

Nonlinear optical properties of electromagnetically-induced-transparency medium interacting with two quantized fields

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We study linear and nonlinear optical properties of electromagnetically induced transparency (EIT) medium interacting with two quantized laser fields for adiabatic EIT case. We show that EIT medium exhibits normal dispersion. Kerr and higher order nonlinear refractive-index coefficients are also calculated in a completely analytical form. It is indicated that EIT medium exhibits giant resonantly enhanced nonlinearities. We discuss the response of the EIT medium to nonclassical light fields and find that the polarization vanishes when the probe laser is initially in a nonclassical state of no single-photon coherence.

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I. INTRODUCTION

In the last ten years, much attention has been paid to understandings and applications of systems exhibiting electromagnetically induced transparency (EIT) [1]. EIT is a powerful technique that can be used to make an otherwise absorbing medium transparent to a probe laser on resonance with a certain atomic transition by applying a coupling laser under the circumstances, while retaining the large and desirable nonlinear optical properties associated with the resonant response of the medium. Preparation of matter in such an EIT state would provide us with a new type of optical material of interest both its own right, and in many applications to fundamental and applied physics. There has been a large number of theoretical contributions in nonlinear optics using EIT [1, 2, 3, 4, 5]. The essence of EIT nonlinear optics is to create strongly-coupled Raman coherence for a three-level system and to control optical responses of the system. EIT has been observed in several different experimental configurations [5, 6, 7]. Recently, considerable attention has been drawn to slow group velocities of light and high nonlinearities in the conditions of EIT. Extremely slow group velocities of light pulses have been observed in Bose-Einstein condensate of ultracold sodium atoms [8, 9], in an optically dense hot rubidium gas [10], in rubidium vapor with slow ground state relaxation [11], and in crystals doped by rare-earth ions [12]. It has been shown that the condition of ultraslow light propagation leads to photon switching at an energy cost of one photon per event [13] and to efficient nonlinear processes at energies of a few photons per atomic cross section [14]. A giant cross-Kerr nonlinearity in EIT was suggested by Schmidt and Imamoglu [15], and has been indirectly measured in the experiment [8]. More recently, two groups [16, 17] have independently realized light storage in atomic mediums by using EIT technique.

Theoretically there are two formalisms to treat EIT. One is adiabatic EIT [1] in which both probe and coupling resonant lasers are adiabatically applied. After the system reaches a steady state, EIT occurs for arbitrary intensities of the probe and coupling lasers. The other is

transient-state EIT [4]. In this case, resonant probe and coupling lasers are simultaneously applied. EIT happens only when the intensity of the coupling laser is much larger than that of the probe.

Conventionally, both coupling and probe lasers were treated as classical, external fields. A disadvantage of the external field approach is that it can not deal with atom-photon and photon-photon quantum entanglement which is of importance not only because of the fundamental physics involved, but also for their potential technological applications such as quantum computation and quantum communication [18]. Recent studies on EIT have indicated that the possibility to coherently control the propagations of quantum probe light pulses in atomic media. This opens up interesting applications such as quantum state memories, generation of squeezed and entangled atomic states, quantum information processing, and as narrow-band sources of nonclassical radiation. In particular, with a quantum treatment of the probe laser, Fleischhauer and Lukin [19] have recently been able to predict the formation of dark-state polaritons in the propagation of light pulses through quantum entanglement of atomic and probe-photon states. This has been confirmed in the latest light storage experiments [16, 17]. This lesson teaches us that the quantum description of laser is more fundamental than the classical one, having advantages in uncovering new effects involving quantum nature of photons.

In a previous paper [20], We have developed a fully quantum treatment of EIT in a vapor of three-level Λ -type atoms. Both the probe and coupling lasers with arbitrary intensities are quantized, and treated on the same footing. The purpose of the present paper is to study the linear and nonlinear optical properties of an adiabatic EIT system interacting with two quantized fields and investigate the response of the EIT medium to nonclassical light fields.

This paper is organized as follows. In Sec. II, we set up our model and give an adiabatic solution of the full Hamiltonian. In Sec. III and IV, we investigate the linear and nonlinear optical properties of the EIT system. In Sec. V, we discuss the response of the EIT medium

to nonclassical light fields. Finally, we summarize our results and make concluding remarks in Sec. VI.

II. THE MODEL AND ADIABATIC SOLUTION

Let us consider a three-level atom, with energy levels $E_1 < E_3 < E_2$, interacting with two quantized laser fields, in the Λ -type configuration (see Fig. 1): The lower two levels $|1\rangle$ and $|3\rangle$ are coupled to the upper level $|2\rangle$. Going over to an interaction picture with respect to $H_0 = \sum_m E_m |m\rangle\langle m| - \hbar[\Delta_1(|2\rangle\langle 2| + |3\rangle\langle 3|) + \omega_1 \hat{a}_1^\dagger \hat{a}_1 + \omega_2 \hat{a}_2^\dagger \hat{a}_2]$, under the rotating-wave approximation, we obtain the total Hamiltonian of the system in the form

$$\hat{H} = \hbar\Delta_1|2\rangle\langle 2| + \hbar(\Delta_1 - \Delta_2)|3\rangle\langle 3| - \hbar(g_1\hat{a}_1|2\rangle\langle 1| + g_2\hat{a}_2|2\rangle\langle 3| + H.c.), \quad (1)$$

where $|m\rangle$ ($m = 1, 2, 3$) are atomic states, \hat{a}_j and \hat{a}_j^\dagger ($j = 1, 2$) the annihilation and creation operators of the probe and coupling laser modes, and the two coupling constants are defined by $g_1 = \mu_{21}\mathcal{E}_1/\hbar$ and $g_2 = \mu_{23}\mathcal{E}_2/\hbar$ with μ_{ij} denoting a transition dipole-matrix element between states $|i\rangle$ and $|j\rangle$, $\mathcal{E}_i = \sqrt{\hbar\omega_i/2\epsilon_0 V}$ being the electric field per photon for light of frequency ω_i in a mode volume V .

