Limits on tradeoffs between third-order optical nonlinearity, absorption loss, and pulse duration in self-induced transparency and real excitation

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Self-induced transparency in a resonant two-level system creates a 2π-soliton pulse, which realizes large optical nonlinearities with a small absorption loss α for a short pulse duration τ. Quantum-mechanical zero-point field fluctuations introduce an ultimate dissipation loss in such a resonant and coherent process and place fundamental limits on the $\chi^{(3)}/\alpha\tau$ value. This value is independent of the dipole moment, atomic density, and pulse duration and is uniquely determined by an optical wavelength, e.g., $\chi^{(3)}/\alpha\tau \sim 1.2\times10^{21}\lambda^4$ esu cm/s. The similar limit on $\chi^{(3)}/\alpha\tau$ value is obtained in a usual operation mode, when the pulse duration becomes much longer than the atomic decay constants and the real excitation of the atoms occurs instead of the virtual excitation in self-induced transparency. The implication of these limits on optical squeezed-state generation, quantum nondemolition measurement, and reversible logic is discussed.

I. INTRODUCTION

Optical nonlinear processes can be divided into two categories: nonresonant-coherent and resonant-incoherent. If a field frequency is well detuned from an atomic transition frequency, the excitation of atoms is virtual; thus the response time τ is fast and the absorption loss α is small, while the third-order susceptibility $\chi^{(3)}$ is usually small. On the other hand, when a field frequency is close to an atomic transition frequency, real excitation of atoms occurs, resulting in the $\chi^{(3)}$ coefficient being resonantly enhanced while both the τ and α values become large. It is believed that some trade-offs exist between the $\chi^{(3)}$ coefficient and the α and τ values. In fact, a $\chi^{(3)}/\alpha\tau$ value is more or less constant for various nonlinear materials.[1] However, so far there is no answer to the question of whether there are any fundamental limits on a $\chi^{(3)}/\alpha\tau$ value.

In a previous paper,[2] we demonstrated new optical nonlinearities (of the third category) based on a 2π-soliton pulse in self-induced transparency (SIT). The 2π-soliton pulse excites all the atoms into an upper state with the leading edge of the pulse and stimulates a down conversion to the ground state with the trailing edge of the pulse. If the atoms are decoupled from all the reservoirs and are free from any relaxation processes, the 2π-soliton pulse propagates without suffering from absorption loss at all. The 2π-soliton pulse features photon-number-dependent self-phase modulation during its propagation and has mutual-phase modulation during its collision with the other soliton. These characteristics have the potential to realize an extremely large $\chi^{(3)}$ coefficient. Moreover, the pulse duration can be much shorter than the relaxation time constants of the atoms. At first sight, it seems to be an ideal nonlinear optical process. However, a certain atom-reservoir coupling cannot be eliminated in principle. It is a radiative decay of atomic dipoles due to quantum-mechanical zero-point field fluctuations. Of course, it is possible to eliminate a field mode which carries zero-point field fluctuations and to inhibit an atom's spontaneous emission by the technique of cavity quantum electrodynamics.[3] In such a case, however, coupling between the atoms and the 2π-soliton pulse is also inhibited. This spontaneous emission decay introduces a finite absorption loss to the 2π-soliton pulse and places the fundamental limits on the $\chi^{(3)}/\alpha\tau$ value achieved with the 2π solitons in SIT.

This paper is organized as follows. In Sec. II, the limit on $\chi^{(3)}/\alpha\tau$ value defined by self-phase modulation in SIT is derived. Moreover, it is demonstrated that the degree of squeezing using self-phase modulation is limited by such limits on $\chi^{(3)}/\alpha\tau$ value. In Sec. III, the limit on $\chi^{(3)}/\alpha\tau$ value defined by mutual-phase modulation in SIT is derived. The necessary minimum photon numbers in quantum nondemolition (QND) measurement and reversible logic are limited by such limits on $\chi^{(3)}/\alpha\tau$ value.

