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Dispersive properties and large Kerr nonlinearities using dipole-induced transparency in a single-sided cavity

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We calculate the dispersive properties of a single-sided cavity coupled to a single dipole. We show that when a field is resonant with the dipole and the Purcell factor exceeds the bare cavity reflection coefficient, the field experiences a π phase shift relative to reflection from a bare cavity. We then show that optically Stark shifting the dipole resonance with a second field creates large Kerr nonlinearities. An approximate expression for the total number of photons needed to create a π phase shift is derived.

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Optical nonlinearities play an important role in quantum optics, quantum information processing, and also in design of practical quantum electronic devices. One of the most commonly encountered nonlinearities is the Kerr effect, which has a large number of applications for optical detection, all-optical switching, and quantum computation [1,2]. The main difficulty in achieving these applications is that conventional materials offer only a very small nonlinear response, which is significantly outweighed by linear absorption. Furthermore, applications in quantum optics and quantum information often require that nonlinearities be created by a small number of photons, or sometimes even a single photon. There are very few situations where one can even approach this regime.

One of the few cases where Kerr nonlinearities with a small number of photons can be observed is in atomic gases using electromagnetically induced transparency (EIT) [3]. The central idea for these schemes was originally presented by Schmidt and Imamoglu [4]. This scheme exploits EIT in a four-state atom where the interaction between two weak pulses is mediated by a strong coupling laser. Due to the large atomic coherence of these systems, cross phase modulation and two-photon absorption can be observed with only a small number of photons [5]. A limitations of the Schmidt-Imamoglu proposal is that the large group velocity mismatch between the two interacting pulses puts a lower limit on the required intensity for a full π phase shift to a few photons per cubic wavelength [6]. This group velocity mismatch problem can in principle be overcome [7,8].

A cavity coupled to a dipole can exhibit similar properties to an atomic three-level system in EIT [9]. When the cavity-dipole system is driven by an external field, the cavity field can destructively interfere in an analogous way to the excited state population of a three-level atom. For this reason, we refer to this effect as dipole induced transparency (DIT). In this paper, we explore the dispersive and nonlinear properties of DIT for a field driving a single-sided cavity, a cavity with one input and one reflected port as shown in Fig. 1, interacting with a resonant dipole. The parameter that characterizes this interaction is the Purcell factor, which is the ratio of the dipole decay rate when coupled to a cavity to the decay rate when decoupled. We show that when the Purcell factor is much larger than one, losses due to cavity leakage and dipole absorption are cancelled. This is a manifestation of destructive interference which inhibits the light from entering the cavity. At the same time, whenever the Purcell factor exceeds the bare cavity reflectivity, the presence of a dipole imposes a zero-phase shift on the reflected field that is resonant with the dipole frequency, whereas a bare cavity would impose a π phase shift.

This change of phase has been previously studied in the regime where the vacuum Rabi frequency of the dipole, denoted g, exceeded the cavity and dipole decay rates [10]. In this regime, which we refer to as the high-Q regime, the cavity mode is fully split into a lower and upper polariton (normal mode splitting). An important point of this paper is that one does not need the high-Q regime to observe this π phase shift. The shift is achieved any time the Purcell factor exceeds the bare cavity reflectivity. In the "bad cavity" limit, defined as the limit where the dipole lifetime is much longer than the cavity lifetime [11], this condition can be achieved for much smaller values of g. We show that the large dipole-induced phase shifts in DIT are sharply peaked near the resonant frequency of the emitter, creating a highly dispersive region with large group delays. At this same region, absorption due to cavity losses and dipole decay are cancelled out. Thus, DIT provides a special condition where we can drive the dipole on resonance and create large phase shifts while not suffering from absorption.

In the second part of the paper, we show that these cavity reflection properties can be used to create large Kerr nonlinearities. Nonlinearities using cavity quantum electrodynamics have been theoretically studied [12], and cross phase modulation angles as large as 16° have been experimentally demonstrated [13]. These cross phase modulation angles

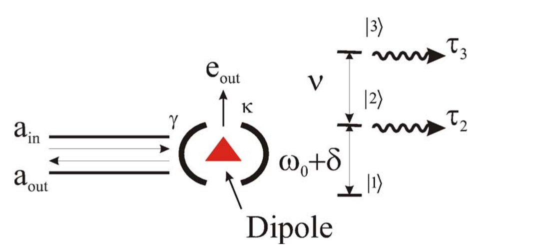

FIG. 1. (Color online) Setup for large Kerr nonlinearities off of reflection from a cavity coupled to a single dipole.

were limited because of the need to detune the field from the resonant frequency of the dipole in order to reduce intracavity losses. Here, we show that by using DIT in a single-sided configuration, one can resonantly drive the dipole to induce large phase shifts without suffering from intracavity losses. We show that with presently achievable cavity lifetimes and vacuum Rabi frequencies, a single cavity photon can achieve a full nonlinear π phase shift using only a single emitter. In contrast to the Schmidt-Imamoglu proposal [4] using EIT without a cavity, our proposal only requires a three-level system instead of a four-level system. Furthermore, the coupling is naturally achieved by cavity-dipole interaction, alleviating the need for a third coupling laser.

