Optical Nonlinearities of Excitonic Self-Induced-Transparency Solitons: Toward Ultimate Realization of Squeezed States and Quantum Nondemolition Measurement

K. Watanabe, H. Nakano, A. Honold,[(a)] and Y. Yamamoto

Nippon Telegraph and Telephone Corporation, Basic Research Laboratories, Musashino-Shi, Tokyo 180, Japan

(Received 20 December 1988)

Self-induced transparency in a resonant two-level system creates a 2π-soliton pulse, which produces large optical nonlinearities with a very fast response time and an extremely small loss. The self-phase modulation of a 2π soliton and the mutual-phase modulation by the collision of two 2π solitons in the excitonic range of the spectrum in CdS can achieve an effective $\chi^{(3)}$ coefficient of 10^{-6} to $10^{-4.5}$ esu for a pulse duration of 1 to 3 ps. The quantum theory of 2π solitons predicts this new nonlinear process can realize squeezed states and quantum nondemolition measurement 20 dB below the standard quantum limit.

PACS numbers: 42.50.Qg, 32.90.+a, 42.50.Dv

Optical nonlinear processes can be divided into two categories: nonresonant coherent and resonant incoherent.[1] If a field frequency is well detuned from the atomic transition frequency, the response time is fast and the absorption loss is negligible, but the $\chi^{(3)}$ coefficient is usually small. On the other hand, when the field frequency is close to the atomic frequency, the $\chi^{(3)}$ coefficient is enhanced, but the response time becomes slow and the absorption loss is high. These tradeoffs between the $\chi^{(3)}$ coefficient and the response time or the absorption loss impose a serious limitation on both nonlinear optical switches in classical optics[2] and squeezed-state generation and quantum nondemolition measurement in quantum optics.[3] This Letter demonstrates the possibility of new optical nonlinearities based on the resonant and coherent coupling between the field and the atoms.

Suppose the pulse duration τ is much shorter than the energy and phase decay time constants T_1, T_2 and the area $A=(p_{21}/\hbar)\int E\,dt$ of a secant-hyperbolic shaped pulse is a multiple of 2π. Here p_{21} is the atomic dipole moment and E is the electric field envelope. The pulse propagates without being absorbed irrespective of the pulse frequency detuning from the atomic frequency and the inhomogeneous broadening of the atomic frequencies.[4] This is a so-called 2π soliton. The Maxwell-Bloch equations for self-induced transparency (SIT) can be solved by the inverse scattering method formula.[5] The 2π-soliton solution is uniquely determined by the complex eigenvalue $\xi=\alpha+i\beta$ of the inverse scattering (Zakharov-Shabat) equations.[5] The field envelope function for the 2π soliton, except for an $\exp[i(\omega_0 t-k_0 z)]$ term, is expressed as

$$E=\frac{2\hbar}{p_{21}}\frac{2\beta v\exp(i\Omega^2\alpha z/2v^2\kappa_0)\exp[-2i\alpha(vt-z)]\exp(i\phi_0)}{\cosh\{2\beta v[t-(z/v)(1+\Omega^2/4v^2\kappa_1)]\}}. \tag{1}$$

Here $\Omega^2=\omega n p_{21}^2/2\hbar\epsilon$, $\epsilon=\epsilon_0 n_0^2$ is the dielectric constant, n is the atomic density, $v=c/n_0$ is the nonresonant light velocity, $\alpha=(\omega-\omega_0)/2v$ represents the nonlinear frequency shift, and ω is a real soliton frequency. The pulse height β is proportional to the photon number N_p, i.e., $\beta=\omega p_{21}^2 N_p/16\hbar v^2\epsilon A_P$, and determines the pulse duration $\tau=1/2\beta v$. Here A_P is the cross-section area of the pulse. If the line-shape function of the atomic systems is assumed to be a delta function $\delta(\omega-\omega_{21})$, then the coefficients κ_0 and κ_1, the group velocity V, and the phase shift ϕ are written as

$$V\equiv\frac{v}{1+\Omega^2/4v^2\kappa_1}=\frac{v}{1+\Omega^2/[(\omega-\omega_{21})^2+1/\tau^2]} \tag{2}$$

and

$$\phi\equiv\frac{\Omega^2\alpha z}{2v^2\kappa_0}=\frac{\Omega^2(\omega-\omega_{21})z/v}{(\omega-\omega_{21})^2+1/\tau^2}. \tag{3}$$