II. LIMIT ON $X^{(3)}/\alpha\tau$ VALUE IN SELF-PHASE MODULATION

The self-phase modulation ϕ_{self} of the 2π-soliton pulse is given by Eq. (3) in Ref. 2. The effective $\chi^{(3)}$ coefficient is defined as the derivative of ϕ_{self} with respect to the intensity $I_P = N_P\hbar\omega/A_P\tau$ of the soliton, giving

$$\chi^{(3)}_{\text{self}} \equiv \frac{c^2\epsilon_0^2}{\omega z}\frac{d\phi_{\text{self}}}{dI_P} = \frac{p_{21}^4 n\tau_s^3}{8\hbar^3}\frac{\Delta\omega\tau_s}{[1+(\Delta\omega\tau_s)^2]^2} . \quad (1)$$

Here N_P is the soliton photon number, A_P is the cross-sectional area, $\tau_s = \tau/2$ is the soliton pulse half-duration, p_{21} is the atomic moment, n is the atomic density, and $\Delta\omega = \omega - \omega_{21}$ is the field frequency ω detuned from the atomic transition frequency ω_{12}. We assume that the atoms are in free space. Generalization of Eq. (1) to a dielectric matrix with refractive index μ is straightforward.[2]

The spontaneous-emission lifetime (longitudinal relaxation time constant) T_1 for the atoms in free space is

$$T_1 = \frac{h\epsilon_0 c^3}{2\omega^3 p_{21}^2} . \tag{2}$$

This uniquely determines the upper limit on a transverse relaxation time constant to be $T_2 = 2T_1$. Here we assumed that the quantum-mechanical zero-point field fluctuation is only one reservoir which dissipates the atomic coherence and energy. Once the atoms have finite T_1 and T_2 time constants, the soliton loses energy partly because the dipole moment loses phase memory and also because the upper-state incoherently decays to the ground state. The decrease in the soliton photon number is described by[4]

$$\frac{d}{dz}\left[\frac{N_P}{A_P}\right]^2 = -64\frac{n\hbar c\epsilon_0}{\omega p_{21}^2}\left[\frac{1}{T_2}\frac{1}{1+(\Delta\omega\tau_s)^2}+\frac{2}{3}\left[\frac{1}{T_1}-\frac{1}{T_2}\right]\frac{1}{[1+(\Delta\omega\tau_s)^2]^2}\right] . \tag{3}$$

Even though the energy-dissipation process is nonlinear, as shown in Eq. (3), the effective linear absorption coefficient α can still be derived in a small-absorption limit,

$$\alpha = \frac{np_{21}^4\omega^4\tau_s^2}{12\pi\hbar^2\epsilon_0^2 c^4}\left[\frac{3}{1+(\Delta\omega\tau_s)^2}+\frac{2}{[1+(\Delta\omega\tau_s)^2]^2}\right] , \tag{4}$$

where Eq. (2) is used for T_1 and $T_2 = 2T_1$.

From Eqs. (1) and (3), the $\chi^{(3)}/\alpha\tau$ value for a self-phase modulation process is given by

$$\frac{\chi_{\text{self}}^{(3)}}{\alpha\tau} = \frac{3\pi\epsilon_0^2 c^4}{4\hbar\omega^4}\frac{\Delta\omega\tau_s}{3[1+(\Delta\omega\tau_s)^2]+2} . \tag{5}$$

The $\chi^{(3)}/\alpha\tau$ value is zero both at the resonance point, $\Delta\omega = 0$, and far from the resonance point, $\Delta\omega \gg 1/\tau_s$. The maximum $\chi^{(3)}/\alpha\tau$ value, in esu cm/s, is obtained at the optimum frequency detuning, $\Delta\omega_{\text{opt}} = \sqrt{\tfrac{5}{3}}(1/\tau_s)$:

$$\left.\frac{\chi_{\text{self}}^{(3)}}{\alpha\tau}\right|_{\Delta\omega_{\text{opt}}} = \left[\frac{3}{5}\right]^{1/2}\frac{\epsilon_0^2\lambda^4}{64\pi^2 h} \sim 1.18\times10^{21}\lambda^4 . \tag{6}$$

Here λ is an optical wavelength in centimeters. The $\chi^{(3)}/\alpha\tau$ value is independent of the dipole moment p_{21}, the pulse duration τ, and the atomic density n. It is uniquely determined by the optical wavelength λ and is proportional to λ^4.