Eigenvalues of above Hamiltonian can be generally expressed as the following form

$$\begin{aligned} E_{n_1, n_2}^{(+)} &= \left[-\frac{q}{2} + \sqrt{\left(\frac{q}{2}\right)^2 + \left(\frac{p}{3}\right)^3} \right]^{1/3} \\ &\quad + \left[-\frac{q}{2} - \sqrt{\left(\frac{q}{2}\right)^2 + \left(\frac{p}{3}\right)^3} \right]^{1/3} - \frac{1}{3}A, \\ E_{n_1, n_2}^{(-)} &= \Lambda_1 \left[-\frac{q}{2} + \sqrt{\left(\frac{q}{2}\right)^2 + \left(\frac{p}{3}\right)^3} \right]^{1/3} \\ &\quad + \Lambda_2 \left[-\frac{q}{2} - \sqrt{\left(\frac{q}{2}\right)^2 + \left(\frac{p}{3}\right)^3} \right]^{1/3} - \frac{1}{3}A, \quad (2) \\ E_{n_1, n_2}^{(0)} &= \Lambda_2 \left[-\frac{q}{2} + \sqrt{\left(\frac{q}{2}\right)^2 + \left(\frac{p}{3}\right)^3} \right]^{1/3} \\ &\quad + \Lambda_1 \left[-\frac{q}{2} - \sqrt{\left(\frac{q}{2}\right)^2 + \left(\frac{p}{3}\right)^3} \right]^{1/3} - \frac{1}{3}A, \end{aligned}$$

where the two constants $\Lambda_1 = \exp(i2\pi/3)$ and $\Lambda_2 = \exp(-i2\pi/3)$, other parameters are given by

$$p = B - \frac{1}{3}A^2, \quad q = C - \frac{1}{3}AB + \frac{2}{27}A^3, \quad (3)$$

with

$$\begin{aligned} A &= -2\Delta_1 + \Delta_2, & C &= g_1^2 n_1 (\Delta_1 - \Delta_2), \\ B &= \Delta_1(\Delta_1 - \Delta_2) - g_2^2(n_2 + 1) - g_1^2 n_1. \end{aligned} \quad (4)$$

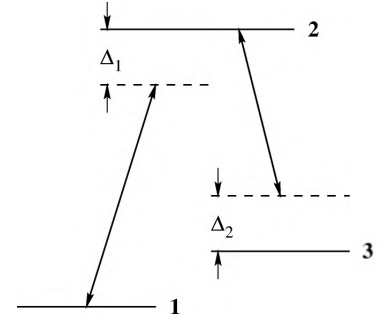


FIG. 1: A sketch of the atomic energy levels coupled to the coupling and probe fields.

The three eigenvalues $E_{n_1, n_2}^{(+)}$, $E_{n_1, n_2}^{(-)}$ and $E_{n_1, n_2}^{(0)}$ are unequal and real when p and q obey the inequality $q^2/4 + p^3/27 < 0$.

Since we are concerned with behaviors of the system in the vicinity of resonance, assuming the two detunings Δ_1 and Δ_2 are small, we can develop a perturbation theory in which we retain only terms linear in Δ_1 and Δ_2 . Then eigenvalues in Eq. (2) become the following simple expressions

$$\begin{aligned} E_{n_1, n_2}^{(\pm)} &= \frac{\Omega_1^2 + 2\Omega_2^2}{2\Omega^2} \Delta_1 - \frac{\Omega_2^2}{2\Omega^2} \Delta_2 \pm \frac{\Omega}{2}, \\ E_{n_1, n_2}^{(0)} &= \frac{\Omega_1^2}{\Omega^2} (\Delta_1 - \Delta_2), \end{aligned} \quad (5)$$

where we have defined $\Omega_1 = 2g_1\sqrt{n_1}$, $\Omega_2 = 2g_2\sqrt{n_2 + 1}$ and $\Omega = \sqrt{\Omega_1^2 + \Omega_2^2}$.

Eigenstates of the Hamiltonian (1) are called dressed states. Dressed states corresponding to above eigenvalues have the following form:

$$\begin{aligned} |\phi_{n_1, n_2}^{(i)}\rangle &= a_i|1, n_1, n_2\rangle + b_i|2, n_1 - 1, n_2\rangle \\ &\quad + c_i|3, n_1 - 1, n_2 + 1\rangle, \end{aligned} \quad (6)$$

where $i = \pm, 0$. Up to first-order perturbation theory in the detunings Δ_1 and Δ_2 , the coefficients in Eq. (6) are given by

$$\begin{aligned} a_{\pm} &= -\frac{\Omega_1}{\sqrt{2}\Omega} \left[1 \mp \frac{\Omega_1^2 + 4\Omega_2^2}{2\Omega^3} \Delta_1 \pm \frac{3\Omega_2^2}{2\Omega^3} \Delta_2 \right], \\ b_{\pm} &= \pm \frac{1}{\sqrt{2}} \left[1 \pm \frac{\Omega_1^2}{2\Omega^3} \Delta_1 \pm \frac{\Omega_2^2}{2\Omega^3} \right], \\ c_{\pm} &= -\frac{\Omega_2}{\sqrt{2}\Omega} \left[1 \pm \frac{3\Omega_1^2}{2\Omega^3} \Delta_1 \mp \frac{4\Omega_1^2 + 3\Omega_2^2}{2\Omega^3} \Delta_2 \right], \end{aligned} \quad (7)$$

and

$$a_0 = -\frac{\Omega_2}{\Omega}, \quad b_0 = \frac{2\Omega_1\Omega_2}{\Omega^3} (\Delta_1 - \Delta_2), \quad c_0 = \frac{\Omega_1}{\Omega}. \quad (8)$$

The complete set of dressed states for the system under consideration comprises the states $|\phi_{n_1, n_2}^{(i)}\rangle$ ($i = \pm, 0$) for

each $n_1 > 0$, and $n_2 \geq 0$ together with other two states $|1, 0, n_2\rangle$ and $|3, n_1, 0\rangle$ with zero eigenvalues. Knowing this set allows us to determine the time evolution of the atom-field system for any initial configuration.