The system we study is shown in Fig. 1. An external waveguide field is reflected off of a single-sided cavity containing a dipole. We define $\hat{\mathbf{a}}_{\text{in}}$ and $\hat{\mathbf{a}}_{\text{out}}$ as the flux operators for the input and reflected field, while $\hat{\mathbf{e}}_{\text{out}}$ is the flux operator for parasitic leaky modes. We define γ as the energy coupling rate from cavity to waveguide, while κ is the energy coupling rate into leaky modes. The dipole has three states, denoted $|1\rangle$, $|2\rangle$, and $|3\rangle$, with transition frequencies $\omega_0+\delta$ and ν, where ω_0 denotes the central frequency of the cavity, and cavity coupling strengths g_1 and g_2. We assume that $\delta, \omega_0-\nu < \gamma+\kappa$, so that both transitions can couple to the cavity mode. The lifetime of states $|2\rangle$ and $|3\rangle$ are given by τ_2 and τ_3, respectively. In the first part of the paper, we only consider the 1-2 transition to calculate the dispersive properties of the cavity in the presence of a dipole. We set $g_2=0$, and the state $|3\rangle$ plays no role in these calculations. The system is simply described by a cavity coupled to a two-level dipole, which is equivalent to the system investigated by Duan and Kimble [10]. In the second part of the paper, we add level $|3\rangle$ into the system. Three-level atomic systems have been extensively studied for applications, such as cavity EIT [14] and light storage [15]. In the system considered here, the 2-3 transition does not drive the cavity, and is used only to create an optical Stark shift on state $|2\rangle$.

We denote σ_-^{12} and σ_-^{23} as the lowering operators for the dipole, and $\hat{\mathbf{b}}$ as the bosonic annihilation operator for the cavity field. The Hamiltonian of the system is given by

$$H = H_s + \sum_{n=1}^{N} \hbar \,[g_1 \hat{\mathbf{b}}^\dagger \sigma_-^{12} + g_2(\hat{\mathbf{b}} + \hat{\mathbf{b}}^\dagger)\sigma_-^{23} + \text{H.c.}], \qquad (1)$$

where g_1 and g_2 are the vacuum Rabi frequencies for the 1-2 and 2-3 transitions, respectively. H_s is the Hamiltonian of the uncoupled systems and the cavity-waveguide interaction terms. In order to properly derive the Stark shift term, we do not yet make the rotating wave approximation for the interaction between the cavity and σ_-^{23}.

Using the above Hamiltonian, along with standard cavity input-output formalism [16], we can derive the Heisenberg picture equations of motion for the operators in the "weak excitation limit," where the dipole is predominantly in state $|1\rangle$. In this limit, we can assume $\sigma_z^{12} \approx -1$, and σ_z^{23} is very small and time invariant. The condition for this assumption to be valid is given by $\langle \sigma_+ \sigma_- \rangle \ll 1$, which is equivalent to the condition $\langle \hat{\mathbf{a}}_{in}^\dagger \hat{\mathbf{a}}_{in} \rangle \ll g_1^2/\gamma$ for an input field that is resonant

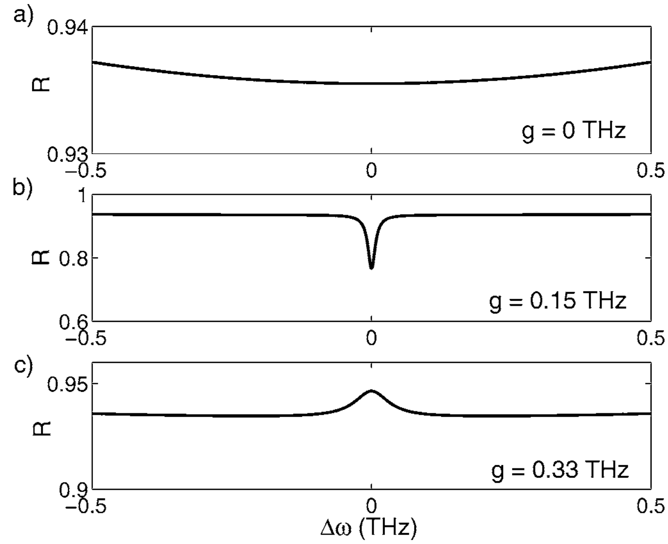

FIG. 2. Cavity reflection coefficient for different values of g: (a) $g=0.3$ THz, (b) $g=0.15$ THz, (c) $g=0.33$ THz.