The spontaneous lifetime, Eq. (2), represents the total radiative decay rate into all continuum modes in free space. If an atom is put into a single-mode waveguide surrounded by superconducting walls (or equivalent optical high-reflectivity mirrors),[3] the atom is decoupled from all continuum modes except one guided mode. The spontaneous-emission lifetime for such a case is modified to be

$$T_1^* = \frac{\hbar\epsilon_0 c}{\omega p_{21}^2}A_P . \tag{7}$$

Since the cross-sectional area A_P of the single-mode waveguide must be in the order of $\sim\lambda^2/4$, Eq. (7) is almost equal to Eq. (2), and the fundamental limit, Eq. (6) for the $\chi^{(3)}/\alpha\tau$ value, is preserved.

The effective linear absorption coefficient (6) holds only for $\tau \ll T_1, T_2$. In an opposite limiting case, $\tau \gg T_1, T_2$, the self-induced transparency breaks down and the real excitation of atoms occurs. For such a case, the $\chi^{(3)}$ and α value are calculated by the usual theory, giving[5]

$$\chi^{(3)} = \frac{p_{21}^4 n T_2^3}{2\hbar^3}\frac{\Delta\omega T_2}{[1+(\Delta\omega T_2)^2]^2} , \tag{8}$$

$$\alpha = \frac{np_{21}^4\omega^4 T_2^2}{2\pi\hbar^2\epsilon_0^2 c^4}\frac{1}{1+(\Delta\omega T_2)^2} . \tag{9}$$

It is interesting to note that if the SIT pulse duration τ is replaced by the transverse relaxation time constant T_2 in Eqs. (1) and (4), they are reduced to the usual real excitation $\chi^{(3)}$ and α values given by Eqs. (8) and (9). The $\chi^{(3)}/\alpha\tau$ value is given by

$$\frac{\chi^{(3)}}{\alpha\tau} = \frac{T_1}{\tau}\frac{\epsilon_0^2}{8\pi^2 h}\lambda^4 . \tag{10}$$

Here the optimum detuning, $\Delta\omega_{\text{opt}} = 1/T_2$, and $T_2 = 2T_1$ are assumed. The $\chi^{(3)}/\alpha\tau$ value depends on the pulse duration τ and the longitudinal time constant T_1. The $\chi^{(3)}/\alpha\tau$ value increases with decreasing τ and approaches the limit (6) when $\tau \simeq 10T_1$. In other words, the real excitation $\chi^{(3)}$ process for $\tau \gg T_1, T_2$ has the nearly equal limit (6) under the minimum allowable pulse duration $\tau \simeq 10T_1$.

Figure 1 illustrates the fundamental limits on the $\chi^{(3)}/\alpha\tau$ value as a function of the wavelength. $\chi^{(3)}/\alpha\tau$ values reported so far for various nonlinear materials[6–15] are also shown for comparison. These experimental results are not for the 2π soliton in self-induced transparency, but are for the real excitation $\chi^{(3)}$ process. The relation $\tau = 10T_1$ is assumed for comparing the theory and experiments.

The quantum phase diffusion (photon-number-dependent self-phase modulation) in a $\chi^{(3)}$ medium produces a number-phase squeezed state.[16] Optical solitons are also squeezed by this process.[2,17] However, an absorption loss in the $\chi^{(3)}$ medium introduces a vacuum field fluctuation and partly destroys squeezing. The quantum noise reduction is thus limited. The degree of squeezing can easily be calculated by the operator evolution equation with a dissipation-fluctuation term, the result being

$$\frac{\langle\Delta\hat{n}^2\rangle}{\langle\hat{n}\rangle} = \frac{3}{8(2^{1/3})}\left[\frac{\epsilon_0^2\lambda^4}{\pi h N_P}\frac{\alpha\tau}{\chi^{(3)}}\right]^{2/3} , \tag{11}$$