We assume that the atom is initially in the ground state, while the coupling and probe lasers in a coherent state $|\alpha, \beta\rangle$, with α and β being supposed to be real for simplicity. Namely the initial state of the atom-field system is assumed to be

$$|\Psi(0)\rangle = |1\rangle \otimes |\alpha, \beta\rangle \equiv \sum_{n_1, n_2=0}^{\infty} C_{n_1, n_2} |1, n_1, n_2\rangle, \quad (9)$$

where the coefficient C_{n_1, n_2} is given by

$$C_{n_1, n_2} = \exp[-(\alpha^2 + \beta^2)/2] \frac{\alpha^{n_1} \beta^{n_2}}{\sqrt{n_1! n_2!}}. \quad (10)$$

Eq. (9) indicates that there is no atom-photon and photon-photon entanglement in the initial state of the atom-field system. In the usual treatment with both lasers being classical external fields, one is concerned with a steady state of the atomic system. The counterpart of the steady state in our fully quantum treatment, according to the commonly used *adiabatic hypothesis* in quantum scattering theory and quantum field theory[23], is the state that evolves from the initial state (9) with the couplings g_1 and g_2 *adiabatically turned on*. Physically this is equivalent to having localized laser pulses before they enter the atomic vapor, with the pulse shape sufficiently smooth. In conformity to the Adiabatic Theorem, we need to identify a linear combination of the dressed states (2) that tends to the initial state (9) if we take the limits $g_1, g_2 \rightarrow 0$ (or $\Omega_1, \Omega_2 \rightarrow 0$). According to Eqs. (3-5), the ordering of the limits $\Omega_1 \rightarrow 0$ and $\Omega_2 \rightarrow 0$ is important. Corresponding to the actual conditions in which EIT is observed, the correct ordering is first $\Omega_1 \rightarrow 0$ and then $\Omega_2 \rightarrow 0$. In our interaction picture, this procedure selects the $i = 0$ state in Eq. (6). Transforming it back to the Schrödinger picture, we identify the following state as the state that evolves adiabatically from the initial state:

$$|\Psi(t)\rangle = \sum_{n_1, n_2=0}^{\infty} C_{n_1, n_2} \exp[-ie_{n_1, n_2}^{(0)} t] |\phi_{n_1, n_2}^{(0)}\rangle. \quad (11)$$

where we have introduced the following notation

$$e_{n_1, n_2}^{(0)} = E_1 + \omega_1 n_1 + \omega_2 n_2 + E_{n_1, n_2}^{(0)}. \quad (12)$$

The state given in Eq. (11) can be viewed as the counterpart of the usual "steady" state in our treatment. We see that the population of the upper level $|2\rangle$ is zero up to first order in detunings Δ_1 and Δ_2 . This means that there is no absorption, implying the phenomenon of EIT. This is the adiabatic EIT. In the next section we will see that for transient-state EIT to occur the usual treatment requires the coupling laser be much stronger than the probe laser, i.e., $\Omega_2 \gg \Omega_1$; then most atoms are populated in the ground state. The adiabatic EIT does not

require this, so broader conditions are allowed: The coupling and probe lasers can be equally strong or even the probe laser is stronger. Then atoms can be in a more general superposition of two atomic states, which are entangled with the coupling and probe fields.

From Eq. (11) one can get the total density operator of the atom-field system. After taking trace over the field states, one finds the atomic reduced density operator to be

$$\begin{aligned} \rho^A(t) = \sum_{n_1, n_2=0}^{\infty} \{ & |D_1(n_1, n_2)|^2 |1\rangle\langle 1| \\ & + |D_2(n_1 + 1, n_2)|^2 |2\rangle\langle 2| \\ & + |D_3(n_1 + 1, n_2 - 1)|^2 |3\rangle\langle 3| \\ & + [D_1(n_1, n_2) D_2^*(n_1 + 1, n_2)] |1\rangle\langle 2| \\ & + D_1(n_1, n_2) D_3^*(n_1 + 1, n_2 - 1) |1\rangle\langle 3| \\ & + D_2(n_1 + 1, n_2) D_3^*(n_1 + 1, n_2 - 1) |2\rangle\langle 3| \\ & + H.c. \}, \end{aligned} \quad (13)$$

where D-coefficients are defined by

$$\begin{aligned} D_1(n_1, n_2) &= a_0 C_{n_1, n_2} \exp[-ie_{n_1, n_2}^{(0)} t], \\ D_2(n_1, n_2) &= b_0 C_{n_1, n_2} \exp[-ie_{n_1, n_2}^{(0)} t], \\ D_3(n_1, n_2) &= c_0 C_{n_1, n_2} \exp[-ie_{n_1, n_2}^{(0)} t], \end{aligned} \quad (14)$$

where $a_0, b_0, abd c_0$ are given by Eq. (8).

III. DISPERSION AND GROUP VELOCITY

When an electromagnetic wave of frequency ν propagates through a nonabsorptive but dispersive medium, the phase velocity is c/n with c being the speed of light in vacuum, and n the refractive index which is related to susceptibility of the medium χ by the relation $n = \sqrt{1 + \chi}$, while the group velocity is $c/[n + \nu(dn/d\nu)]$. So long as $dn/d\nu > 0$ which corresponds to normal dispersive, the group velocity is less than the phase velocity. In what follows we shall indicate that under EIT conditions both probe and coupling lasers exhibit normal dispersion, and their group velocities are greatly reduced while they have the same phase velocity c .

