Exciton-exciton interaction and optical nonlinearity in biased semiconductor quantum wells

Tohya Hiroshima
Opto-Electronics Research Laboratories, NEC Corporation, 4-1-1 Miyazaki, Miyamae-ku, Kawasaki 213, Japan

Eiichi Hanamura
Department of Applied Physics, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113, Japan

Masamichi Yamanishi
Department of Physical Electronics, Hiroshima University, Saijo-cho, Higashi-Hiroshima 724, Japan
(Received 23 November 1987)

We investigate theoretically the excitonic optical nonlinearity in a semiconductor quantum well subject to an electrostatic field normal to the quantum-well plane, basing our calculation on the Hamiltonian of interacting bosons for the system of many quasi-two-dimensional excitons. It is pointed out that the dipole-dipole interaction between excitons plays an essential role in this optical nonlinearity. Numerical results of the third-order optical susceptibility for a GaAs quantum-well structure are also presented.

I. INTRODUCTION

The nonlinear-optical properties of exciton systems are due to the exciton-exciton interaction, i.e., the deviation from ideal boson behavior of excitons. For example, the exchange interaction between excitons plays an important role[1,2] in the recently discovered "optical Stark effect,"[3–5] which is an ultrafast dynamical blue shift and a bleaching of the exciton resonances in semiconductor quantum wells (QW's) irradiated by an intense laser beam in the transparency region below the absorption edge. Very recently one of the authors (M.Y.)[6] and Chemla *et al.*[7] have proposed independently a new mechanism of the ultrafast optical nonlinearity due to virtual (nonresonant) excitations in QW structures biased by an electrostatic field, in which the electrostatic screening field produced by the virtually induced static dipole moment modifies the optical properties of the QW material, as in the quantum-confined Stark effect.[8] When this new effect is viewed as an excitonic optical nonlinearity, it also relies on the anharmonic interaction between excitons. In the present case, the anharmonicity originates from the dipole-dipole interaction between excitons, which is enhanced under an electrostatic field normal to the QW plane. This is in marked contrast to the optical Stark effect. Furthermore, the present optical nonlinearity accompanies electrostatic screening due to virtually induced polarization through exciton states.

The aim of this paper is to present a rigorous description of this electrostatic-field-induced optical nonlinearity, taking into account the excitonic effect. The excitonic effect was not treated fully in Refs. 6 and 7, since the mutual interaction between excitons was not considered. This excitonic effect must be included when the off-resonance energy is of the same order of or less than the exciton binding energy. To be specific, considerations are focused only on the stationary properties of this effect. Furthermore, the electron spin is disregarded initially, although it is included in the final expressions. Based on the Hamiltonian of interacting bosons for the system of many quasi-two-dimensional excitons, we derive the nonresonant third-order optical susceptibility $\chi^{(3)}(\omega;\omega,-\omega,\omega)$ for degenerate four-wave mixing from first principles. The numerical results are also presented for a GaAs QW.

II. THEORY

First of all, let us consider the system of interacting quasi-two-dimensional excitons in a semiconductor QW. The QW considered here is a single QW layer within a multi-QW structure with well thickness L_z and barrier thickness L_B along the z direction, so that the quantization length of this direction is $L=L_z+L_B$. The Hamiltonian of the system of quasi-two-dimensional excitons in the boson space is given by

$$H=\sum_{\nu}\sum_{\mathbf{K}_\parallel}\varepsilon_\nu(\mathbf{K}_\parallel)C^\dagger_{\nu\mathbf{K}_\parallel}C_{\nu\mathbf{K}_\parallel}+\frac{1}{2V}\sum_{\nu_1,\nu_2,\nu_3,\nu_4}\sum_{\mathbf{Q}}\sum_{\mathbf{K}_\parallel\mathbf{K}'_\parallel}V^{\nu_4,\nu_3}_{\nu_1,\nu_2}(\mathbf{Q},\mathbf{K}_\parallel,\mathbf{K}'_\parallel)\times C^\dagger_{\nu_4\mathbf{K}_\parallel+\mathbf{q}_\parallel}C^\dagger_{\nu_3\mathbf{K}'_\parallel-\mathbf{q}_\parallel}C_{\nu_2\mathbf{K}'_\parallel}C_{\nu_1\mathbf{K}_\parallel}\ , \tag{1}$$