The phase shift (3) represents the self-phase modulation of the 2π soliton, and the effective $\chi^{(3)}$ coefficient for self-phase modulation is evaluated by comparing (3) to the self-phase modulation of a normal rectangular pulse of duration τ,

$$\chi_{\text{self}}^{(3)}=\frac{p_{21}^4 n\tau^2}{16\hbar^3}\frac{\omega-\omega_{21}}{(\omega-\omega_{21})^2+1/\tau^2}. \tag{4}$$

$\chi_{\text{self}}^{(3)}$ is zero both on resonance $\omega=\omega_{21}$ and far from resonance $|\omega-\omega_{21}|\gg 1/\tau$. The maximum $\chi_{\text{self}}^{(3)}$ occurs at the optimum frequency detuning, $\Delta\omega_{\text{opt}}=|\omega-\omega_{21}|=1/\tau$, and is proportional to p_{21}^4, n, and τ^3.

If two 2π solitons collide, i.e., a slow soliton is overtaken by a fast soliton, the soliton phase is scattered according to[5]

$$\phi_2=2\left[\tan^{-1}\left(\frac{\beta_2+\beta_1}{\alpha_2-\alpha_1}\right)-\tan^{-1}\left(\frac{\beta_2-\beta_1}{\alpha_2-\alpha_1}\right)\right]. \tag{5}$$

The subscripts 1 and 2 refer to the two solitons. The self-phase modulation term (3) is not included in (5). If the frequency difference of the two solitons exceeds the inverse of the soliton pulse duration, $|\omega_1-\omega_2| \gg 1/\tau_1, 1/\tau_2$, the mutual-phase modulation $\partial\phi_2/\partial N_{p1} = p_{21}^2\omega/2(\omega_1-\omega_2)\hbar v\epsilon A_P$ is much larger than the other contributions $\partial\phi_2/\partial N_{p2}$, $\partial\phi_2/\partial\alpha_1$, and $\partial\phi_2/\partial\alpha_2$. The mutual-phase modulation is not dependent on the frequency detuning from ω_{12}, but is only dependent on the frequency difference $\omega_1-\omega_2$. The interaction length z_{eff}, in which the two 2π solitons pass through each other, is given by

$$Z_{\text{eff}} \equiv \frac{(1/\beta_1+1/\beta_2)}{v\,|1/V_1-1/V_2|} = \frac{2v}{\Omega^2}(\tau_1+\tau_2)\frac{[(\omega_1-\omega_{21})^2+1/\tau_1^2][(\omega_2-\omega_{21})^2+1/\tau_2^2]}{|(\omega_1-\omega_2)(\omega_1+\omega_2-2\omega_{21})+(1/\tau_1-1/\tau_2)(1/\tau_2+1/\tau_1)|}. \tag{6}$$

The mutual-phase modulation increases with decreasing $|\omega_1-\omega_2|$, but the interaction length also increases. The effective $\chi^{(3)}$ coefficient for mutual-phase modulation is evaluated by comparing $\partial\phi_2/\partial N_{p1}$ to the mutual-phase modulation between two normal pulses for the same interaction length z_{eff},

$$\chi^{(3)}_{\text{mutual}} = \frac{v\epsilon p_{21}^2\tau}{4\hbar^2\omega(\omega_1-\omega_2)z_{\text{eff}}}. \tag{7}$$

$\chi^{(3)}_{\text{mutual}}$ is not zero even on resonance $\omega_2=\omega_{21}$, where $\chi^{(3)}_{\text{self}}$ is zero. This is the important feature of SIT solitons for optical switching and quantum nondemolition measurement applications, because the phase shift is only dependent on the other soliton's photon number.

If the atomic system has finite T_1 and T_2 constants, the 2π soliton suffers from energy loss.[4] The soliton pulse duration τ has to be much smaller than $T_s=(2T_1^{-1}+T_2^{-1})^{-1}$ in order to allow the absorption loss to be small. When the energy loss is small, the energy-loss coefficient is approximated by

$$\alpha_{\text{eff}} = \frac{1}{6}\frac{n\omega p_{21}^2\tau^2}{\hbar v\epsilon T_s}. \tag{8}$$

Next let us consider the SIT solitons in the excitonic range of the spectrum in semiconductors. A free exciton propagating in the same direction as the light interacts coherently with the electric field of the light forming a mixed excitation of both, a so-called polariton. However, zero-dimensional bound excitons trapped by impurities cannot propagate, and they can be considered as static "atoms." In such a case, 2π solitons are produced very efficiently, because the bound excitons feature large dipole moments and long-decay-time constants.

