Calculation of the second-order optical nonlinear susceptibilities in biased $Al_xGa_{1-x}As$ quantum wells

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The second-order optical nonlinear susceptibilities $\chi^{(2)}$ of biased $Al_xGa_{1-x}As$ quantum wells (QW's) are theoretically estimated. The contributions of Wannier excitonic states and nonexcitonic excited states to $\chi^{(2)}$ are discussed. It is shown that the transitions between a Wannier excitonic state and nonexcitonic excited states contribute dominantly to $\chi^{(2)}$ of biased $Al_xGa_{1-x}As$ QW's. The $\chi^{(2)}$ values are calculated for various QW widths and electric fields. The strong resonance of exciton energy and pumping-light energy, the strong oscillator strength of Wannier exciton states, and the large dipole of Wannier excitons result in large $\chi^{(2)}$ values greater than 5×10^{-10} m/V for QW's with well widths of less than 70 Å. Because QW's have large $\chi^{(2)}$ values in the wavelength range near 1.55 μm, they are good candidates for nonlinear optical devices for communication systems.

I. INTRODUCTION

In the 1.3–1.55 μm optical telecommunications wavelength range, powerful wavelength conversion and state squeezing functions[1] can be performed using second-order nonlinear effects. To realize such a device, materials with large second-order optical nonlinear susceptibilities $\chi^{(2)}$ are required.

Asymmetric quantum-well (QW) structures of III-V compound semiconductors like biased QW's are expected to have $\chi^{(2)}$ two orders of magnitude larger than those of bulk III-V compound semiconductors.[2] Bulk III-V compound semiconductors have some of the largest reported $\chi^{(2)}$.[3] Although III-V compound semiconductors are not suitable for second-harmonic generation (SHG) applications due to their optical loss for photons with energy greater than the band-gap energy, they can be used for wavelength conversion devices or parametric amplification with pumping light near the band-gap energy and signal light at half the band-gap energy where the semiconductor is almost transparent. In addition to a possible large $\chi^{(2)}$, III-V compound semiconductor QW's can be used as the core of a waveguide structure with tight optical confinement. Further, growing the waveguide and pump laser on the same substrate is very attractive. Strong optical-confinement waveguide structure and integration with a pumping laser allow the high optical power density indispensable to nonlinear optical effects. In the case of bulk III-V semiconductors, it is difficult to achieve the phase-matching condition because of the crystal symmetry. However, with QW's, a pseudo-phase-matching structure[4] can easily be constructed by application of a periodic electric field.

In spite of these attractive features, the $\chi^{(2)}$ of biased QW structures in the wavelength range around 1 μm has not been thoroughly investigated. Several experiments and discussions[5,6] at the wavelength of 10 μm using subband states in the conduction band of QW's have reported large values of $\chi^{(2)}$ on the order of 1.0×10^{-9} m/V. Unfortunately, these values cannot be used in the 1-μm wavelength range, because neither the pump light nor the signal light is resonant with the subband energy. Khurgin[7] discussed the $\chi^{(2)}$ of various asymmetric $Al_xGa_{1-x}As$ QW's theoretically in the case when the photon energy of the beam is resonant with the interband transition energy, and showed that $\chi^{(2)}$ was of the same order of magnitude as for bulk GaAs. On the other hand, Shimizu[2] pointed out that the Wannier exciton states, which were not considered in Khurgin's calculation, play a large role in second-order nonlinear optical phenomena, and estimated a large $\chi^{(2)}$ of 8×10^{-6} esu (3×10^{-9} m/V), even when QW width and electric field were not optimized. As the oscillator strength of the Wannier excitons depends on well width and electric field, optimization of the well width and the electric field will yield significant improvements in $\chi^{(2)}$.

In this paper, we calculate the $\chi^{(2)}$ of $Al_xGa_{1-x}As$ QW's for signal light of wavelength 1.55 μm and pumping light near 0.78-μm wavelength. The transition between the Wannier exciton state and nonexcitonic excited states is shown to be dominant among the various transitions contributing to $\chi^{(2)}$. We optimize the electric field for various QW widths and obtain large $\chi^{(2)}$ values for small QW widths with relatively high applied electric field. The values are shown to be sufficient for use in nonlinear optical devices such as parametric amplifiers in the optical communication wavelength range.