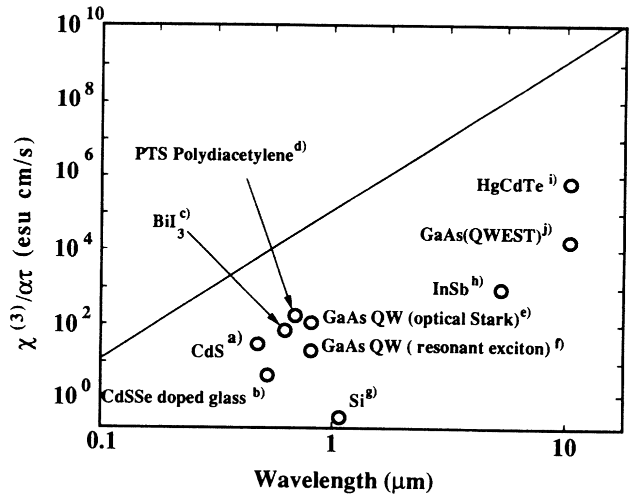

FIG. 1. The fundamental limits on $\chi^{(3)}/\alpha\tau$ values as a function of the wavelength, and the estimated values for the various nonlinear materials from the experimental data. In the estimation, $\tau=10T_1$ is assumed. (a)Reference 6; (b)Ref. 7; (c)Ref. 8; (d)Ref. 9; (e)Ref. 10; (f)Ref. 11; (g)Ref. 12; (h)Ref. 13; (i)Ref. 14; (j)Ref. 15.

where $A_P=\lambda^2/4$ is used. If Eq. (6) is used in Eq. (11), the maximum degree of squeezing is uniquely determined by the soliton photon number, $\langle \Delta \hat{n}^2 \rangle / \langle \hat{n} \rangle \simeq 10/N_P^{2/3}$. In order to generate a squeezed soliton, the soliton photon number must be greater than 50.

III. LIMIT ON $\chi^{(3)}/\alpha\tau$ VALUE IN MUTUAL-PHASE MODULATION

The mutual-phase modulation ϕ_{mutual} between two 2π-soliton pulses during the collision process is given by Eq. (5) in Ref. 2. The effective $\chi^{(3)}$ coefficient is defined as the derivative of the phase shift $\phi_{\text{mutual},1}$ of soliton 1 with respect to the intensity I_{p2} of soliton 2, giving

$$\chi^{(3)}_{\text{mutual}} = \frac{p_{21}^4 n \tau_s^3}{32\hbar^3} \frac{\Delta\omega\tau_s}{1+(\Delta\omega\tau_s)^2} . \tag{12}$$

Here we assume that the frequency ω of soliton 1 is slightly detuned from the atomic transition frequency ω_{21} and that the frequency of soliton 2 is equal to ω_{12}. Since soliton 2 is on resonance, it suffers from a higher absorption loss than soliton 1:

$$\alpha_2 = \frac{5np_{21}^4 \omega^4 \tau_s^2}{12\pi\hbar^2 \epsilon_0^2 c^4} > \alpha_1 . \tag{13}$$

From (12) and (13), we obtain the $\chi^{(3)}/\alpha\tau$ value for the mutual-phase modulation process:

$$\frac{\chi^{(3)}_{\text{mutual}}}{\alpha\tau} = \frac{3\pi\epsilon_0^2 c^4}{80\hbar\omega^4} \frac{\Delta\omega\tau_s}{1+(\Delta\omega\tau_s)^2} . \tag{14}$$

The maximum $\chi^{(3)}/\alpha\tau$ value, in esu cm/s, is obtained at the optimum detuning, $\Delta\omega_{\text{opt}}=1/\tau_s$, as

$$\left.\frac{\chi^{(3)}_{\text{mutual}}}{\alpha\tau}\right|_{\Delta\omega_{\text{opt}}} = \frac{3}{80}\frac{\epsilon_0^2\lambda^4}{(4\pi)^2 h} \sim 2.3\times 10^{20}\lambda^4 . \tag{15}$$

$\chi^{(3)}_{\text{mutual}}/\alpha\tau$ is slightly smaller than $\chi^{(3)}_{\text{self}}/\alpha\tau$.

