Absorptionless self-phase-modulation via dark-state electromagnetically induced transparency

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We study a combination of two-level electromagnetically induced transparency and dark states, and show that desirable features of both phenomena can be obtained in a single system. In particular, large self-phase modulation can be produced without pump or probe absorption, and without spontaneous-emission noise. We point out possible application of our system as a source for squeezed light.

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I. INTRODUCTION

The closely related phenomena of dark states and electromagnetically induced transparency (EIT) have greatly enriched the fields of nonlinear and quantum optics in recent years. Although these two processes produce similar effects, they have important distinctions. Atomic dark states [1,2] are coherent superpositions of the two lower levels in a three-level lambda (Λ) system that are not coupled to the excited state by the two applied laser fields. An atom prepared in a dark state will not absorb the incident fields, have no excited-state population, and thus no spontaneous emission. Unfortunately for many applications in nonlinear optics, self-phase modulation also vanishes along with absorption. Electromagnetically induced transparency [3,4] is a pump-probe effect that provides transparency in a variety of systems [5]. While dark states are specific to Λ configurations, EIT occurs more generally: in two-level [6], cascaded three-level [7], and four-level systems [8]. In these systems there can be large third-order susceptibility, and thus self-phase modulation, in the absence of absorption of the probe field [9]. Unfortunately, spontaneous emission cannot be eliminated completely in these systems. The presence of spontaneous emission with associated noise limits the use of these EIT systems for noise-sensitive applications such as squeezed-light generation [10]. One possible exception using four fields is the double lambda system [11], which has been shown to produce a large third-order nonlinear susceptibility in the absence of absorption [9].

In this paper, we combine the benefits of dark states and two-level EIT to create a system that has no absorption, while still possessing large self-phase modulation (SPM) of the signal field. There is no absorption of the pump fields and, in addition, spontaneous-emission noise is eliminated because there is no population in the excited state. Such a system would be useful for applications such as squeezed-light generation via self-phase modulation and spatial soliton propagation. It should be noted that there is a recent study on the linear absorption of such a system [12]. Gain profiles have also been studied in a lambda system, where a single pump field is allowed to couple to both arms of the system [13].

The present paper is organized as follows. Section II presents the theoretical formulation, while Sec. III outlines the special features and drawbacks of the two-level and the three-level Λ systems and shows that these systems do not achieve the goals individually. In Sec. IV, a theoretical model of the dark-state electromagnetically induced transparency (DS-EIT) system is formulated for experiments in a sodium cell. The analysis first considers only radiative broadening, then includes the effects of collisional and Doppler broadening.

II. THEORETICAL FORMULATION

We will consider a closed three-level Λ system with two applied fields of the form $\tilde{E}(t)=E_b e^{-i\omega_b t}+E_d e^{-i\omega_d t}+\text{c.c}$, each coupling to a lower level, with corresponding Rabi with frequencies $\Omega_b=2\mu_{ab}E_b/\hbar$ and $\Omega_d=2\mu_{ac}E_d/\hbar$. Detunings of these fields with respect to the upper bare atomic state $|a\rangle$ are $\Delta_b=\omega_b-\omega_{ab}$ and $\Delta_d=\omega_d-\omega_{ac}$. Here, we generalized the E_d field to be comprised of a carrier and a modulation field in the form:

$$E_d(t)e^{-i\omega_d t}=E_c e^{-i\omega_c t}+E_s e^{-i\omega_s t}. \tag{2.1}$$

The population decay rates γ_b and γ_c are from $|a\rangle$ to the lower states $|b\rangle$ and $|c\rangle$. γ_1 and γ_2 are the population exchange rates between $|c\rangle$ to $|b\rangle$ and $|b\rangle$ to $|c\rangle$, respectively. The coherence dephasing rates are

$$\gamma_{ab}=\frac{\gamma_b+\gamma_c+\gamma_2}{2}+\gamma_{ab}^{coll},$$

$$\gamma_{ca}=\frac{\gamma_b+\gamma_c+\gamma_1}{2}+\gamma_{ca}^{coll},$$

$$\gamma_{cb}=\frac{\gamma_1+\gamma_2}{2}+\gamma_{cb}^{coll}, \tag{2.2}$$

where the phase decay due to elastic collisions is designated by the superscript *coll*. The complex dephasing rates are defined as