with the dipole [9] (this condition is well satisfied in our regime of interest). Using this approximation, the operator equations of motion are given by

$$\frac{d\hat{\mathbf{b}}}{dt} = (-i\omega_0 - \gamma/2 - \kappa/2)\hat{\mathbf{b}} - \sqrt{\gamma}\hat{\mathbf{a}}_{\text{in}} - \sqrt{\kappa}\hat{\mathbf{e}}_{\text{in}} - ig_1\sigma_-^{12}, \qquad (2)$$

$$\frac{d\sigma_-^{12}}{dt} = \left(-i(\omega_0+\delta) - \frac{1}{2\tau_2}\right)\sigma_-^{12} - ig_1\hat{\mathbf{b}} - ig_2(\hat{\mathbf{b}}+\hat{\mathbf{b}}^\dagger)\sigma_-^{12}\sigma_-^{23} + \hat{\mathbf{f}} \qquad (3)$$

$$\frac{d\sigma_-^{23}}{dt} = \left(-i\nu - \frac{1}{2\tau_3}\right)\sigma_-^{23} - ig_2(\hat{\mathbf{b}}+\hat{\mathbf{b}}^\dagger)\sigma_z^{23} + \hat{\mathbf{h}}. \qquad (4)$$

The operators $\hat{\mathbf{f}}$ and $\hat{\mathbf{h}}$ are noise operators that are needed to conserve the commutation relation of the dipole operators. The input and output fields are related by $\hat{\mathbf{a}}_{\text{out}} - \hat{\mathbf{a}}_{\text{in}} = \sqrt{\gamma}\hat{\mathbf{b}}$.

We begin by first studying the dispersive properties of reflected light with only states $|1\rangle$ and $|2\rangle$, and set $g_2=0$. In this case, if we apply an input field at frequency ω, the reflection coefficient can be solved in the frequency domain using Eqs. (2) and (3) with the correct input-output relationships to give

$$r(\omega) = \frac{-i\Delta\omega + \dfrac{g_1^2}{-i(\Delta\omega+\delta) + 1/2\tau_2} - \gamma/2 + \kappa/2}{-i\Delta\omega + \dfrac{g_1^2}{-i(\Delta\omega+\delta) + 1/2\tau_2} + \gamma/2 + \kappa/2}. \qquad (5)$$

We define the amplitude and phase of the reflection coefficient by $r=\sqrt{R}e^{i\Phi_r}$, where R is the reflectivity of the cavity and Φ_r is the phase shift imposed on the reflected field.

Figure 2 plots the cavity reflectivity as a function of detuning from the cavity resonance for several values of g_1. We set $\gamma=6$ THz and $\kappa=0.1$ THz, which is the decay rate of a cavity with a quality factor of $Q=10000$ (here Q is defined as

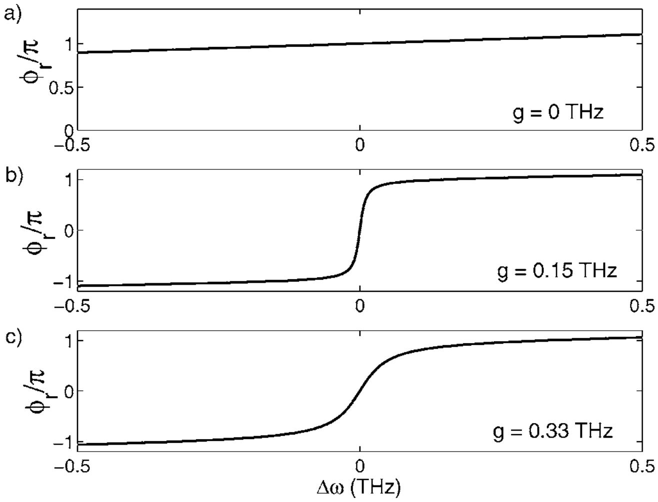

FIG. 3. Phase shift after reflection from cavity, normalized by π, for several values of g: (a) $g=0$ THz, (b) $g=0.15$ THz, (c) $g=0.33$ THz.