In order to get the susceptibility of the medium, we need to calculate the polarization of the atomic medium which is related to off-diagonal elements of the atomic reduced density operator through the following relation:

$$P = N(\mu_{12}\rho_{21}^A + \mu_{32}\rho_{23}^A) + c.c., \quad (15)$$

where N is the number density of atoms, μ_{ij} denotes a transition dipole-matrix element between states $|i\rangle$ and $|j\rangle$.

Transferring the induced polarization of the atomic medium P to a Fourier representation of a frequency ω , one can define the susceptibility of the medium $\chi(\omega)$ by

$$P(\omega) = \epsilon_0 \chi_s(\omega) E(\omega), \quad (16)$$

where $E(\omega)$ is the Fourier component of the mean value of the total electric field at the frequency ω , ϵ_0 the free space permittivity.

From Eqs. (13) and (14) we get the optical coherence of the system at time t

$$\begin{aligned}\rho_{21}^A(t) &= \sum_{n_1, n_2=0}^{\infty} P(n_1, n_2) a_0^*(n_1, n_2) b_0(n_1 + 1, n_2) \\ &\quad \times \frac{\alpha}{\sqrt{n_1 + 1}} \exp \left\{ i \left[e_{n_1, n_2}^{(0)} - e_{n_1+1, n_2}^{(0)} \right] t \right\}, \quad (17) \\ \rho_{23}^A(t) &= \sum_{n_1, n_2=0}^{\infty} P(n_1, n_2) b_0(n_1 + 1, n_2) c_0^*(n_1 + 1, n_2 - 1) \\ &\quad \times \frac{\sqrt{n_2} |\alpha|^2}{(n_1 + 1) \beta^*} \exp \left\{ i \left[e_{n_1+1, n_2-1}^{(0)} - e_{n_1+1, n_2}^{(0)} \right] t \right\},\end{aligned}$$

where the weighting factor is given by

$$P(n_1, n_2) = \frac{\bar{n}_\alpha^{n_1} \bar{n}_\beta^{n_2}}{n_1! n_2!} e^{-(\bar{n}_\alpha + \bar{n}_\beta)}, \quad (18)$$

with $\bar{n}_\alpha = |\alpha|^2$ and $\bar{n}_\beta = |\beta|^2$, respectively, being the initial mean photon numbers of the probe and coupling lasers in the coherent state $|\alpha, \beta\rangle$.

In the case $\bar{n}_\alpha \gg 1$ and $\bar{n}_\beta \gg 1$ ($\bar{n} \sim 10^4$ for a recent experiment of light storage [16], $\bar{n} \sim 10^4$), called the large- n limit below, the summation over n_1 and n_2 in Eq. (17) can be performed approximately. If one notices that the weighting function $P(n_1, n_2)$ will peak at values $n_1 = \bar{n}_\alpha$ and $n_2 = \bar{n}_\beta$ with relatively narrow dispersion: $(\langle n_\alpha^2 \rangle - \bar{n}_\alpha^2)^{1/2} = \sqrt{\bar{n}_\alpha}$ and $(\langle n_\beta^2 \rangle - \bar{n}_\beta^2)^{1/2} = \sqrt{\bar{n}_\beta}$, this implies that the photon distributions are sharply peaked around their mean values. So the rapid oscillations in the time record in Eq. (17) are just dominant oscillations which occur for $n_1 = \bar{n}_\alpha$ and $n_2 = \bar{n}_\beta$, i.e., $\Omega_1 = \bar{\Omega}_1(n_1 = \bar{n}_\alpha)$ and $\Omega_2 = \bar{\Omega}_2(n_2 = \bar{n}_\beta)$. Indeed we can expand the Rabi frequencies $\Omega_1 = 2g_1\sqrt{n_1}$ and $\Omega_2 = 2g_2\sqrt{n_2 + 1}$ about \bar{n}_α and \bar{n}_β to obtain

$$\begin{aligned}2g_1\sqrt{n_1} &= 2g_1\sqrt{\bar{n}_\alpha} + \frac{1}{\sqrt{\bar{n}_\alpha}}(n_1 - \bar{n}_\alpha) \\ &\quad - \frac{1}{4\bar{n}_\alpha^{3/2}}(n_1 - \bar{n}_\alpha)^2 + \dots \\ 2g_2\sqrt{n_2 + 1} &= 2g_2\sqrt{\bar{n}_\beta + 1} + \frac{1}{\sqrt{\bar{n}_\beta}}(n_2 - \bar{n}_\beta) \\ &\quad - \frac{1}{4\bar{n}_\beta^{3/2}}(n_2 - \bar{n}_\beta)^2 + \dots \quad (19)\end{aligned}$$

To the lowest order of $n_1 - \bar{n}_\alpha$ and $n_2 - \bar{n}_\beta$, we get $\Omega_1 = \bar{\Omega}_1(\bar{n}_\alpha)$ and $\Omega_2 = \bar{\Omega}_2(\bar{n}_\beta)$.

After substituting the lowest-order values of the Rabi frequencies, $a_0(n_1, n_2)$, $b_0(n_1, n_2)$, and $e_{n_1, n_2}^{(0)}$ in Eq. (17) are independent of n_1 and n_2 . The summations over n_1 and n_2 in Eq. (17) are carried out only for $P(n_1, n_2)/\sqrt{n_1 + 1}$ and $P(n_1, n_2)\sqrt{n_2}/(n_1 + 1)$. These

can be done easily. Then we obtain the Fourier component of the optical coherence of the adiabatically prepared state at the frequency ω

$$\begin{aligned}\rho_{21}^A(\omega) &= \bar{a}_0 \bar{b}_0 [\delta(\omega - \omega_1) + \delta(\omega + \omega_1)], \\ \rho_{23}^A(\omega) &= \bar{c}_0 \bar{b}_0 [\delta(\omega - \omega_2) + \delta(\omega + \omega_2)], \quad (20)\end{aligned}$$

where $\bar{a}_0 = a_0(\bar{n}_\alpha, \bar{n}_\beta)$, $\bar{b}_0 = b_0(\bar{n}_\alpha, \bar{n}_\beta)$, and $\bar{c}_0 = c_0(\bar{n}_\alpha, \bar{n}_\beta)$.