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$$\Gamma_{ab}=\gamma_{ab}-i\Delta_b=\Gamma_{ba}^*,$$

$$\Gamma_{ca}=\gamma_{ca}+i\Delta_c=\Gamma_{ac}^*,$$

$$\Gamma_{cb}=\gamma_{cb}+i\Delta_c-i\Delta_b=\Gamma_{bc}^*. \quad (2.3)$$

The quantity $\rho_{ij}=\sigma_{ij}\mu_{ij}e^{-i\omega_{ij}t}$ is the dipole moment expectation value (for $i\neq j$) and the population (for $i=j$). Starting from the general density-matrix equations of motion and using the population conservation rule for closed atomic systems,

$$1=\rho_{aa}+\rho_{bb}+\rho_{cc}, \quad (2.4)$$

we eliminate ρ_{aa} leaving

$$\dot{\rho}_{bb}-\gamma_b=-(\gamma_b+\gamma_2)\rho_{bb}-(\gamma_b-\gamma_1)\rho_{cc}+\frac{i}{2}\Omega_b^*\rho_{ab}-\frac{i}{2}\Omega_b\rho_{ba},$$

$$\dot{\rho}_{cc}-\gamma_c=-(\gamma_c-\gamma_2)\rho_{bb}-(\gamma_c+\gamma_1)\rho_{cc}+\frac{i}{2}\Omega_d^*\rho_{ac}-\frac{i}{2}\Omega_d\rho_{ca},$$

$$\dot{\rho}_{ab}+\frac{i}{2}\Omega_b=i\Omega_b\rho_{bb}+\frac{i}{2}\Omega_b\rho_{cc}-\Gamma_{ab}\rho_{ab}+\frac{i}{2}\Omega_d\rho_{cb},$$

$$\dot{\rho}_{ac}+\frac{i}{2}\Omega_d=\frac{i}{2}\Omega_d\rho_{bb}+i\Omega_d\rho_{cc}-\Gamma_{ac}\rho_{ac}+\frac{i}{2}\Omega_b\rho_{bc},$$

$$\dot{\rho}_{bc}=-\frac{i}{2}\Omega_d\rho_{ba}+\frac{i}{2}\Omega_b^*\rho_{ac}-\Gamma_{bc}\rho_{bc},$$

$$\dot{\rho}_{ba}-\frac{i}{2}\Omega_b^*=-i\Omega_b^*\rho_{bb}-\frac{i}{2}\Omega_b^*\rho_{cc}-\Gamma_{ba}\rho_{ba}-\frac{i}{2}\Omega_d^*\rho_{bc},$$

$$\dot{\rho}_{ca}-\frac{i}{2}\Omega_d^*=-\frac{i}{2}\Omega_d^*\rho_{bb}-i\Omega_d^*\rho_{cc}-\Gamma_{ca}\rho_{ca}-\frac{i}{2}\Omega_b^*\rho_{cb},$$

$$\dot{\rho}_{cb}=\frac{i}{2}\Omega_d^*\rho_{ab}-\frac{i}{2}\Omega_b\rho_{ca}-\Gamma_{cb}\rho_{cb}. \quad (2.5)$$

These equations can be written compactly in matrix form as

$$\dot{R}+\Sigma=MR, \quad (2.6)$$

where we define

$$R\equiv(\rho_{bb},\ \rho_{cc},\ \rho_{ab},\ \rho_{ac},\ \rho_{bc},\ \rho_{ba},\ \rho_{ca},\ \rho_{cb})^T \quad (2.7)$$

and Σ is a constant vector.

In this paper, we consider the effect of two strong fields on the behavior of a weak signal field. To explicitly show the third weaker signal field, we define

$$\Omega_d=\Omega_c+\Omega_s e^{-i\delta t}, \quad (2.8)$$

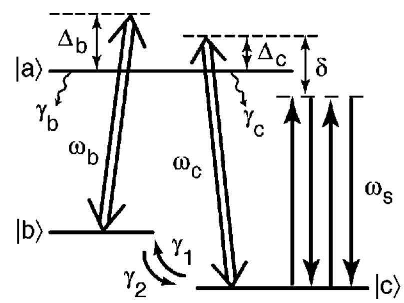

FIG. 1. The dark-state electromagnetically induced transparency system.

where Ω_c is the Rabi frequency of the strong pump field E_c, Ω_s is that of the weak signal field E_s and δ is the detuning between E_s and E_c ($\delta=\Delta_s-\Delta_c=\omega_s-\omega_c$), see Fig. 1. The matrix M and vector Σ are each partitioned into terms with different time dependence

$$M=M_0+e^{-i\delta t}\Omega_s M_1+e^{i\delta t}\Omega_s^* M_{-1}, \quad (2.9)$$

and

$$\Sigma=\Sigma_0+e^{-i\delta t}\Omega_s\Sigma_1+e^{i\delta t}\Omega_s^*\Sigma_{-1}. \quad (2.10)$$