II. THEORETICAL MODEL OF $\chi^{(2)}$ IN QW's

We consider the situation of parametric amplification or difference-frequency generation in the wavelength range near 1 μm. To use the pump beam efficiently, its photon energy is selected near the resonance of the interband transition but several meV below the resonance of the Wannier exciton. If the QW is transparent for all

waves (pumping wave ω, signal wave ω_s, and idler wave ω_i), overall permutation symmetry holds and the $\chi^{(2)}$ of difference-frequency generation is the same as that of sum-frequency generation. According to nonlinear-response theories, the microscopic expression of $\chi^{(2)}_{ijk}(\omega;\omega_s,\omega_i)$, where i, j, and k are the polarization direction of pumping wave, signal wave, and idler wave, is given as[8]

$$\chi^{(2)}_{ijk}(\omega;\omega_s,\omega_i)=-\frac{N_z q^3}{\epsilon_0\hbar^2}\sum_{n1,n2\neq g}\frac{\langle g|r_i|n1\rangle\langle n1|r_j|n2\rangle\langle n2|r_k|g\rangle}{(\omega-\omega_{n1-g}+i\Gamma)(\omega_i-\omega_{n2-g}+i\Gamma)}+(j,\omega_s)\Longleftrightarrow(k,\omega_i)\ . \tag{1}$$

Here N_z is the number of wells per unit length in the direction perpendicular to the QW plane, ϵ_0 is the permittivity in vacuum, and q is the charge of the electrons. $|g\rangle$ is the ground state, $|n1\rangle$ and $|n2\rangle$ are excited states, and $1/\Gamma_{nn'}$ is the relaxation time between states n and n'. We assume $\Gamma_{nn'}=\Gamma_{n'n}=\Gamma$. $(j,\omega_s)\Longleftrightarrow(k,\omega_i)$ means a term with k exchanged for j and ω_s for ω_i. Nonresonant terms are ignored.

Equation (1) is derived under the assumption that the population in the ground state $\rho^{(0)}_{gg}$ equals unity, and that in the excited states $\rho^{(0)}_{nn}$ equals zero. Even at room temperature, this assumption is a good approximation in our case, because the transition energy from the ground state to the excited states, $E_{\rm tr}$, which is of the same order as the band-gap energy (≥ 1.5 eV), is 50 times larger than the room-temperature thermal energy kT (≈ 30 meV) and

$$\rho^{(0)}_{nn}/\rho^{(0)}_{gg}\approx\exp(-E_{\rm tr}/kT)\approx 2\times 10^{-22}\approx 0\ ,$$

which means $\rho^{(0)}_{gg}\approx 1$.

Combinations of $|n1\rangle$ and $|n2\rangle$ are classified into three categories according to the relation with Wannier excitonic states.

(a) The contribution of transitions between nonexcitonic excited states. In the first case examined, both $|n1\rangle$ and $|n2\rangle$ are nonexcitonic excited states. Equation (1) may be written as

$$\chi^{(2)}_{ijk}(\omega;\omega_s,\omega_i)_{\text{band-band}}=-\frac{N_z q^3}{\epsilon_0\hbar^2}\sum_{\mathbf{k}_\parallel}\frac{\langle {\rm hh}(\mathbf{k}_\parallel)|r_i|c_1(\mathbf{k}_\parallel)\rangle\langle c_1(\mathbf{k}_\parallel)|r_j|c_2(\mathbf{k}_\parallel)\rangle\langle c_2(\mathbf{k}_\parallel)|r_k|{\rm hh}(\mathbf{k}_\parallel)\rangle}{[\omega-\omega_{\text{hh-}c1}(\mathbf{k}_\parallel)+i\Gamma][\omega_1-\omega_{n2-g}(\mathbf{k}_\parallel)+i\Gamma]}\ , \tag{2}$$