The Fourier component of the mean value of the total electric field $E(\omega)$ can be calculated from the adiabatically prepared state (11). In the large n approximation, we find that

$$\begin{aligned}E(\omega) &= \mathcal{E}_1 \alpha [\delta(\omega - \omega_1) + \delta(\omega + \omega_1)] \\ &\quad + \mathcal{E}_2 \beta [\delta(\omega - \omega_2) + \delta(\omega + \omega_2)], \quad (21)\end{aligned}$$

where $\mathcal{E}_i = (\hbar\omega_i/2\epsilon_0 V)^{1/2}$ ($i = 1, 2$) with V the quantized volume.

From Eq. (21) we obtain $E(\omega_1) = \mathcal{E}_1 \alpha$ and $E(\omega_2) = \mathcal{E}_2 \beta$, which imply that the mean values of the electric fields are unchanged when the pulses enter the medium. This is in agreement with the argument in Ref. [14] for the case of the classical probe and coupling laser fields. Since the power density is also unchanged as the pulses enter the medium, the energy density in the medium must increase. It is this increase that leads the spatial compression of a light pulse in the medium.

We first consider the propagation of the probe laser. For the frequency of the probe laser ω_1 , substituting Eqs. (20) and (21) into Eqs. (15) and (16) and retaining only terms linear in the detunings Δ_1 and Δ_2 we obtain the following expression of the susceptibility

$$\chi_s(\omega_1) = -\frac{4N|\mu_{12}|^2 \bar{\Omega}_2^2 (\Delta_1 - \Delta_2)}{\hbar\epsilon_0 (\bar{\Omega}_1^2 + \bar{\Omega}_2^2)^2}, \quad (22)$$

where $\bar{\Omega}_1 = \Omega_1(\bar{n}_\alpha, \bar{n}_\beta)$ and $\bar{\Omega}_2 = \Omega_2(\bar{n}_\alpha, \bar{n}_\beta)$ are the Rabi frequencies of the coupling and probe lasers, respectively. It should be pointed out that the expression (22) is just a perturbation result of the susceptibility for the small detunings up to the first order of Δ_1 and Δ_2 . Eq. (22) indicates that the imaginary part of the susceptibility vanishes under the first order approximation of the detunings. When the EIT occurs, we have $\Delta_1 = 0 = \Delta_2$. Then both real and imaginary parts of the susceptibility are zero. Hence, the Kramers-Kronig relation is not violated. When the coupling laser is on resonance, Eq. (22) reduces to the result in our previous paper [20]. It is interesting to note that this result is valid for arbitrary ratio of $\bar{\Omega}_1/\bar{\Omega}_2$.

We can see that Eq. (22) exhibits the signature of EIT: the linear susceptibility vanishes at the resonance ($\Delta_1 = 0 = \Delta_2$), which implies that the probe laser has the phase velocity c . From Eq. (22) we can also know that at resonance, $d\chi_s/d\omega_1 > 0$. Hence we can conclude that the medium exhibits the normal dispersion.

A dispersive medium can be characterized by a light group velocity. Steep dispersion is associated with larger

modifications of the group velocity. At the resonance frequency the group velocity of the probe laser pulse is related to the derivative of the susceptibility through the relation: $v_{pg}^{(s)} = c/[1 + (\omega_1/2)(d\chi_s/d\omega_1)]$, where the derivative of $\chi_s(\omega_1)$ is evaluated at the probe frequency ω_1 with c the speed of light in vacuum. Thus we have

$$v_{pg}^{(s)} = v_{pg}^0 \frac{(\bar{\Omega}_1^2 + \bar{\Omega}_2^2)^2}{\bar{\Omega}_2^4}, \quad (23)$$

where v_{pg}^0 is the usual expression for the group velocity [8] given by

$$v_{pg}^0 = \frac{\hbar c \epsilon_0 \bar{\Omega}_2^2}{2\omega_1 |\mu_{12}|^2 N}. \quad (24)$$

Eq. (23) indicates that in general the group velocity of the probe laser depends on the Rabi frequency of the coupling as well as the probe laser. The $\bar{\Omega}_1$ dependence of the group velocity is a new result of the fully quantum formalism, closely related to the higher order nonlinear susceptibilities we will discuss below.

These general results should be compared with earlier studies of EIT-based dispersive properties [24, 25]. In order to do this, we expand the susceptibility given in Eq. (22) in terms of the powers of $\bar{\Omega}_1/\bar{\Omega}_2$. When $\bar{\Omega}_1 \ll \bar{\Omega}_2$, the first term of this expansion gives us the linear susceptibility

$$\chi_s^{(1)}(\omega_1) = -\frac{4N|\mu_{12}|^2(\Delta_1 - \Delta_2)}{\hbar \epsilon_0 \bar{\Omega}_2^2}, \quad (25)$$

which is the same as Eq. (16a) in Ref. [25]. Note that the Rabi frequencies here are twice of those in Ref. [25] by definition.

On the other hand, when the probe and coupling lasers are on resonance, i.e., $\Delta_1 = 0 = \Delta_2$, from Eq. (25) we find that

$$\begin{aligned} \chi_s^{(1)}(\omega_1) &= 0, \quad \frac{d\chi_s^{(1)}}{d\omega_1} = \frac{4|\mu_{12}|^2 N}{\hbar \epsilon_0 \bar{\Omega}_2^2}, \\ \frac{d^2 \chi_s^{(1)}}{d\omega_1^2} &= 0. \end{aligned} \quad (26)$$

Thus, at the lowest order we recover results in Ref. [24] (see Table 1 in [24]), when the decay rates of states are neglected at the resonant frequency of the probe laser. Similarly, keeping only the lowest-order term in $\bar{\Omega}_1/\bar{\Omega}_2$, Eq. (23) will essentially give the same expression for the group velocity as reported in Ref. [8], i.e., the expression given by Eq. (24), which does not depend on $\bar{\Omega}_1$ and involves the contributions only from linear susceptibility.