Substituting these definitions into Eq. (2.6), we obtain

$$\dot{R}+\Sigma_0+e^{-i\delta t}\Omega_s\Sigma_1+e^{i\delta t}\Omega_s^*\Sigma_{-1}=(M_0+e^{-i\delta t}\Omega_s M_1+e^{i\delta t}\Omega_s^* M_{-1})R. \quad (2.11)$$

A simple application of Floquet's theorem shows that the stationary solution R to Eq. (2.11) will have only terms at the harmonics of the detuning δ. We assume that E_s is weak enough so that the Floquet harmonic expansion can be truncated at the third order

$$R=R_0+e^{-i\delta t}\Omega_s R_1+e^{i\delta t}\Omega_s^* R_{-1}+e^{-2i\delta t}(\Omega_s)^2R_2+|\Omega_s|^2H_0+e^{2i\delta t}(\Omega_s^*)^2R_{-2}+e^{-3i\delta t}(\Omega_s)^3R_3+e^{-i\delta t}\Omega_s|\Omega_s|^2H_1+e^{i\delta t}\Omega_s^*|\Omega_s|^2H_{-1}+e^{3i\delta t}(\Omega_s^*)^3R_{-3}. \quad (2.12)$$

The coefficients are obtained by substituting Eq. (2.12) into Eq. (2.11) and equating the coefficients of the different harmonics of δ and corresponding powers of Ω_s. The resulting equations are

$$R_1=(M_0+i\delta)^{-1}(\Sigma_1-M_1R_0), \quad (2.13)$$

$$H_1=-(M_0+i\delta)^{-1}(M_1H_0+M_{-1}R_2), \quad (2.14)$$

etc.

The linear susceptibility of the signal field can be obtained from the fourth element of the R_1 vector ($\rho_{ac}^{(1)}$) and the nonlinear susceptibility (for self-phase modulation) from the fourth element of the H_1 vector ($\rho_{ac}^{(3)}$). The total susceptibilities of the pump fields E_b and E_c can be obtained from

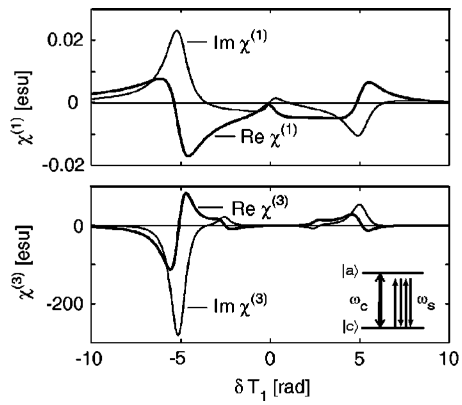

FIG. 2. Linear and nonlinear response of a homogeneously broadened, two-level system with $\Delta_c T_1 = 1$ and $\Omega_c T_1 = 5$.

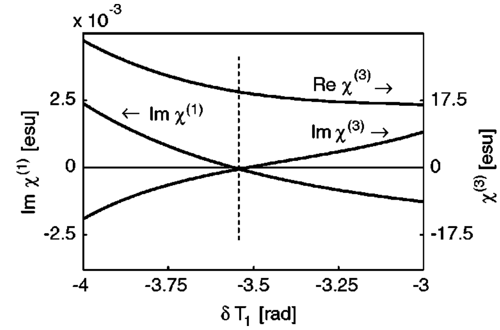

FIG. 3. Expanded view of Fig. 2, showing that the self-phase modulation coefficient (proportional to Re $\chi^{(3)}$) is nonzero at the detuning where there is no absorption of the signal field.

the third and fourth element of the R_0 vector ($\rho_{ab}^{(0)}$, $\rho_{ac}^{(0)}$), respectively. The population in the lower states $|b\rangle$ and $|c\rangle$ are from the first and second elements of the R_0 vector (ρ_{bb} and ρ_{cc}), respectively, and using Eq. (2.4), the population in the excited state can be calculated. These procedures give analytic expressions for $\chi^{(1)}$, $\chi^{(3)}$, etc. but these expressions are omitted here for brevity.

III. BUILDING BLOCKS

The combination of dark states and EIT produces a rich range of phenomena. To make sense the inherent complexity, and to see why we find it necessary to use the combination, we find it useful to examine limiting cases in which the various phenomena can be introduced separately. We begin with EIT in a simple two-level system, and then proceed to look at a three-level system.