The mutual-phase modulation between the signal and probe pulses in a $\chi^{(3)}$ medium results in the quantum nondemolition measurement of the signal photon number.[18] The collision of two solitons also results in the quantum nondemolition measurement of the signal photon number.[19] Without attenuating the signal photon number, it can be measured via the probe phase shift. As demonstrated in Ref. 2, the collision of two 2π solitons results in a more efficient quantum nondemolition measurement than the collision of two fiber solitons,[19] when soliton 1 at ω is assigned as the signal and soliton 2 at ω_{21} is assigned as the probe. With this assignment, the signal soliton has less absorption loss, due to frequency detuning, and the probe soliton is free from self-phase modulation, due to on-resonant excitation of the atoms. Suppose that the probe soliton is prepared in an optimum phase-squeezed state; then the probe phase noise is given by $\langle \Delta\hat{\phi}_P^2 \rangle_{SS} \sim 1/4\langle \hat{N}_P \rangle^2$, rather than $\langle \Delta\hat{\phi}_P^2 \rangle_{CS} = 1/4\langle \hat{N}_P \rangle$ for a coherent state. The measurement error $(\langle \Delta\hat{N}_S^2 \rangle_{\text{meas}})^{1/2}$ of the signal photon number, defined by the signal-to-quantum-noise ratio of unity, is given by

$$(\langle \Delta\hat{N}_S^2 \rangle_{\text{meas}})^{1/2} = \frac{\pi^2 c^4 \epsilon_0^2 \tau_s}{2\hbar\omega^4 \langle \hat{N}_P \rangle z \chi^{(3)}_{\text{mutual}}} . \tag{16}$$

In order to achieve measurement accuracy for "one photon" $(\langle \Delta\hat{N}_S^2 \rangle_{\text{meas}})^{1/2}=1$, the probe soliton loses energy according to

$$\hbar\omega\langle \hat{N}_P \rangle \alpha z = \frac{c\epsilon_0^2\lambda^3}{8\pi}\left[\frac{\alpha\tau}{\chi^{(3)}_{\text{mutual}}}\right] \simeq 40h\nu . \tag{17}$$

Here we use Eq. (15) in the second equality. This is the ultimate energy dissipation of the probe per one quantum nondemolition measurement. The energy dissipation of the signal can be arbitrarily decreased by a large probe photon number. There is no fundamental limit on the energy dissipation of the signal. This makes the quantum nondemolition measurement to be a physically realizable notion.

The mutual-phase modulation in a $\chi^{(3)}$ medium also can be used to realize reversible optical logic, for instance, an optical Fredkin gate.[20] Without dissipating the signal photon number, universal logic operations such as AND, OR, NOT, and COPY can be constructed. For this purpose, the signal soliton at ω must be phase modulated by π with the control soliton at ω_{12}, which requires

$$\frac{\hbar\omega^4\langle \hat{N}_P \rangle z \chi^{(3)}_{\text{mutual}}}{\pi^3 c^4 \epsilon_0^2 \tau_s} = 1 . \tag{18}$$

The control soliton loses its energy according to

$$\hbar\omega\langle \hat{N}_P \rangle \alpha z = \frac{c\epsilon_0^2\lambda^3}{4}\left[\frac{\alpha\tau}{\chi^{(3)}_{\text{mutual}}}\right] \simeq 260h\nu . \tag{19}$$

There exists a minimum required energy dissipation per one logic operation.

IV. CONCLUSION

It is shown that the limits on the trade-offs between a third-order susceptibility $\chi^{(3)}$, linear absorption loss α, and pulse duration τ in SIT and real excitation. The maximum value for a figure of merit, $\chi^{(3)}/\alpha\tau$, is uniquely determined by the optical wavelength and is proportional to λ^4. The limits on optical squeezed-state generation, quantum nondemolition measurement, and reversible logic are determined by this trade-off relation. The calculation is based on the 2π soliton in self-induced transparency at one-photon resonance. The same limit is obtained for the real excitation $\chi^{(3)}$ process under the optimum condition. It is still an open question as to whether this limit is a universal one or not, even though all the experimental results reported so far are below this limit, as shown in Fig. 1. Specifically, the relation does not necessarily hold a $\chi^{(3)}$ process enhanced either by two-photon resonance or by excitonic gigantic dipoles.

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