where $V=LS$ with the quantization area S in the QW plane, and $C^\dagger_{\nu\mathbf{K}_\parallel}$ ($C_{\nu\mathbf{K}_\parallel}$) is the creation (annihilation) operator for an exciton with quantum number ν, translational momentum $\hbar\mathbf{K}_\parallel$ in the QW plane, and energy $\varepsilon_\nu(\mathbf{K}_\parallel)$. This operator obeys boson commutation relations. The quantum number ν represents both the internal state for the electron-hole relative motion ($1s,2s,\ldots$) and the indices of electron and hole subbands (n_e,n_h).

The exciton-exciton interaction with the wave-vector transfer $\mathbf{Q}=(\mathbf{q}_{\parallel},q_z)$ is described by $V^{\nu_4,\nu_3}_{\nu_1,\nu_2}(\mathbf{Q},\mathbf{K}_{\parallel},\mathbf{K}'_{\parallel})$. The Hamiltonian [Eq. (1)] is nothing but the quasi-two-dimensional version of the many-exciton Hamiltonian in the bulk crystal.[9,10]

The term $V^{\nu_4,\nu_3}_{\nu_1,\nu_2}(\mathbf{Q},\mathbf{K}_{\parallel},\mathbf{K}'_{\parallel})$ is composed of two parts; the purely Coulombic interaction, $f^{\nu_4,\nu_3}_{\nu_1,\nu_2}(\mathbf{Q})$, and the interaction which contains the exchange of constituent particles of excitons. Under an electrostatic field normal to the QW plane, the purely Coulombic interaction is enhanced due to the distortion of the exciton envelope function along the z direction, and it plays an essential role in the electrostatic-field-induced optical nonlinearity considered in this paper. Therefore, the exchange term is skipped. When the wave-vector transfer $\mathbf{Q}$ is small, $f^{\nu_4,\nu_3}_{\nu_1,\nu_2}(\mathbf{Q})$ is written as follows:

$$f^{\nu_4,\nu_3}_{\nu_1,\nu_2}(\mathbf{Q})=V(\mathbf{Q})\sum_{\mathbf{p},\mathbf{p}'}\sum_{P_z,P'_z}\sum_{p_z,p'_z}[\psi^*_{\nu_4}(\mathbf{p}-\alpha\mathbf{q}_{\parallel};P_z-q_z,p_z)-\psi^*_{\nu_4}(\mathbf{p}+\beta\mathbf{q}_{\parallel};P_z,p_z+q_z)]$$
$$\times[\psi^*_{\nu_3}(\mathbf{p}'+\alpha\mathbf{q}_{\parallel};P'_z+q_z,p'_z)-\psi^*_{\nu_3}(\mathbf{p}'-\beta q_{\parallel};P'_z,p'_z-q_z)]\psi_{\nu_2}(\mathbf{p}',P'_z,p'_z)\psi_{\nu_1}(\mathbf{p};P_z,p_z)\ , \quad (2)$$

where $V(\mathbf{Q})=4\pi e^2/(\epsilon_s\mathbf{Q}^2)$, and ϵ_s is the static dielectric constant, $\alpha=m_h/(m_e+m_h)$, and $\beta=1-\alpha$ with m_e (m_h) the in-plane effective mass of an electron (hole). The function $\psi_\nu(\mathbf{k};P_z,p_z)$ is the Fourier transformation of the ν-exciton envelope function, $\Psi_\nu(\mathbf{r};z_e,z_h)$;