A. EIT in a two-level system

We first analyze the two-level system [6] as a candidate for meeting the goals described above. EIT-like features can be found in a two-level system by applying two fields to the same transition. This simple system can be studied as a limiting case of the theory from Sec. II. We remove lower level $|b\rangle$ by setting $\Omega_b T_1$, $\gamma_1 T_1$, and $\gamma_b T_1$ to zero and $\gamma_c T_1 = 1$, where T_1 is the excited-state lifetime. We set $\gamma_2 T_1 \approx 10^{-10}$ to prevent numerical problems with inverting a singular matrix but the predictions obtained in this way are indistinguishable from those obtained with $\gamma_2 T_1 = 0$ as calculated in Refs. [6,9]. Here, we assume radiative decay ($T_2 = 2T_1$) unless stated otherwise. For comparison, the number density used in all numerical plots is identical to that of the Doppler broadened DS-EIT case; working with the hyperfine levels at the $D1$ line in a sodium cell at 300 °C, the number density is 2.3×10^{14} cm^{-3}.

The first-order probe absorption spectrum in this two-level case is often called the Mollow absorption spectrum [14]. A graph of the predictions of our theory for this spectrum in the case $\Delta_c T_1 = 1$ and $\Omega_c T_1 = 5$ is presented in the upper portion of Fig. 2. The strong pump field splits both the levels into doublets. Probing the system with a weak signal field on the same transition, we see a strong absorption peak on the red (lower frequency) side of resonance and gain on the blue side. These features are found at frequencies displaced by the generalized Rabi frequency ($\Omega_c' = \sqrt{\Omega_c^2 + \Delta_c^2}$) on either side of the bare-atom resonance. The difference in the amplitude between the absorption peak and the gain peak can be attributed to the one-photon detuning. Taking the analysis to third-order, we can identify a set of parameters where the SPM is relatively large while the linear absorption vanishes for the signal field (Fig. 3). An inherent disadvantage of a two-level system is that there is absorption of the pump field. Thus, as the pump Rabi frequency is reduced with propagation, the transparency window shifts and the result is a loss of transparency for the signal field. Another consequence is that there is substantial population in the excited state producing spontaneous emission. The population of the excited state of the two-level system in the above example is approximately 0.45. This system fails to satisfy all the goals that we have set.

B. Nonlinear optics of coherent population trapping

One of the important features of a regular three-level Λ system is that a dark state can be created. Similar to EIT, there is a transparency window for the signal field when the atom is excited at the two-photon resonance. With the two fields (pump and signal) coupling the Λ system (with no coupling between lower states), two coherent superpositions of the lower atomic basis states can be defined as

$$|+\rangle = \frac{\Omega_b}{\Omega'}|b\rangle + \frac{\Omega_s}{\Omega'}|c\rangle$$

$$|-\rangle = \frac{\Omega_s}{\Omega'}|b\rangle - \frac{\Omega_b}{\Omega'}|c\rangle, \tag{3.1}$$

where $\Omega' = \sqrt{\Omega_b^2 + \Omega_s^2}$. The state $|+\rangle$ is called the "bright" state as it is coupled via the pump and signal fields to the excited atomic state $|a\rangle$. On the other hand, the "dark state" $|-\rangle$ is completely decoupled. Then, if the atom begins in an arbitrary linear combination of the two lower levels, the amplitude that is initially in the bright state will be pumped up into the excited state, where it can spontaneously decay into either the bright or the dark state. In a few spontaneous emis-

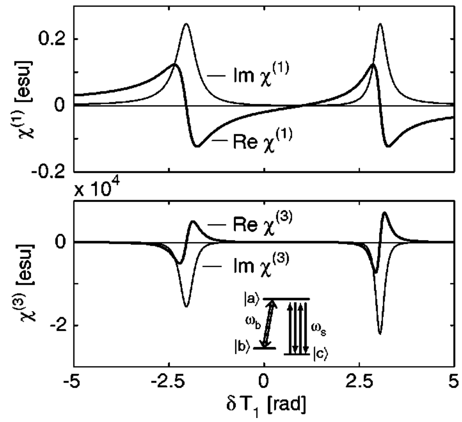

FIG. 4. Linear and nonlinear response of a homogeneously broadened, three-level Λ atom with $\Delta_b T_1 = 0$ and $\Omega_b T_1 = 5$. Note that the SPM coefficient vanishes at the frequency of induced transparency.