where $|{\rm hh}(\mathbf{k}_\parallel)\rangle$ is the lowest subband state of the heavy-hole band for a wave vector $\mathbf{k}_\parallel$ in the plane of the QW, while $|c_1(\mathbf{k}_\parallel)\rangle$ and $|c_2(\mathbf{k}_\parallel)\rangle$ are the lowest and the second-lowest subband states. A dipole transition between $|c_2(\mathbf{k}_\parallel)\rangle$ and $|{\rm hh}(\mathbf{k}_\parallel)\rangle$ is possible when an electric field is applied, because the symmetry of the wave function is broken. Khurgin[7] calculated $\chi^{(2)}$ for this transition and the transition between hole subbands. A value of $\chi^{(2)}=6\times 10^{-12}$ m/V ($d_{13}=1.2\times 10^{-11}$ m/V) was estimated for a 50-Å QW. This value is almost the same as that of bulk GaAs.[3] Using the resonance transition only at the point $\mathbf{k}_\parallel=\mathbf{0}$ and a large 75-meV detuning between pump photon energy and interband energy to avoid band-tail absorption results in this discouragingly small value. As a result, we cannot decrease the denominator $[\omega-\omega_{\text{hh-}c1}(\mathbf{k}_\parallel)+i\Gamma]$ of Eq. (2). Even if the band-tail absorption can be ignored in applications such as parametric amplification, the detuning cannot be decreased to less than 15 meV because it is necessary to avoid the absorption of Wannier excitons whose binding energy is about 10 meV. This decrease of the detuning increases $\chi^{(2)}$ by a factor of 5. However, this value is one order of magnitude smaller than the process which includes Wannier exciton states, as shown later.

(b) The contribution of transitions between Wannier exciton states. In the second case considered both $|n1\rangle$ and $|n2\rangle$ are Wannier exciton states.

$$\chi^{(2)}_{ijk}(\omega;\omega_s,\omega_i)_{\text{ex-ex}}=-\frac{N_z q^3}{\epsilon_0\hbar^2}\frac{\langle g|r_i|\Psi_{\rm ex1}\rangle\langle\Psi_{\rm ex1}|r_j|\Psi_{\rm ex2}\rangle\langle\Psi_{\rm ex2}|r_k|g\rangle}{(\omega-\omega_{\text{ex1-}g}+i\Gamma)(\omega_i-\omega_{\text{ex2-}g}+i\Gamma)}\ . \tag{3}$$

The Wannier exciton states in a QW are written as the linear combination of the product of a free-electron and a hole state,[9–11]

$$\Psi_{\rm ex}=\sum_{\mathbf{k}_\parallel}A(\mathbf{k}_\parallel)\varphi_e(z_e)u_c(\mathbf{r}_e)\varphi_h(z_h)u_v(\mathbf{r}_h)e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\ , \tag{4}$$

where $u_c(\mathbf{r}_e)$ and $u_v(\mathbf{r}_h)$ are the periodic parts of the band-edge Bloch functions of the conduction band, primarily composed of s-state functions, and of the valence band, composed of p-state functions. $\varphi_e(z_e)$ and $\varphi_h(z_h)$ are confined-state envelope functions for the direction z, perpendicular to the QW plane. $A(\mathbf{k}_\parallel)$ is the Fourier transform of the in-plane envelope wave function of the exciton, $\Phi_{\rm ex}(\mathbf{r}_\parallel)$, and is given as

$$A(\mathbf{k}_\parallel)=\int\int d^2\mathbf{r}_\parallel e^{-i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\Phi_{\rm ex}(\mathbf{r}_\parallel)\ . \tag{5}$$

We take

$$\Phi_{ex}(\mathbf{r}_\parallel)=\left[\frac{2}{\pi}\right]^{1/2}\frac{1}{\lambda_{ex}}e^{-|\mathbf{r}_\parallel|/\lambda_{ex}} . \tag{6}$$

The two-dimensional exciton radius λ_{ex} can be determined by a variational method which maximizes the exciton binding energy of

$$E_B(\lambda_{ex})=-\frac{\hbar^2}{2\mu\lambda_{ex}^2}+\frac{q^2}{4\pi\epsilon}\left\langle\Phi\left|\frac{1}{\sqrt{|\mathbf{r}|^2+(z_e-z_h)^2}}\right|\Phi\right\rangle , \tag{7}$$

where μ is the reduced effective mass parallel to the quantum-well layers. Similar to the Wannier exciton oscillator strength formulation,[9-11] the matrix element $\langle g|r_i|\Psi_{ex}\rangle$ in Eq. (3) can be written, using Eq. (4),

$$\langle g|r_i|\Psi_{ex}\rangle=\sum_{\mathbf{k}_\parallel}A(\mathbf{k}_\parallel)\frac{\hbar}{im_0(E_c-E_v)}\langle u_c|p_j|u_v\rangle_{cell}\times\langle\varphi_e(z_e)|\varphi_h(z_h)\rangle . \tag{8}$$