The refractive index of the medium can be generally obtained from the susceptibility (22) by definition $n = \sqrt{1 + \chi}$. Making use of Eqs. (22) and (23), we find that the refractive index change in the vicinity of the resonance can be expressed in terms of the group velocity of the probe laser as

$$\Delta n_s(\omega_1) = \frac{\lambda_1 (\Delta_1 - \Delta_2)}{2\pi v_{pg}^{(s)}}. \quad (27)$$

It is worthwhile to note that this refractive-index change involves the contributions of all orders of nonlinear susceptibilities due to the intensity dependence of the group velocity in (23). For the slow light experiment of $v_{pg}^{(s)} = 17$ m/s with parameters [8] $\Delta_1 = 1.3 \times 10^6$ rad/s, $\Delta_2 = 0$, and $\lambda_1 = 589$ nm, from Eq.(27) we obtain $\Delta n_s(\omega_1) = 8.2 \times 10^{-3}$. This value is the same as that indirectly measured in Ref. [8]. It should be pointed out that the much larger change of refractive-index coefficients or the much larger normal dispersive of the medium which leads to much slower group velocity, is the direct result of EIT but not the result of strong coupling laser. In fact, a strong coupling laser is unnecessary for the adiabatic EIT situation.

We then consider the propagation of the coupling laser. For the frequency of the coupling laser ω_2 , from Eqs. (16), and (20) we obtain the following susceptibility

$$\chi_s(\omega_2) = \frac{4N|\mu_{32}|^2 \bar{\Omega}_1^2 (\Delta_1 - \Delta_2)}{\hbar \epsilon_0 (\bar{\Omega}_1^2 + \bar{\Omega}_2^2)^2}. \quad (28)$$

Eq. (28) indicates that at resonance, we have $\chi_s(\omega_2) = 0$, and $d\chi_s(\omega_2)/d\omega_2 > 0$. This means that the coupling laser has the phase velocity c and exhibits the normal dispersion.

When $\bar{\Omega}_1 \ll \bar{\Omega}_2$, we expand the susceptibility given in Eq. (28) in terms of powers of $\bar{\Omega}_1/\bar{\Omega}_2$. The first term of this expansion gives us the linear susceptibility of the coupling laser

$$\chi_s^{(1)}(\omega_2) = \frac{4N|\mu_{32}|^2 \bar{\Omega}_1^2 (\Delta_1 - \Delta_2)}{\hbar \epsilon_0 \bar{\Omega}_2^4}. \quad (29)$$

Thus, at the lowest order we recover results in Ref. [25] (Eq.(B2) in Ref. [25]).

Making use of the susceptibility (28), we obtain the group velocity for the coupling laser pulse

$$v_{cg}^{(s)} = v_{cg}^0 \frac{(\bar{\Omega}_1^2 + \bar{\Omega}_2^2)^2}{\bar{\Omega}_1^4}, \quad (30)$$

where v_{cg}^0 is given by

$$v_{cg}^0 = \frac{\hbar c \epsilon_0 \bar{\Omega}_1^2}{2\omega_2 |\mu_{32}|^2 N} = v_{pg}^0 \frac{\omega_1 I_1 |\mu_{12}|^2}{\omega_2 I_2 |\mu_{32}|^2}. \quad (31)$$

where v_{pg}^0 has given in Eq. (24), $I_1 = 2\epsilon_0 c \alpha^2$ and $I_2 = 2\epsilon_0 c \beta^2$ are the intensities of the incident probe and coupling lasers, respectively. For the slow light experiment of $v_{pg}^{(s)} = 17$ m/s with parameters [8] $I_1 = 1$ mW/cm², $I_2 = 40$ mW/cm², $\omega_1/\omega_2 \approx 1$ and $\mu_{12}/\mu_{32} \approx 1.22$, from Eqs. (30) and (31) we obtain $v_{cg}^{(s)} \approx 1020$ m/s. Hence the group velocity of the coupling laser is greatly reduced, although it is much greater than that of the probe laser.

The refractive-index change in the vicinity of the resonance is given by

$$\Delta n_s(\omega_2) = \frac{2N|\mu_{32}|^2 \bar{\Omega}_1^2 (\Delta_1 - \Delta_2)}{\hbar \epsilon_0 (\bar{\Omega}_1^2 + \bar{\Omega}_2^2)^2}, \quad (32)$$

which can be expressed as the following simple form

$$\Delta n_s(\omega_2) = \frac{\lambda_2 (\Delta_1 - \Delta_2)}{2\pi v_{cg}^{(s)}}. \quad (33)$$

For the slow light experiment of $v_{pg}^{(s)} = 17$ m/s with parameters [8] $\Delta_1 = 1.3 \times 10^6$ rad/s, $\Delta_2 = 0$, and $\lambda_2 \approx 589$ nm, from Eq.(33) we obtain $\Delta n_s(\omega_2) = 2.1 \times 10^{-4}$. This value is less one order of magnitude than that of the probe laser $\Delta n_s(\omega_1) = 8.2 \times 10^{-3}$.

