omega_1$ and $\delta\omega_2$ are two photon detunings of the laser beams from each Raman transition, respectively. The positions of the Raman resonances as the probe laser frequency is scanned can be independently controlled by a combination of Zeeman and Stark shifts of the hyperfine states.

We proceed with an analysis of the scheme of Fig. 1. With Δ much larger than the decay width Γ, we can adiabatically eliminate the probability amplitudes of the excited states. We also take the two beams to be weak enough such that the power broadening of the Raman transitions can be ignored. With these assumptions, the polarization of the medium at the two laser frequencies can be written as [17]

$$\begin{aligned} P_p &= \epsilon_0\chi^{(1)}E_p + \epsilon_0\chi^{(3)}|E_c|^2E_p, \\ P_c &= \epsilon_0\chi^{(1)}E_c + \epsilon_0\chi^{(3)*}|E_p|^2E_c, \end{aligned} \tag{1}$$

where $\chi^{(1)}$ and $\chi^{(3)}$ are the linear and nonlinear third-order susceptibilities of the medium, respectively. These quantities are [17]

$$\begin{aligned} \chi^{(1)} &= \frac{N}{\epsilon_0\hbar}\frac{|\mu_{ij}|^2}{\Delta + j\Gamma}, \\ \chi^{(3)} &= \frac{N}{4\epsilon_0\hbar^3}\frac{1}{|\Delta + j\Gamma|^2}\left(\frac{|\mu_{ij}|^2|\mu_{jk}|^2}{\delta\omega_1 - j\gamma_1} + \frac{|\mu_{il}|^2|\mu_{lm}|^2}{\delta\omega_2 + j\gamma_2}\right), \end{aligned} \tag{2}$$

where N is the atomic density, μ_{ij} are the dipole matrix elements between relevant states, Γ is the decay rate of the excited state, and γ_1 and γ_2 are dephasing rates of the Raman transitions, respectively. As expected, the third-order nonlinear susceptibility is a sum of two terms due to two Raman transitions. It is the interference of these two terms that results in the enhancement of the real part while resulting in the vanishing imaginary part. Figure 2 shows the real (solid line) and the imaginary (dashed line) parts of $\chi^{(3)}$ as the probe laser frequency is scanned for three different values of the separation of Raman resonances, $\delta = 10\gamma, 5\gamma$, and γ, respectively. Here, for simplicity, we assume that the two Raman transitions have the same parameters, including an identical Raman linewidth of $\gamma_1 = \gamma_2 \equiv \gamma$. At the midpoint between the two resonances, $\delta\omega_1 = \delta\omega_2$, the imaginary part vanishes while the real part is enhanced due to constructive interference.

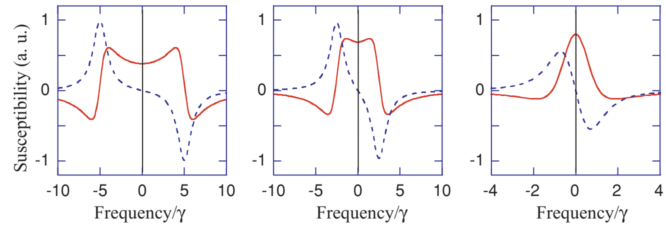

FIG. 2. (Color online) The real (solid line) and imaginary (dashed line) parts of the nonlinear susceptibility as a function of probe laser frequency for three different values of the separation of two Raman resonances, $\delta = 10\gamma$, 5γ, and γ. At the midpoint between the two resonances, the real part is enhanced due to constructive interference whereas the imaginary part vanishes due to destructive interference.

At the point of the vanishing imaginary part, the third-order nonlinear susceptibility is purely real and is given by

$$\chi^{(3)} = \frac{N}{2\epsilon_0\hbar^3}\frac{|\mu_{ij}|^2|\mu_{jk}|^2}{|\Delta + j\Gamma|^2}\frac{\delta/2}{(\delta/2)^2 + \gamma^2}. \tag{3}$$

While deriving Eq. (3), for simplicity, we have again assumed the two Raman resonances to have identical parameters. From Eq. (3), we note that the value of the nonlinear susceptibility strongly depends on the separation of the two Raman resonances, δ. The maximum nonlinear susceptibility is obtained when $\delta = 2\gamma$ and is given by

$$\chi^{(3)}_{\max} = \frac{N}{4\epsilon_0\hbar^3}\frac{|\mu_{ij}|^2|\mu_{jk}|^2}{|\Delta + j\Gamma|^2}\frac{1}{\gamma}. \tag{4}$$

Equation (4) is the central result of this paper. The nonlinear susceptibility is purely real and, in the limit of very long dephasing and decay time of the Raman transitions ($\gamma \approx 0$), it can become arbitrarily large.