$$\psi_\nu(\mathbf{k};P_z,p_z)=\frac{1}{L\sqrt{S}}\int d^2r\int dz_e\int dz_h\Psi_\nu(\mathbf{r};z_e,z_h)\exp(-i\mathbf{k}\cdot\mathbf{r}-iP_zz_e+ip_zz_h)\ ,$$

where z_e (z_h) is the coordinate of the electron (hole) along the z direction (normal to the QW plane), and $\mathbf{r}$ is the relative coordinate of the electron-hole pair (exciton) in the QW plane. As mentioned later, the relevant translational wave vectors of excitons are zero so that $\mathbf{q}_{\parallel}=0$. Furthermore, we take $q_z\to 0$ in the spirit of the mean-field approximation. At that time, Eq. (2) reduces to the following equation:

$$f^{\nu_4,\nu_3}_{\nu_1,\nu_2}(0)=\frac{4\pi}{\epsilon_s}P^{\nu_4}_{\nu_1}P^{\nu_3}_{\nu_2}\ , \quad (3)$$

where $P^{\nu}_{\nu'}=e\langle\Psi_\nu|z_h-z_e|\Psi_{\nu'}\rangle$, the dipole moment which is induced along the z direction through ν- and ν'-exciton states elongated along this direction. The interaction term with $\nu_1=\nu_2=\nu_3=\nu_4=(1,1;1s)$ is dominant and represents the dipole-dipole interaction between two excitons in the lowest state ($1s$) for the electron-hole relative motion in the lowest subbands ($n_e=1$, $n_h=1$) of electron and hole. This term vanishes in the symmetric QW structures without the electrostatic field. On the other hand, the interaction terms with different ν's, e.g., the terms with $\nu_1=\nu_2=(1,1;1s)$ and $\nu_3=\nu_4=(1,1;2p)$, are finite even without the field but are much smaller than the dominant one.

On the basis of the Hamiltonian [Eq. (1)] and the effective exciton-exciton interaction [Eq. (3)], we consider a semiconductor QW subject to an electrostatic field F along the z direction and a classical monochromatic laser field $E_p^*\exp(i\omega_p t)+E_p\exp(-i\omega_p t)$ in the transparency region below the lowest exciton resonance. The laser field is assumed to be polarized on the QW plane. The exciton-laser field interaction is described in the dipolar approximation. Furthermore, the rotating-wave approximation is imposed. The total effective Hamiltonian is thus given by

$$\tilde{H}=\sum_\nu e_\nu C^\dagger_\nu C_\nu+\frac{2\pi}{V\epsilon_s}\sum_{\nu_1,\nu_2,\nu_3,\nu_4}P^{\nu_4}_{\nu_1}P^{\nu_3}_{\nu_2}C^\dagger_{\nu_4}C^\dagger_{\nu_3}C_{\nu_2}C_{\nu_1}$$
$$-\sum_\nu(\mu_\nu E_p^*C_\nu+\text{H.c.})\ , \quad (4)$$

where translational wave vectors are dropped from exciton operators because relevant translational wave vectors are zero in the present configuration. In Eq. (4), $e_\nu=\varepsilon_\nu(\mathbf{K}_{\parallel}=\mathbf{0})-\hbar\omega_p$, the off-resonance energy, and μ_ν is the transition dipole momentum between the ν exciton and the ground state;

$$\mu_\nu=\frac{\sqrt{S}e\hbar M_{\mathrm{CV}}}{m_0\varepsilon_\nu(0)}\int dz\Psi_\nu(0;z,z)\ ,$$

where m_0 is the free-electron mass and M_{CV} is the optical matrix element between the valence and conduction bands.[11]