IV. NONLINEAR REFRACTIVE INDICES

We note that in the conventional approach, it is difficult to obtain nonlinear susceptibilities higher than $\chi^{(1)}$ and $\chi^{(3)}$. However, in our formalism we can get arbitrary higher-order nonlinear susceptibilities once for all. For the propagation of the probe laser it is easy to see that the higher-order terms of the expansion in Eq. (22) give rise to higher-order nonlinear susceptibilities defined by

$$\chi_s(\omega_1) = \chi_s^{(1)}(\omega_1) + \chi_s^{(3)}(\omega_1)|E(\omega_1)|^2 + \chi_s^{(5)}(\omega_1)|E(\omega_1)|^4 + \chi_s^{(7)}(\omega_1)|E(\omega_1)|^6 + \dots (34)$$

where $\chi_s^{(1)}(\omega_1)$ is the linear susceptibility given in Eq. (25), and $\chi_s^{(k)}(\omega_1)$ ($k \geq 3$) represent the k th-order nonlinear susceptibility.

Again, assuming that the coupling laser is stronger than the probe, we expand the susceptibility (22) in powers of $\bar{\Omega}_1/\bar{\Omega}_2$. Rewriting this expansion in the form of Eq. (34), we find

$$\begin{aligned} \chi_s^{(3)}(\omega_1) &= \frac{8\hbar\epsilon_0 c^2 |\mu_{12}|^4 N}{|\mu_{32}|^4 I_2^2} (\Delta_1 - \Delta_2), \\ \chi_s^{(5)}(\omega_1) &= -\frac{24\hbar\epsilon_0^2 c^3 |\mu_{12}|^6 N}{|\mu_{32}|^6 I_2^3} (\Delta_1 - \Delta_2), \\ \chi_s^{(7)}(\omega_1) &= \frac{192\hbar\epsilon_0^3 c^4 |\mu_{12}|^8 N}{3|\mu_{32}|^8 I_2^4} (\Delta_1 - \Delta_2), \end{aligned} \quad (35)$$

where $\chi_s^{(1)}(\omega_1)$ is given by Eq. (25).

A light wave propagating through a nonlinear medium collects a nonlinear phase shift which is described by the expansion of the refractive index on powers of the light intensity [26]:

$$n_s = n_0^{(s)} + n_2^{(s)}|E_1|^2 + n_4^{(s)}|E_1|^4 + n_6^{(s)}|E_1|^6 + \dots (36)$$

where the first nonlinear correction to the refractive index is the Kerr coefficient $n_2^{(s)}$, which is related to $\chi_s^{(3)}(\omega_1)$. $n_k^{(s)}$ ($k \geq 4$) are higher-order nonlinear refractive-index coefficients, related to higher-order nonlinear susceptibilities up to $\chi_s^{(k+1)}(\omega_1)$.

Making use of Eq. (34), we find that the linear and nonlinear refractive-index coefficients can be expressed

in terms of the linear and nonlinear susceptibilities such as

$$\begin{aligned} n_0^{(s)}(\omega) &= 1 + \chi_s^{(1)}(\omega), \quad n_2^{(s)}(\omega) = \frac{1}{2n_0} \chi_s^{(3)}(\omega), \\ n_4^{(s)}(\omega) &= \frac{1}{2n_0} \chi_s^{(5)}(\omega) - \frac{1}{8n_0^3} \left(\chi_s^{(3)}(\omega) \right)^2, \\ n_6^{(s)}(\omega) &= \frac{1}{2n_0} \chi_s^{(7)}(\omega) - \frac{1}{4n_0^3} \chi_s^{(3)}(\omega) \chi_s^{(5)}(\omega) \\ &\quad + \frac{1}{16n_0^5} \left(\chi_s^{(3)}(\omega) \right)^3. \end{aligned} \quad (37)$$

Making use of Eq. (35), we obtain from Eq. (37) nonlinear refractive index coefficients:

$$\begin{aligned} n_2^{(s)} &= -\frac{2\epsilon_0 c}{I_2} \chi_s^{(3)}(\omega_1) = \frac{2\epsilon_0 c (\Delta_1 - \Delta_2)}{\pi I_2} \frac{\lambda_1}{v_{pg}^0}, \\ n_4^{(s)} &= -\frac{3\epsilon_0 c n_2^{(s)}}{I_2}, \quad n_6^{(s)} = -\frac{8\epsilon_0 c n_4^{(s)}}{3I_2}. \end{aligned} \quad (38)$$

Note that the sign of $n_i^{(s)}$ is of significance. In particular, the sign of $n_2^{(s)}$ can be used to examine the self-focusing/-defocusing effect of the medium. When $n_2^{(s)} > 0$, the medium is self-focusing. When $n_2^{(s)} < 0$, the medium is self-defocusing. Eq. (38) tell us that one can manipulate the self-focusing/-defocusing effect of the medium by changing the relative detuning between the coupling and probe lasers.

From Eqs. (35) and (38) we can see that to increase the value of nonlinear refractive-index coefficients one may either increase the atomic density or decrease the coupling laser intensity. Note that when the ratio of $\bar{\Omega}_1/\bar{\Omega}_2$ is close to unity, one had better directly deal with the original formula (22), rather than using its power series expansion.

In order to get some flavor of the magnitude of the giant nonlinearities derived above, we calculate the nonlinear refractive-index coefficients using the parameters ($I_2 = 40$ mW/cm², $\Delta_1 = 1.3 \times 10^6$ rad/s, $\Delta_2 = 0$, and $\lambda_1 = 589$ nm) in the ultraslow light experiment reported in Ref. [8], in which a light pulse speed 17 m/s was observed. We estimate that under these conditions, $n_2^{(s)} = 1.9 \times 10^{-7}$ m²/V², $n_4^{(s)} = -3.8 \times 10^{-12}$ m⁴/V⁴, and $n_6^{(s)} = 6.7 \times 10^{-17}$ m⁶/V⁶. In terms of a common practical unit, $n_2^{(s)} = 0.36$ cm²/W, $n_4^{(s)} = -13.0$ cm⁴/W², and $n_6^{(s)} = 4.5 \times 10^2$ cm⁶/W³. Hence, this value of the Kerr nonlinearity is consistent with that indirectly measured in Ref. [8], almost 10⁶ times greater than that measured in cold Cs atoms [8, 27], and $\sim 10^{12}$ times greater than that measured in other materials [28]. The fourth-order refractive-index coefficient $n_4^{(s)}$ is $\sim 10^{22}$ times greater than that measured in other materials [28].