$Q=\omega_0/\kappa$). The dipole is assumed to be resonant with the center frequency of the cavity so that $\delta=0$, while $\tau_2=1$ ns, a value taken from experimental measurements on quantum dots [17].

When $g_1=0$ the cavity is not perfectly reflecting due to the coupling to leaky modes as shown in panel (a) of the figure. Introducing a small g_1 increases the cavity loss on resonance, as shown in panel (b), because the dipole behaves as an absorber. However, when g_1 is increased to higher values, the cavity reflection improves. This can be understood from the reflection coefficient at $\Delta\omega=0$, which is given by

$$r(\omega_0) = (F_p - r_0)/(F_p + 1), \tag{6}$$

where $F_p=4\tau_2 g^2/(\gamma+\kappa)$ is the Purcell factor, and $r_0=(\gamma-\kappa)/(\gamma+\kappa)$ is the reflection coefficient for a bare cavity with no dipole. When $F_p\approx r_0$, the cavity is very lossy because most of the field is absorbed by the dipole. However, when $F_p \gg r_0$, the cavity becomes very reflective again, with one significant change. The reflection coefficient changes sign and becomes positive. Thus, there is a π phase shift change between the case $F_p > r_0$ and $F_p < r_0$. In previous work by Duan and Kimble [10], it has been shown that this change in reflection phase can be used to perform a controlled phase gate by performing successive reflections of two photons off of a cavity. In that proposal, the authors considered only the high-Q regime for implementing this phase change. Equation (6) shows that the Duan-Kimble proposal works even in the limit that g is much smaller than the cavity lifetime, provided the Purcell factor exceeds r_0. We need only create sufficiently large values for F_p so that the cavity is not lossy.

We next plot Φ_r, normalized by π, in Fig. 3 for different values of g_1. An important feature of the reflection coefficient is that it is highly dispersive near $\Delta\omega=0$. The slope of the dispersion can be calculated from Eq. (5), and takes on a particularly simple form near zero detuning when $F_p \gg 1$. In this region, we have $\Phi_r=\arctan((\gamma+\kappa/2)\Delta\omega/g_1^2)$. Thus, the phase shift quickly changes to π at a detuning of $\Delta\omega \approx g_1^2/(\gamma+\kappa)$. The slope of Φ_r gives the group delay a pulse experiences from cavity reflection. Near-zero detuning, this group delay is simply $T_g=(\gamma+\kappa)/g_1^2$. The second derivative of Φ_r near zero detuning vanishes, ensuring that the reflected pulse preserves its shape, and is not distorted by the cavity.

The sharp dispersive feature of the reflection coefficient allows the possibility to achieve large Kerr nonlinearity. If one can shift the dispersion curve by a very small amount, on the order of $\Delta\omega \approx g_1^2/(\gamma/2+\kappa/2)$, the reflection phase can be changed from π to 0. The shift in dispersion can be created by optically Stark shifting the 1-2 transition, which can be implemented by adding level $|3\rangle$ to the system, and applying a second off-resonant field on the 2-3 transition.

To calculate the optical Stark shift, we assume that the input field has two frequency components, one at ω and the other at $\nu+\Delta$. The component at $\nu+\Delta$ is the field responsible for creating a Stark shift. The response of the cavity at this frequency is given by substituting $\hat{\mathbf{a}}_{\text{in}}=\hat{\mathbf{a}}_{\nu+\Delta}e^{-i(\nu+\Delta)t}$ and taking the Fourier transform of Eq. (2) at frequency $\nu+\Delta$. Assuming the frequency $\nu+\Delta$ is within the linewidth of the cavity we have $\hat{\mathbf{b}}_{\nu+\Delta} \approx -2\hat{\mathbf{a}}_{\nu+\Delta}/\sqrt{\gamma}$. We assume that σ_-^{23} is driven mainly by the field component at frequency $\nu+\Delta$. Using this approximation, the detuning of σ_-^{12} in the presence of the Stark field, denoted δ_s, becomes $\delta_s=\delta+\hat{\mathbf{S}}$, where

$$\hat{\mathbf{S}} = \frac{i2g_2^2\hat{\mathbf{b}}^\dagger_{\nu+\Delta}\hat{\mathbf{b}}_{\nu+\Delta}}{i\Delta + \dfrac{1}{2\tau_3}}. \tag{7}$$

The Stark operator $\hat{\mathbf{S}}$ has a real and imaginary component. The real component gives the optical Stark shift, while the imaginary component represents loss due to two-photon absorption. When $\Delta \gg 1/\tau_3$ this operator represents an energy shift that is proportional to the number of photons at frequency $\nu+\Delta$.