The ground state of the system of coherently pumped excitons described by the Hamiltonian [Eq. (4)] is written by $U|0\rangle$, where $|0\rangle$ is the vacuum of the exciton system, and U is a unitary operator. We assume that U has the following form:

$$U=\exp\left[\sum_\nu(x_\nu C^\dagger_\nu-x^*_\nu C_\nu)\right]\ ,$$

where x_ν is the variational parameter (c number) determined by the requirement that $\tilde{E}=\langle 0|U^{-1}\tilde{H}U|0\rangle$ should be minimized. From $\partial\tilde{E}/\partial x^*_\nu=0$, the following equation is obtained:

$$e_\nu x_\nu + \sum_{\nu'} F_{\nu\nu'} x_{\nu'} - \mu_\nu E_p = 0 \ , \tag{5}$$

where

$$F_{\nu\nu'} = \frac{4\pi}{V\epsilon_s} P^\nu_{\nu'} \sum_{\nu_1,\nu_2} P^{\nu_2}_{\nu_1} x^*_{\nu_2} x_{\nu_1} \ . \tag{6}$$

Here μ_ν and $P^\nu_{\nu'}$ are assumed to be real without loss of generality. In this case, $P^{\nu'}_\nu = P^\nu_{\nu'}$. Equation (6) is also rewritten as

$$F_{\nu\nu'} = \frac{4\pi}{V\epsilon_s} P^\nu_{\nu'} \sum_{\nu_1,\nu_2} \langle 0 \mid U^{-1}(P^{\nu_2}_{\nu_1} C^\dagger_{\nu_2} C_{\nu_1}) U \mid 0 \rangle \equiv F_d P^\nu_{\nu'} \ . \tag{7}$$

This expression represents the interaction energy between the dipole moment $P^\nu_{\nu'}$ and the depolarization field F_d which partially screens the applied electrostatic field. This means that the real electrostatic field which works along the z direction is not F, but $F+F_d$. Therefore, e_ν and μ_ν in Eq. (5) should be evaluated at $F+F_d$. To solve Eq. (5) by the iteration in E_p, e_ν and μ_ν are expanded in F_d;

$$e_\nu = e^{(0)}_\nu + F_d(\partial e_\nu/\partial F) \ , \tag{8a}$$

$$\mu_\nu = \mu^{(0)}_\nu + F_d(\partial \mu_\nu/\partial F) \ . \tag{8b}$$

Substituting Eqs. (8a) and (8b) into Eq. (5), $x_\nu = x^{(1)}_\nu + x^{(3)}_\nu + \cdots$ is obtained as follows:

$$x^{(1)}_\nu = \frac{E_p \mu^{(0)}_\nu}{e^{(0)}_\nu} \ , \tag{9a}$$

$$x^{(3)}_\nu = \frac{E_p F_d}{e^{(0)}_\nu} \left[-\sum_{\nu'} P^\nu_{\nu'} \frac{\mu^{(0)}_{\nu'}}{e^{(0)}_{\nu'}} - \frac{\mu^{(0)}_\nu}{e^{(0)}_\nu} \frac{\partial e_\nu}{\partial F} + \frac{\partial \mu_\nu}{\partial F} \right] , \tag{9b}$$

where

$$F_d = \frac{4\pi}{\epsilon_s V} \mid E_p \mid^2 P_0 \ , \tag{10}$$

and

$$P_0 = 2 \sum_{\nu,\nu'} \frac{\mu^{(0)}_\nu \mu^{(0)}_{\nu'}}{e^{(0)}_\nu e^{(0)}_{\nu'}} P^{\nu'}_\nu \ . \tag{11}$$

In Eq. (11), the factor 2 in front of the summation is the spin weight. The polarization density P driven by the laser field is given by

$$P = \frac{2}{V} \sum_\nu \mu_\nu \langle 0 \mid U^{-1} C_\nu U \mid 0 \rangle = \frac{2}{V} \sum_\nu \mu_\nu x_\nu \ . \tag{12}$$