From the nonlinear susceptibility of Eq. (4), we can also find the expression for the intensity-dependent refractive index, $n_2 = \eta\chi^{(3)}$, where $\eta = \sqrt{\mu_0/\epsilon_0}$. Before proceeding further, we evaluate the intensity-dependent refractive index for experimentally achievable parameters. We consider an ultracold ^{87}Rb atomic cloud with $N = 10^{14}$ atoms/cm^3. We take $\Delta = 10\Gamma$, where Γ is the decay rate of $5P_{1/2}$, $2\Gamma = 2\pi \times 5.74$ MHz. We assume a Raman transition linewidth of $\gamma = 2\pi \times 10$ kHz. With these modest parameters, we calculate an intensity-dependent refractive index of $n_2 = 75.8$ cm^2/W, which is comparable to what has been achieved in recent EIT experiments.

We proceed with the evaluation of the nonlinear phase shift of the probe laser due to few-photon control laser pulses. For this purpose, we consider a control laser beam pulse of Gaussian temporal shape that contains n_c photons. By using Eqs. (1)–(4), the nonlinear phase accumulation of the probe laser beam and the corresponding linear power absorption coefficient can be derived (for the specific scheme of Fig. 1):

$$\begin{aligned} \phi_{\text{nonlinear}} &= \frac{3}{32\sqrt{\pi}}n_c(N\sigma L)\frac{\Gamma^2}{\Delta^2 + \Gamma^2}\frac{\sigma}{A}\frac{1}{\gamma\tau}, \\ \alpha_{\text{linear}} &= \frac{1}{4}(N\sigma L)\frac{\Gamma^2}{\Delta^2 + \Gamma^2}, \end{aligned} \tag{5}$$

where $\sigma = \lambda^2/2\pi$ is the atomic cross section, A is the transverse area of the beam, and τ is the temporal Gaussian

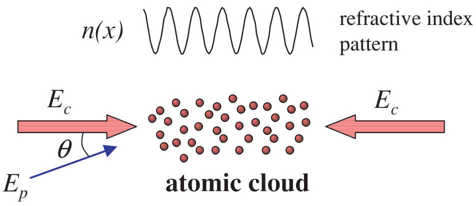

FIG. 3. (Color online) The setup for constructing an all-optical mirror. The counterpropagating control lasers form a standing-wave pattern which results in a periodic variation of the refractive index. As a result, a photonic band gap is formed for the probe laser beam and the probe wave is reflected off the medium.

width of the pulse. While deriving Eq. (5), we have assumed that the excited states are purely lifetime broadened and neglected nonradiative broadening effects such as collisions. As expected, Eq. (5) shows that the nonlinear phase accumulation is intrinsically related to the linear loss. If we assume the ideal case of $\tau \approx 1/\gamma$ and consider tightly focused beams, $A \approx \sigma$, Eq. (5) reduces to

$$\phi_{\text{nonlinear}} = \frac{3}{8\sqrt{\pi}} n_c \alpha_{\text{linear}}. \tag{6}$$

From Eq. (6), if we limit the linear power loss to 50% ($\alpha_{\text{linear}} = 0.7$), a single control photon ($n_c = 1$) per atomic cross section causes a nonlinear phase shift of 0.15 rad. A nonlinear phase shift of π rad would therefore require about 21 control laser photons per atomic cross section.

We next discuss a type of optically controlled optical device using our scheme [19]. Due to the intensity-dependent refractive index, an intensity pattern on the control laser produces a refractive-index pattern for the probe laser beam. By using an appropriate intensity pattern, one can, therefore, engineer an all-optical device. Figure 3 shows a simple scheme where we consider an ultracold atomic cloud interacting with a probe and a counterpropagating pair of control laser beams. Due to the standing-wave intensity pattern of the control laser, the probe wave experiences a periodic variation of the refractive index. Under these conditions, within a certain frequency range, a photonic band gap is produced and the propagation of the probe wave is forbidden inside the medium. Within the photonic band gap, the incident probe laser cannot penetrate the medium and is reflected. The idea of utilizing a periodic variation of the refractive index was motivated by the recent work of Lukin and colleagues, who proposed and experimentally demonstrated optically induced photonic band gaps using EIT and slow light [20,21].