First, we consider the case where there is exactly one photon in the cavity at frequency $\nu+\Delta$, in which case we can substitute $\hat{\mathbf{b}}^\dagger_{\nu+\Delta}\hat{\mathbf{b}}_{\nu+\Delta}=1$. We assume $\Delta \gg \tau_3$ so that the Stark operator causes only a phase shift. To calculate the Stark shift, we must have a value for g_2. In the case of a quantum dot, we can use the single exciton and biexciton transitions for the 1-2 and 2-3 transitions. In this case, it is reasonable to assume that $g_2=g_1=g$, because both transitions represent an absorption of a photon by an exciton in the quantum dot. For other systems, the value of g_2 must be measured or calculated from the matrix element between $|2\rangle$ and $|3\rangle$.

Making the assumption that $g_1=g_2=g$, Fig. 4 plots the phase of the reflection coefficient for $\Delta=\infty$ (no Stark shift) and $\Delta=-3g$, with $g=0.3$ THz. The curve corresponding to $\Delta=\infty$ is identical to the one in Fig. 3. By introducting a Stark field, the resonant frequency of the first transition is shifted by $2g^2/\Delta$. The phase shift curve will therefore be translated to the new resonant frequency of the dipole. As the figure shows, $\Delta=-3g$ is enough to change the reflection phase from 0 to π in the presence of a single photon. In general, when $|\Delta|=\kappa/2+\gamma/2$, we have a frequency shift of $2g^2/(\kappa+\gamma)$, which is sufficiently large to change the phase by π. This

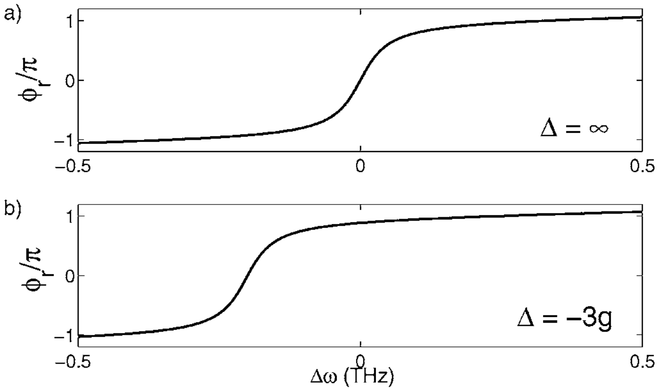

FIG. 4. Reflection phase shift in presence of a single photon inside the cavity at frequency $\nu+\Delta$. It is assumed that $g_1=g_2=g=0.3$ THz. (a) $\Delta=\infty$, (b) $\Delta=-3g$.

means that a single photon inside the cavity can provide a π phase shift if it is whithin the cavity linewidth.

In order to maintain one photon in the cavity with high probability, the Stark field must provide at least one photon per cavity lifetime γ. This is a reasonable lower bound to ensure the probability of zero-cavity photons is small. At the same time, the Stark field must have the same pulse duration as the reflected field. In order for the reflected field to fit within the linear dispersion region of the cavity reflection coefficient, its duration must be larger than the modified spontaneous emission lifetime of the dipole. This lifetime is given by γ/g^2 in the limit that $\kappa=0$ (losses to waveguide are dominant). The minimum number of photons in the Stark field is then given by multiplying the flux of the field by the minimum pulse duration, which leads to $n_{\mathrm{Stark}}>(\gamma/g_1)^2$. For typical experimental parameters using quantum dots in photonic crystal cavities, we have $g=0.3$ THz and $\gamma=1$ THz. This means that ten photons may be enough to see a significant Stark shift. As we approach the regime $g_1=\gamma$ (the high-Q regime), we can potentially achieve a π phase shift with a single photon. When $g>\gamma$, this phase shift can be achieved by loading the cavity with one photon at the Stark field frequency, and then reflecting a second pulse on resonance with the dipole within the photon lifetime of the cavity. Nevertheless, even in the low-Q regime where tens of photons or more are needed, the amount of nonlinearity is orders of magnitude bigger than conventional methods for cross-phase modulation. Such nonlinearities may find application for single photon QND measurements [1], as well as quantum computation methods that do not require a full π phase shift at the single photon level [18].

The system we consider exhibits large Kerr nonlinearities due to two properties. The first is that the confinement of the cavity field creates large values of g, which in turn generate very strong optical Stark shifts. But there is a second, more critical property. The largest phase difference is experienced near resonance with the dipole. In a normal system, we would not be able to drive a dipole resonantly without simultaneously suffering from large losses. In the case of DIT, however, we can drive the system on resonance and not suffer from absorption. This combination is the essential point of achieving large phase shifts.

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