From Eqs. (8b) and (9), P is calculated as a function of E_p, from which the third-order optical susceptibility $\chi^{(3)}(\omega;\omega,-\omega,\omega)$ is obtained as follows:

$$\chi^{(3)}(\omega;\omega,-\omega,\omega) = \frac{4\pi P_0}{\epsilon_s V^2} \left[-P_0 - 2 \sum_\nu \left[\frac{\mu^{(0)}_\nu}{e^{(0)}_\nu} \right]^2 \frac{\partial e_\nu}{\partial F} + 4 \sum_\nu \frac{\mu^{(0)}_\nu}{e^{(0)}_\nu} \frac{\partial \mu_\nu}{\partial F} \right] , \tag{13}$$

where P_0 is defined in Eq. (11). Note that $\chi^{(3)}$ is independent of S. Also it is noted that the effect of damping is neglected in Eq. (13). This is justified as long as the off-resonance energy is much larger than the linewidth due to the transverse relaxation of the exciton. The third-order optical susceptibility $\chi^{(3)}$ is composed of three terms; the first term represents the saturation effect through the exciton-exciton interaction, which is always negative. On the other hand, the second term and the third term correspond, respectively, to the effect of exciton level shifts and that of the transition dipole moment changes due to the depolarization field F_d. It should be noted that, for small off-resonances, $\chi^{(3)}$ is determined mainly by the first and the second terms.

III. NUMERICAL RESULTS

In this section, the numerical results are presented for a GaAs QW structure, consisting of a GaAs well layer and $Al_{0.6}Ga_{0.4}As$-clad layers. Both the QW thickness L_z and the barrier thickness L_B are set to be 100 Å. In the numerical estimation, it is sufficient to retain the lowest $(1,1;1s)$ exciton state as long as the off-resonance energy is of the same order of or less than the exciton binding energy, although the theory is not restricted to this case. The envelope function for the $(1,1;1s)$ exciton state is written variationally:

$$\Psi(\mathbf{r};z_e,z_h) = \gamma(2/\pi)^{1/2} e^{-\gamma r} f_e(z_e) f_h(z_h) \ ,$$

where $r = \mid \mathbf{r} \mid$, $f_{e(h)}(z)$ is the electron (hole) envelope function, and γ is the variational parameter in the trial function used in the variational calculation of the exciton binding energy.[11] The numerical parameters used in the calculations are the same as cited in Ref. 11, except for the static dielectric constant of the QW material (GaAs): $\epsilon_s = 12.4$.

First of all, let us present the electrostatic-field dependence of $P^{(1,1;1s)}_{(1,1;1s)}$, which determines almost the whole

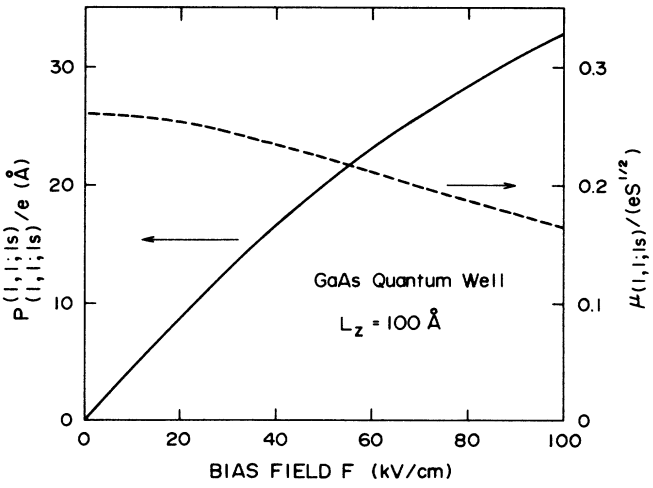

FIG. 1. The electrostatic-bias-field dependence of $P^{(1,1;1s)}_{(1,1;1s)}e$ (solid line) and $\mu_{(1,1;1s)}/e\sqrt{S}$ (dashed line) in a GaAs(100 Å) quantum-well structure.