To calculate the reflectivity of such a medium, we use the coupled mode theory of Yariv and Yeh [22]. Figure 4 shows the results of a calculation for an ultracold ^{87}Rb cloud. Here, we choose the medium parameters and the control laser intensity such that the nonlinear index enhancement is $n_2 I_c^{\max} = 10^{-3}$. Due to the standing-wave pattern, the refractive index varies between $n = 1$ and $n = 1 + 10^{-3}$ with a period of $\lambda_c/2 \approx$ 397 nm. We take the atomic cloud to be sufficiently cold (temperature of about 1 μK) such that two-photon Doppler broadening of the Raman transitions can be ignored. This assumption simplifies the problem considerably since the probe beam interacts with both control lasers in the same way.

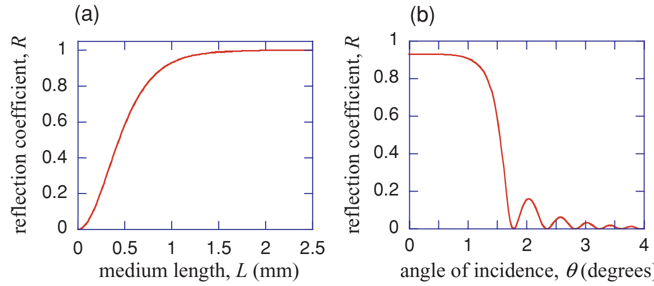

FIG. 4. (Color online) (a) The reflection coefficient for the probe wave, R, as a function of the length of the index-enhanced medium, L, for $n_2 I_c = 10^{-3}$ and $\theta = 0$. The reflection coefficient exceeds 99% for $L = 1.5$ mm. (b) The reflection coefficient as a function of the angle of incidence, θ. The angle of incidence, θ, is defined as the angle between the probe beam and one of the control lasers, as shown in Fig. 3. For $L = 1$ mm, if we set the medium parameters such that there is 50% power loss due to linear absorption ($\alpha_{\text{linear}} = 0.7$), then these results require about $n_c = 13$ control laser photons (in each beam) per atomic cross section.

Figure 4(a) shows the power reflection coefficient, R, for the probe wave as a function of the length of the medium, L. Here, we take the probe wave-propagation direction to coincide with one of the control lasers ($\theta = 0$). For $L = 1.5$ mm, the reflection coefficient exceeds 99%. Figure 4(b) shows the reflection coefficient as a function of the angle of incidence, θ, for a medium length of $L = 1$ mm. The angle of incidence, θ, is defined as the angle between the probe beam and one of the control lasers as shown in Fig. 3. The reflection coefficient remains high for about 1.5° and then drops sharply. For $L = 1$ mm, if we set the medium parameters such that there is 50% power loss due to linear absorption ($\alpha_{\text{linear}} = 0.7$), then the results of Fig. 4 require about $n_c = 13$ control laser photons (in each beam) per atomic cross section.

Before concluding, we would like to draw attention to the recent work of Shapiro and colleagues [23,24] and Gea-Banacloche [25]. By using a multimode quantum-mechanical treatment, these authors argue the impossibility of achieving large nonlinear phase shifts using single-photon wave packets. The key reason is the loss of fidelity due to spontaneous emission. Their results suggest that achieving large nonlinear phase shifts with reliable fidelity would require beam energies at least at the tens of photons level, independent of the specific scheme that is used.

In conclusion, we suggested an alternative approach for achieving a large nonlinear Kerr effect between two weak laser beams while maintaining vanishing nonlinear absorption. Differing from EIT, the linear absorption of the beams is not eliminated and, as a result, high nonlinear phase shifts require beam energies at the level of tens of photons per atomic cross section. The key advantage over EIT is that there is no strong-coupling laser and as a result the total energy requirement is at the level of tens of photons. We have also suggested a type of all-optical distributed Bragg reflector that utilizes periodic variation of the refractive index due to a standing-wave pattern.

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