sion lifetimes all of the population will be pumped into the dark state, effectively decoupling the atom from the two applied fields. The transparency of the medium to the propagation of the pump and signal fields is not caused merely by the Autler-Townes splitting (the signal experiences two absorption peaks at the detuning of half the generalized Rabi frequency, $\Omega_b' = \sqrt{\Omega_b^2 + \Delta_b^2}$) but a destructive interference that cancels all absorption. Upon further analysis of the third-order susceptibility, one finds that the real part of $\chi^{(3)}$ also vanishes as the linear absorption vanishes. In fact, all susceptibilities vanish at exact two-photon resonance where a perfect dark state is created. These phenomena are illustrated in Fig. 4 for the case in which we set $\Delta_c T_1$, $\gamma_1 T_1$, $\gamma_2 T_1$, and $\Omega_c T_1$ to zero. With $\Delta_b T_1$ fixed, δT_1 is the two-photon detuning as well as the one-photon detuning of the signal field. We also take $\gamma_b T_1 = 3/8$, $\gamma_c T_1 = 5/8$, $\Omega_b T_1 = 5$, and $\Delta_b T_1 = 1$ in our illustrated example. While the system is ideal in the sense that there is no absorption or spontaneous emission, the desirable property of self-phase modulation also vanishes.

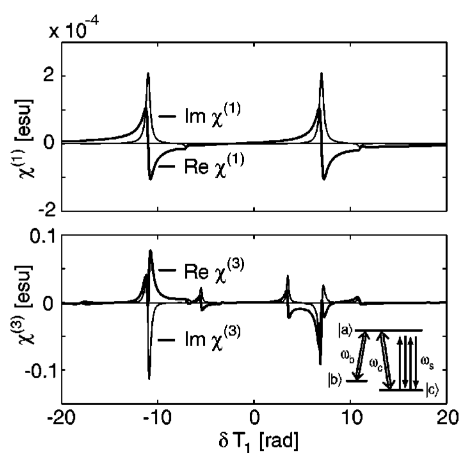

FIG. 5. Linear and nonlinear response of the radiatively broadened DS-EIT system with $\Delta_b T_1 = 2$, $\Delta_c T_1 = 0$, $\Omega_b T_1 = 0.5$, and $\Omega_c T_1 = 18$.

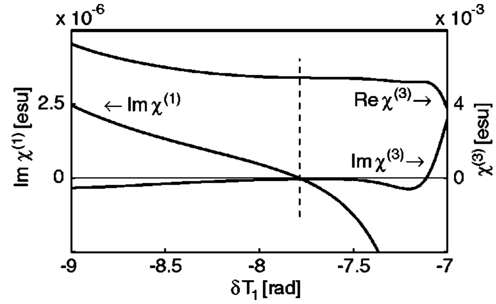

FIG. 6. Expanded view of Fig. 5, showing the SPM coefficient is nonvanishing at the frequency where the linear and nonlinear absorption vanish.

IV. THE DARK-STATE ELECTROMAGNETICALLY INDUCED TRANSPARENCY SYSTEM

With a suitable combination of the two systems considered above we can reap the benefits of each, while eliminating the limitations. In the DS-EIT system (Fig. 1), the two pump fields create the dark state and a third signal field interacts with the coherently prepared system. We can find a set of field parameters so that the SPM term is still large while both the linear and third-order nonlinear absorption are negligibly small. There is neither appreciable population in the excited state nor appreciable absorption of the pump fields. Thus, the transparency window, which is dependent on the Rabi frequency does not shift as the beams are propagated.

As reviewed in the previous section, if we have a perfect dark state, all the responses vanish at two-photon resonance. Thus, we need to find an atomic system for which the two lower levels have a larger direct coupling decay, or we need to detune the fields slightly off two-photon resonance. It is much easier to control the two-photon detuning experimentally. Consider our three-level system with $\Delta_b T_1 = 2$, $\Delta_c T_1 = 0$, $\Omega_b T_1 = 0.5$, $\Omega_c T_1 = 18$, $\gamma_b T_1 = 3/8$, and $\gamma_c T_1 = 5/8$. The predictions of the radiatively broadened model under these conditions are shown in Figs. 5 and 6 around the point of zero absorption with the excited-state population at approximately 10^{-4}. The excited-state population is also plotted as a function of the two pump Rabi frequencies in the absence of the signal field in Fig. 7.