An exciton bound to a neutral donor (the I_2 line) in CdS at 2 K has an absorption peak wavelength of $\lambda=487$ nm, a T_1 time constant of 135 ps, a T_2 time constant between 40 and 270 ps, an exciton density of $n=1\times10^{21}$ m^{-3}, and an oscillator strength of $f_0=26$,[6] that corresponds to the dipole moment $p_{21} \equiv e(f_0\hbar/2m_e\omega)^{1/2}=1\times10^{-28}$ Cm. When the beam radius r is 30 and 0.1 μm, the photon number and the peak power to produce the 2π soliton of 1-ps pulse duration are 5.0×10^8 and 200 W and 5×10^3 and 2 mW, respectively. In Fig. 1, the $\chi^{(3)}_{\text{self}}$ coefficients versus the frequency detuning $\omega-\omega_{21}$ are plotted. For the above mentioned CdS sample with pulse durations of $\tau=1$ and 3 ps the maximum $\chi^{(3)}_{\text{self}}$ values of 10^{-8} and $10^{-6.5}$ esu are obtained at $\Delta\omega_{\text{opt}}=10^{12}$ Hz and 3×10^{11} Hz. If the exciton density is increased to $n=1\times10^{23}$ m^{-3}, which is still well below the maximum density determined by the exciton Bohr radius, the maximum $\chi^{(3)}_{\text{self}}$ values are 10^{-6} and $10^{-4.5}$ esu with the pulse durations 1 and 3 ps, respectively. In Fig. 2, the $\chi^{(3)}_{\text{mutual}}$ coefficients versus the frequency difference $\omega_1-\omega_2$ of the two 2π solitons are plotted. Here we have assumed $\omega_2=\omega_{21}$. The $\chi^{(3)}_{\text{mutual}}$ coefficients are of the same order as the $\chi^{(3)}_{\text{self}}$ coefficients. From (8) the energy-loss coefficient is $\alpha_{\text{eff}}=1.39$ cm^{-1} for $\tau=1$ ps, $T_1=135$ ps, $T_2=270$ ps, and $n=10^{21}$ m^{-3}. The value often used as a figure of merit for $\chi^{(3)}/\alpha\tau$ for nonlinear materials is 10^4 esu cm/s.

Two-dimensional excitons in quantum wells, where the light is propagating perpendicular to the layer, can also be considered as static "atoms." An exciton in a 12-nm-thick GaAs quantum well at 2 K has an absorption peak wavelength of 806 nm, T_1 time constant of 180 ps, T_2 time constant of 12 ps, exciton density of $n=2\times10^{13}$ m^{-2}, and oscillator strength of 15.[7] The values are limited by the imperfections in GaAs crystal and interfaces. By solving the relations between f_0, T_1, T_2, and the

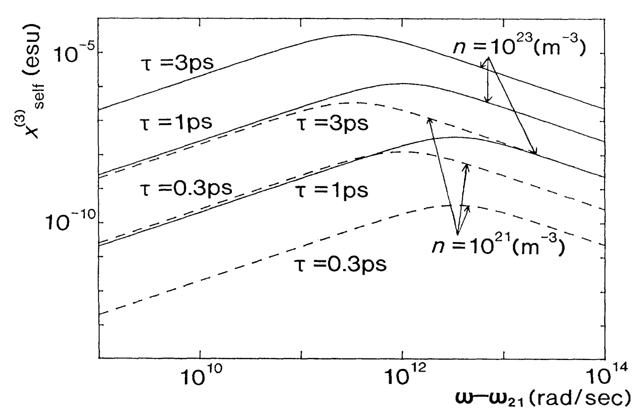

FIG. 1. The effective $\chi^{(3)}$ coefficient defined by the self-phase modulation for 2π solitons in CdS with the exciton density $n=10^{23}$ m^{-3} (solid line) and $n-10^{21}$ m^{-3} (dashed line) vs the angular frequency difference between the soliton frequency ω and the exciton frequency ω_{21}. The parameter is the pulse duration τ. $\chi^{(3)}$ is shown in terms of cgs units, which is obtained from Eq. (4) times 8.1×10^{18}.