The second nonlinear constants n_4 , known as the $\chi^{(5)}$ nonlinear index of refraction, is also an important parameter in contemporary nonlinear optics. Theoretical investigations show that in order to obtain spatial solitary waves a certain relation between n_2 and n_4 must be

satisfied. Specially, Wright and his coworkers [29] indicated that the ratio between the second- and the fourth-order refractive-index coefficients is an essential parameter to obtain stable spatial solitary waves. The lower the ratio $n_2^{(s)}/n_4^{(s)}$, the lower the required power for stable beam propagation. For the atomic medium with EIT, we have $n_2^{(s)}/n_4^{(s)} = -I_2/(3\epsilon_0 c)$. With the parameters in the experiment [8], we estimate $|n_2^{(s)}/n_4^{(s)}| \sim 10^{-2} \text{W/cm}^2$. This ratio is small compared to most other nonlinear media [28] by almost 11 orders of magnitude. From Eq. (37) we can also obtain the ratio between the fourth- and the sixth-order refractive-index coefficients $n_4^{(s)}/n_6^{(s)} = -3I_2/8(\epsilon_0 c)$, which is of the same order as that of $n_2^{(s)}/n_4^{(s)}$.

V. RESPONSE TO NONCLASSICAL FIELDS

We now discuss the influence of non-classical light on atomic polarization. Allowing such a study is one of the advantages of our fully quantum treatment of both the coupling and the probe laser fields. We assume that initially the atom is in the ground state, two laser fields in an arbitrary state $\sum_{n_1, n_2=0}^{\infty} C_{n_1} C_{n_2} |n_1, n_2\rangle$, and the coupling and probe fields are adiabatically applied. The "steady" state of the system is found to be

$$|\Psi(t)\rangle = \sum_{n_1, n_2=0}^{\infty} C_{n_1} C_{n_2} \exp[-ie_{n_1, n_2}^{(0)} t] |\phi_{n_1, n_2}^{(0)}\rangle. \quad (39)$$

The Fourier component of the optical coherence at probe-laser frequency is then given by

$$\rho_{21}^A(\omega_1) = \sum_{n_1, n_2=0}^{\infty} C_{n_1+1} C_{n_1}^* |C_{n_2}|^2 a_0(n_1, n_2) b_0(n_1, n_2). \quad (40)$$

which indicates that the atomic polarization depends upon the single-photon coherence of the initial probe field, i.e., $C_{n_1+1} C_{n_1}^*$. Hence, the atomic polarization is zero, if the probe field is initially in a state of no single-photon coherence.

This point can be understood by the following argument. The polarization is produced by the electric field. When the probe field is initially in a state of no single-photon coherence, the mean value of the electric field in the state vanishes, therefore the polarization of the medium is zero. When the coupling and probe lasers are in the coherent state $|\alpha, \beta\rangle$, Eq. (40) reduces to Eq. (20) in the large- n approximation. Obviously, the large- n approximation should be given up to investigate on the response of the EIT medium to nonclassical laser fields.

VI. CONCLUDING REMARKS

In conclusion we have studied the linear and nonlinear optical properties of EIT medium interacting with two

quantized laser fields for the adiabatic EIT case in terms of a time-independent approach in which both probe and coupling lasers are included as parts of the dynamical system under our consideration, and treated on the same footing. It has been shown that the fully-quantized treatment for both probe and coupling lasers can not only describes the ultraslow light experiments [8] very well, but also sheds new insight for the response of EIT media to nonclassical laser fields. In fact, we have found that very good agreement with experimental results for slow group velocities and the nonlinear refractive index of the probe laser observed in experiments [8]. We have shown that EIT medium exhibits normal dispersion. We have investigated the group velocities of both probe and coupling lasers in adiabatic EIT media. It has been found that the group velocities of both probe and coupling lasers are reduced. It should be pointed out that there are conditions for achieving slow group velocities of both probe and coupling lasers. In the perturbation regime, the weak-probe-strong-coupling-laser configuration leads to the fact that only the probe laser have significantly reduced group velocity. When using a strong-probe-strong-coupling-laser configuration, one transfers appreciable population to the excited state $|3\rangle$ at the early stage of the pulse. This is the familiar coherent population transfer stage. After this stage, both probe and coupling lasers take dual role: they both work partly as a probe and partly as a control laser. The consequence is that both will experience slow down. We have studied refractive-index changes of both probe and coupling lasers in the vicinity of the resonance. We have also calculated nonlinear susceptibilities and nonlinear refractive-index coefficients in a completely analytical form. We have indicated that EIT medium exhibits giant resonantly enhanced nonlinearities. We have discussed the response of the EIT medium to nonclassical light fields, and indicated that the polarization vanishes when the probe laser is initially in a nonclassical state of no single-photon coherence.

Finally, it should be remarked that in our treatment of EIT, which incorporates both probe and coupling lasers as part of the dynamical system, the decay parameters of various levels are ignored. This is a good approximation, since EIT is insensitive to any possible decay of the top level $|2\rangle$: During the adiabatic preparation the population of the level $|2\rangle$ in the dressed state $\phi^{(0)}$ remains negligibly small; see eqs. (6) and (8). Even if we add by hand an imaginary part to the energy of the level $|2\rangle$, it will *not* enter the energy eigenvalue for the dressed state $\phi^{(0)}$. Moreover, the effects of other decay parameters are expected to depend only on their ratios to $\bar{\Omega}_1$ or $\bar{\Omega}_2$, which normally are too small to dramatically change our results.

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