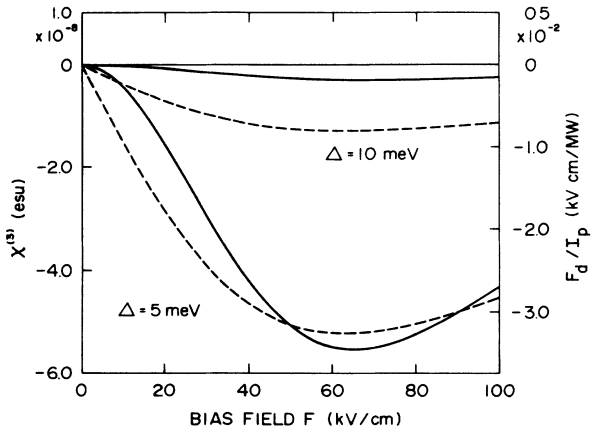

FIG. 2. The electrostatic-bias-field dependence of $\chi^{(3)}$ (solid lines) and F_d/I_p (dashed lines) for two off-resonance energies $\Delta=5$ and 10 meV, in a GaAs(100 Å)-$Al_{0.6}Ga_{0.4}As$(100 Å) quantum-well structure.

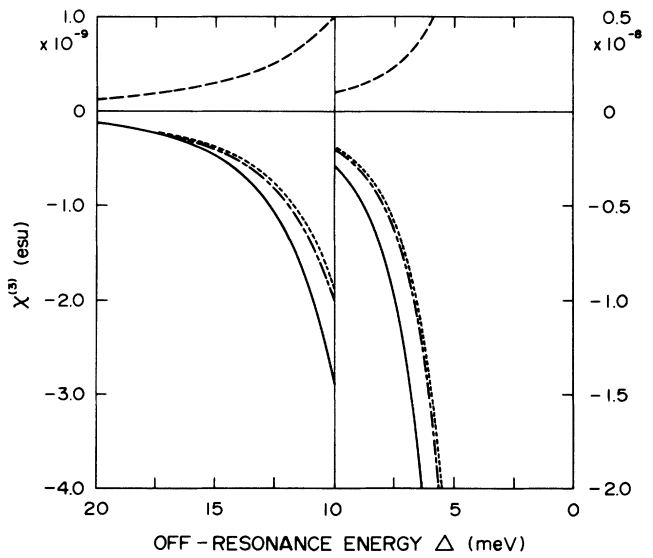

FIG. 3. The off-resonance energy dependence of $\chi^{(3)}$ (solid line) for an electrostatic bias field of 60 kV/cm in a GaAs(100 Å)-$Al_{0.6}Ga_{0.4}As$(100 Å) quantum-well structure. The contributions of the first term, the second term, and the third term of $\chi^{(3)}$ [Eq. (13)] are, respectively, denoted by the dot-dashed line (—·—·—·), the dotted line (· · · ·), and the dashed line (— — —).