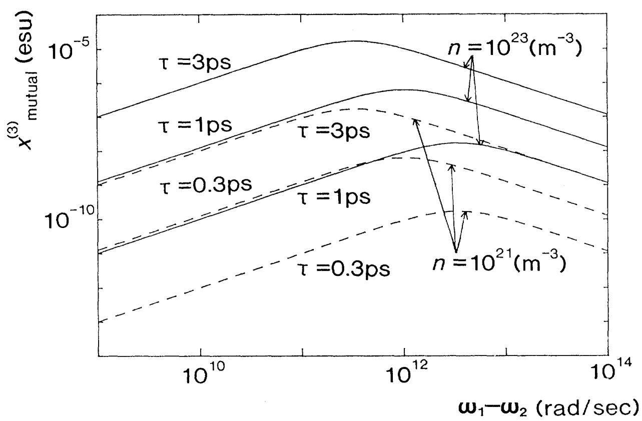

FIG. 2. The effective $\chi^{(3)}$ coefficient defined by the mutual-phase modulation in the collision of two 2π solitons in CdS with the exciton density $n=10^{23}$ m^{-3} (solid line) and $n=10^{21}$ m^{-3} (dashed line) vs the angular frequency difference between the signal soliton frequency ω_1 and the probe soliton frequency ω_2. The parameter is the pulse duration τ.

coherence area A_c (Ref. 8) self-consistently, we obtain $T_1=18$ ps, $T_2=36$ ps, and $f_0=108$ for a perfect sample.[7] The maximum $\chi^{(3)}$ coefficient is about $10^{-6.5}$ esu and the energy-loss coefficient is about 86.7 cm^{-1} for a 1-ps pulse.

Finally let us consider squeezed-state generation and quantum nondemolition measurement using 2π solitons. A quantum mechanical description of pulse propagation requires the definition of modes. One may employ either the Fourier modes defined by the wave number and the time or the local modes defined by the position and the time. However, neither set of modes are normal modes for the coupled Maxwell-Bloch equations. The pulse propagation in a two-level atomic system must be described by complicated coupled multimode equations, if these modes are employed.[9] A 2π soliton (or inverse scattering data, in general) is, on the other hand, a normal mode free from mode coupling. We can describe the propagation of a 2π soliton by a single-mode theory, if this normal mode is employed.[9] In other words, the mode function is given by $\text{sech}(t-z/V)$ and the quadrature component of this mode is detected by the homodyne measurement with the same time-dependent local oscillator pulse.

Suppose we excite the 2π soliton in a coherent state at $z=0$. The N_p dependence of the phase shown by (3) causes quantum phase spreading. Classically, the self-phase modulation and the dispersion balance exactly to form a soliton with a fixed phase. Quantum mechanically, a coherent-state soliton consists of linear superposition of different photon number states,[10] which have different phase velocities and therefore diffuse in phase space. This is similar to the "crescent squeezing" which would occur for a normal pulse in a Kerr medium.[11] The quadrature noise is calculated by the linearized quantum operator equation. At the optimum detuning $\Delta\omega_{\text{opt}}$ for the self-phase modulation, the minimum quadrature noise is expressed as

$$\langle \Delta\hat{a}_1^2 \rangle = \tfrac{1}{4}\exp(-p_{21}^2 n\omega\tau z/4\hbar\epsilon v)\,. \tag{9}$$

The pulse duration τ is defined for the average photon number $\langle \hat{N}_p \rangle$. If we assume the same numerical parameters used in Fig. 1, the quantum noise is reduced by 20 dB below the standard quantum limit at the distance of 400 and 4 μm for $n=10^{21}$ and 10^{23} m^{-3}, respectively, and $\tau=1$ ps. The energy loss is 5% for $T_1=135$ ps and $T_2=270$ ps. This is the most efficient and fastest squeezed-state generation scheme proposed so far.

Let us consider that the two 2π solitons are excited in coherent states at $z=0$ and with an appropriate time difference to achieve the collision. Since the probe soliton phase is shifted in proportion to the signal soliton photon number, the quantum nondemolition measurement of the signal photon number is made by detection of the probe phase shift.[12] The normalized measurement error of the signal photon number is written as

$$\frac{\langle \Delta N_{p1}^2 \rangle_{\text{meas}}}{\langle N_{p1} \rangle^2} = \frac{(\omega_2-\omega_1)^2\tau_1^2}{64\langle \hat{N}_{p2} \rangle}\,. \tag{10}$$

If we assume the same numerical parameters used in Fig. 2 and $\omega_1-\omega_2=\Delta\omega_{\text{opt}}$, $\tau_1=\tau_2=1$ ps, the measurement error $\langle \Delta\hat{N}_{p1}^2 \rangle_{\text{meas}}$ is about 20 dB below the quantum photon number noise $\langle \Delta\hat{N}_{p1}^2 \rangle = \langle \hat{N}_{p1} \rangle$. If $r=0.1$ μm, $n=10^{21}$ m^{-3}, $z_{\text{eff}}=400$ μm, and $\langle \hat{N}_{p2} \rangle = 5\times 10^3$ (2 mW), $\langle \Delta\hat{N}_{p1}^2 \rangle_{\text{meas}}^{1/2}$ is as small as 10^2. This is the most efficient quantum nondemolition measurement scheme proposed so far.