The physics of the rather complex plots can be understood

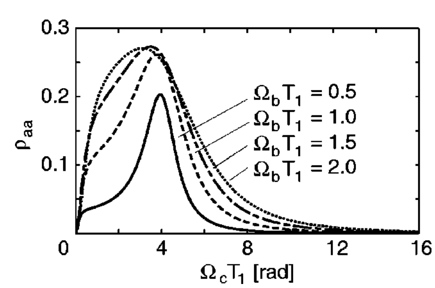

FIG. 7. Excited-state population as a function of the two pump amplitudes (expressed in terms of the normalized Rabi frequencies) for the case $\Delta_b T_1 = 2$. Note that the excited-state population can be neglected through the use of a pump field C sufficiently intense, $\Omega_c T_1 > 15$.

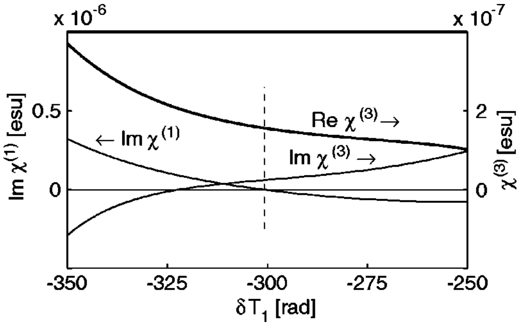

FIG. 8. Response of a DS-EIT system with the inclusion of Doppler broadening. $\Delta_b = 0.8$ GHz, $I_b = 5.08$ W/cm^2 and $I_c = 6.58$ kW/cm^2. Note that linear and nonlinear absorption vanish (around -3 GHz) while the SPM coefficient is retained.

in terms of resonances between the triplets that form the dressed states of the three-level, two strong field system [15]. We will not include such an analysis here, but simply present the graphical results.

Figure 8 shows a similar response when Doppler broadening is included. All three fields are copropagating in a sodium cell. At a cell temperature of 300 °C, a Gaussian distribution for the one-photon detuning is assumed with a width (half width at half maximum) of 0.9 GHz. To prevent the effects from averaging out, argon buffer gas is introduced with a density of 4.6×10^{18} cm^{-3}, which leads to collisional broadening of 0.6 GHz. For $\Delta_b = 0.8$ GHz, $I_b = 5.08$ W/cm^2 and $I_c = 6.58$ kW/cm^2, the excited state population is at 3.4×10^{-4}. At a detuning of -3 GHz, $\mathrm{Im}\,\chi^{(1)} = 8.4\times10^{-10}$ esu, $\mathrm{Im}\,\chi^{(3)} = 2.4\times10^{-8}$ esu, $\mathrm{Re}\,\chi^{(3)} = 1.6\times10^{-7}$ esu (positive), and $\beta_2 = \partial^2/\partial\delta^2\chi^{(1)} = -1.2\times10^{-11}$ s^2/m is of the opposite sign, satisfying one of the key conditions for temporal solitons. Other parameters such as critical energy (power) must also be achieved to establish a temporal (spatial) soliton but since that is beyond the scope of this paper, we will not be going into further details. The nonlinear phase shift is

$$\Delta\phi_{NL} = \frac{2\pi}{\lambda} n_2 I_s L_s, \tag{4.1}$$

where $n_2 = 0.0395\,\mathrm{Re}\,\chi^{(3)}/n_0^2$, $n_0^2 = 1 + 4\pi\,\mathrm{Re}\,\chi^{(1)}$, $\chi^{(1)}$ and $\chi^{(3)}$ are measured in esu, I_s is the signal intensity in W/cm^2, and $L_s = 1/\alpha$ is the absorption length for the signal field. I_c is assumed to be ten times stronger than I_s to prevent back action of the signal field onto the system. Using a cell length of 10 cm with the field absorption lengths much longer than that (L_s is 8.9 m), n_2 is 6.2×10^{-9} cm^2/W, giving a nonlinear phase shift of 4.3 rad. This is close to the value of 4 rad predicted by Blow *et al.* [16] for optimal squeezed-light generation via self-phase modulation.

V. CONCLUSION

We have shown that dark states can provide transparency with no spontaneous-emission noise, but with no SPM. A driven two-level system can enhance SPM with no absorption of the signal field but with appreciable excited-state population due to pump-field absorption. Combining the two, it can be seen from the graphs that the response of the DS-EIT system may be complicated but it does enjoy the benefits from both the two-level and the three-level Λ systems, while overcoming the disadvantages of both. Possible applications of this system are to the generation of spatial solitons or of squeezed light.

ACKNOWLEDGMENT

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