effect. The calculated results are shown in Fig. 1 (the solid line). As a reference, the electrostatic-field dependence of $\mu_{(1,1;1s)}$ is also included by the dashed line in this figure. The application of the electrostatic field normal to the QW plane causes the distortion of electron and hole envelope functions along this direction; the positive and negative charges are pushed against opposite walls of the QW. This results in the increase of $P^{(1,1;1s)}_{(1,1;1s)}$ but the decrease of $\mu_{(1,1;1s)}$, which is proportional to the overlap between electron and hole envelope functions. Figure 2 shows the electrostatic-bias-field dependence of $\chi^{(3)}$ and F_d/I_p, the induced depolarization field normalized by the laser field intensity I_p, for two off-resonance energies, $\Delta=5$ and 10 meV, with respect to the lowest exciton resonance. The laser field intensity I_p is given by $I_p=cn\,|E_p|^2/(2\pi)$ where n is the refractive index and assumed to be 3.5 in the actual calculation. It is apparent from Fig. 2 that $|\chi^{(3)}|$ and $|F_d/I_p|$ reach a maximum around $F=60$ kV/cm, and there is no quadratic dependence of $\chi^{(3)}$ on F, except for $F\lesssim 20$ kV/cm. This suggests that the numerical estimation of $\chi^{(3)}$ with an assumption of quadratic dependence reported by Chemla *et al.*[7] is no longer valid. The decrease of $|\chi^{(3)}|$ and $|F_d/I_p|$ under large F ($\gtrsim 60$ kV/cm) comes from the decrease of $\mu_{(1,1;1s)}$ with increasing F as shown in Fig. 1. Figure 3 shows the dependence of $\chi^{(3)}$ on the off-resonance energy Δ. The electrostatic bias field is set to be 60 kV/cm. For an off-resonance energy $\Delta=5$ meV, the absolute value of $\chi^{(3)}$ is about 5.5×10^{-8} esu, which is large enough to be observed. Under smaller off-resonance $\chi^{(3)}$ is mainly determined by the first and the second terms of Eq. (13), both of which behave as Δ^{-4}, and the contribution of the third term, which behaves as Δ^{-3}, is not so large. Although Δ must be much larger than Γ, the linewidth due to the transverse relaxation of the exciton, this condition is satisfied even for Δ of a few meV at low temperature, because it has been found that Γ is below 1 meV at low temperature ($\lesssim 50$ K).[12]

IV. CONCLUSIONS

In conclusion, we have formulated the excitonic optical nonlinearity in electrostatic-field-biased semiconductor QW's. The third-order optical susceptibility $\chi^{(3)}$ is composed of two parts; the saturation effect due to the dipole-dipole interaction between excitons and the effect of the depolarization field. These effects depend on the distortion of exciton envelope functions due to the application of the electrostatic field normal to the QW layer. This means that suitable QW designing would further enhance $\chi^{(3)}$ in the present mechanism, offering important applications in optical and optoelectronic devices. Such an exploration has just started.[13]

ACKNOWLEDGMENTS

The authors thank Dr. Roy Lang for his continual encouragement throughout this work. Two of the authors (E.H. and M.Y.) acknowledge financial support by a Scientific Research Grant-In-Aid for Specially Promoted Research from the Ministry of Education, Science and Culture of Japan.

[1] S. Schmitt-Rink and D. S. Chemla, Phys. Rev. Lett. **57**, 2752 (1986).

[2] S. Schmitt-Rink, D. S. Chemla, and H. Haug, Phys. Rev. B **37**, 941 (1988).

[3] A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, and H. Morkoç, Phys. Rev. Lett. **56**, 2748 (1986).

[4] A. Von Lehmen, D. S. Chemla, J. E. Zucker, and J. P. Heritage, Opt. Lett. **11**, 609 (1986).

[5]K. Tai, J. Hegarty, and W. T. Tsang, Appl. Phys. Lett. **51**, 152 (1987).
[6]M. Yamanishi, Phys. Rev. Lett. **59**, 1014 (1987).
[7]D. S. Chemla, D. A. B. Miller, and S. Schmitt-Rink, Phys. Rev. Lett. **59**, 1018 (1987).
[8]D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Phys. Rev. Lett. **53**, 2173 (1984).
[9]E. Hanamura, J. Phys. Soc. Jpn. **29**, 50 (1970).
[10]E. Hanamura, J. Phys. Soc. Jpn. **37**, 1545 (1974).
[11]T. Hiroshima and K. Nishi, J. Appl. Phys. **62**, 3360 (1987).
[12]L. Schultheis, A. Honold, J. Kuhl, K. Köhler, and C. W. Tu, Phys. Rev. B **34**, 9027 (1986).
[13]M. Yamanishi and M. Kurosaki, IEEE J. Quantum Electron. **QE-24**, 325 (1988); Superlatt. Microstruct. (to be published).