In conclusion, we have demonstrated the possibility of new optical nonlinearities using SIT solitons in the excitonic wavelength region in semiconductors. The propagation and the collision of 2π solitons give large $\chi^{(3)}$ coefficients with a very fast response time and an extremely small loss. These nonlinear coefficients are the largest among various coherent and fast nonlinear processes in semiconductors and organic materials.[1] The experimental difficulties of producing a squeezed state and demonstrating a quantum nondemolition measurement can be overcome by this new scheme.

The authors wish to thank Professor H. A. Haus of MIT, Professor E. Hanamura of the University of Tokyo, and Professor M. Yamanishi of Hiroshima University for sending unpublished papers on quantum solitons and excitonic nonlinearities. They are also indebted to Dr. T. Takagahara, Dr. H. Ando, and Dr. K. Igeta of Nippon Telegraph and Telephone Corporation for their critical discussions.

[a]On leave from the Max-Planck Institut für Festkörperforschung, Stuttgart, Federal Republic of Germany.

[1]For a recent review, see D. H. Auston *et al.*, Appl. Opt. **26**,

211 (1987).

[2]For a recent review, see IEEE J. Select. Areas Commun. **6**, No. 7 (1988).

[3]For a recent review on squeezed states, see J. Opt. Soc. Am B **4**, Oct. (1987). For a recent article on quantum nondemolition measurement, see N. Imoto, H. A. Haus, and Y. Yamamoto, Phys. Rev. A **32**, 2287 (1985).

[4]S. L. McCall and E. L. Hahn, Phys. Rev. **183**, 457 (1969); G. L. Lamb, Jr., Phys. Rev. Lett. **31**, 196 (1973); H. A. Haus, Rev. Mod. Phys. **51**, 331 (1979).

[5]V. E. Zakharov and A. B. Shabat, Zh. Eksp. Teor. Fiz. **61**, 118 (1972) [Sov. Phys. JETP **34**, 62 (1972)].

[6]M. Dagenais and W. F. Sharfin, J. Opt. Soc. Am. B **2**, 1179 (1985).

[7]A. Honold, L. Schultheis, J. Kuhl, and C. W. Tu, in Proceedings of the Sixteenth International Quantum Electronics Conference, Tokyo, Japan, July 1988 (to be published), paper No. ThB 6; A. Honold (to be published).

[8]J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. Foxon, and R. J. Elliott, Phys. Rev. Lett. **59**, 2337 (1987); E. Hanamura, in "Optical Switching in Low-Dimensional Systems," edited by H. Haug and L. Banyal, NATO Advanced Study Institute Series B, Vol. 194 (Plenum, New York, to be published); M. Yamanishi, *ibid.*

[9]K. Watanabe, H. Nakano, A. Honold, Y. Yamamoto, H. A. Haus, and Y. Lai, in Proceedings of the Fifth Symposium on Quantum Optics, Rotorua, New Zealand, February 1989 (Springer-Verlag, Berlin, to be published).

[10]D. Kaup, J. Math. Phys. **16**, 2036 (1975); L. D. Faddeev, Sov. Sci. Rev. Math. Phys. C **1**, 107 (1980); H. B. Thacker, Rev. Mod. Phys. **53**, 253 (1981); M. Wadachi and M. Sakagami, J. Phys. Soc. Jpn. **53**, 1933 (1984); Y. Lai and H. A. Haus (unpublished).

[11]M. Kitagawa and Y. Yamamoto, Phys. Rev. A **34**, 3974 (1986).

[12]Y. Yamamoto, S. Machida, K. Watanabe, K. Igeta, M. Ueda, and H. A. Haus, in Proceedings of the Eleventh International Conference on Atomic Physics, Paris, France, July 1988, edited by S. Haroche, J. C. Gay, and G. Grynberg (World Scientific, Singapore, to be published); H. A. Haus, K. Watanabe, and Y. Yamamoto, J. Opt. Soc. Am. B (to be published).