The lowest heavy-hole subband $|hh1\rangle$ of the exciton state was chosen as $|\Psi_{ex1}\rangle$, to use the resonance effectively. In this case, i should be selected in the x or y direction parallel to the QW because $\langle u_c|p_z|u_v\rangle_{cell}$ is almost zero near the Γ point of a QW. The matrix element $\langle\Psi_{ex1}|r_j|\Psi_{ex2}\rangle$ is written using Eq. (4) as

$$\begin{aligned}\langle\Psi_{ex1}|r_j|\Psi_{ex2}\rangle&=\langle\Psi_{ex1}|z_h-z_e|\Psi_{ex2}\rangle\\&=\int\int\int_{cell}u_{v1}^*(\mathbf{r}_h)u_{v2}(\mathbf{r}_h)d^3r\int\int\int_{cell}u_{c1}^*(\mathbf{r}_e)u_{c2}(\mathbf{r}_e)d^3r\\&\quad\times\int\int\Phi_{ex1}(\mathbf{r}_\parallel)\Phi_{ex2}(\mathbf{r}_\parallel)d^2\mathbf{r}_\parallel\\&\quad\times\{\langle\varphi_{e1}|z_e|\varphi_{e2}\rangle\langle\varphi_{h1}|\varphi_{h2}\rangle-\langle\varphi_{e1}|\varphi_{e2}\rangle\langle\varphi_{h1}|z_h|\varphi_{h2}\rangle\}.\end{aligned} \tag{9}$$

As the asymmetry of the envelope function occurs only in the z direction, j is selected for z perpendicular to the QW. The factor

$$\int\int\int_{cell}u_{v1}^*(\mathbf{r}_h)u_{v2}(\mathbf{r}_h)d^3r$$

in Eq. (9) is nonzero when $u_{v1}(\mathbf{r}_h)$ and $u_{v2}(\mathbf{r}_h)$ are the same p-state function, so the second-lowest heavy-hole subband exciton may be selected for $|\Psi_{ex2}\rangle$. However, $\langle\varphi_{h1}|\varphi_{h2}\rangle$ and $\langle\varphi_{e1}|\varphi_{e2}\rangle$ are less than 10^{-2} even when an electric field is applied, because $|\varphi_{h1}\rangle$ and $|\varphi_{h2}\rangle$ move in the same direction. From actual calculations, it is found that $\chi_{ijk}^{(2)}(\omega;\omega_s,\omega_i)_{ex\text{-}ex}$ is less than 1×10^{-12} m/V, which is one order of magnitude smaller than that of bulk GaAs.[3]

(c) The contribution of transitions between a Wannier exciton state and nonexcitonic excited states. In the third category studied, $|n1\rangle$ is a Wannier exciton state and $|n2\rangle$ is a nonexcitonic excited state,

$$\chi_{ijk}^{(2)}(\omega;\omega_s,\omega_i)_{ex\text{-}band}=-\frac{N_z q^3}{\epsilon_0\hbar^2}\sum_{\mathbf{k}_\parallel}\frac{\langle g|r_i|\Psi_{ex}\rangle\langle\Psi_{ex}|r_j|\Psi_{cv}(\mathbf{k}_\parallel)\rangle\langle\Psi_{cv}(\mathbf{k}_\parallel)|r_k|g\rangle}{(\omega-\omega_{ex\text{-}g}+i\Gamma)[\omega_i-\omega_{cv\text{-}g}(\mathbf{k}_\parallel)+i\Gamma]} . \tag{10}$$

The nonexcitonic excited state $|\Psi_{cv}(\mathbf{k}_\parallel)\rangle$ is written as

$$\Psi_{cv}(\mathbf{k}_\parallel)=u_c(\mathbf{r}_e)\varphi_e(z_e)e^{i\mathbf{k}_{e\parallel}\cdot\mathbf{r}_{e\parallel}}u_v(\mathbf{r}_h)\varphi_h(z_h)e^{i\mathbf{k}_{h\parallel}\cdot\mathbf{r}_{e\parallel}} . \tag{11}$$

The in-plane wave-vector selection rule $\mathbf{k}_\parallel=\mathbf{k}_{e\parallel}=-\mathbf{k}_{h\parallel}$ can be included under the electric-dipole transition. Equation (8) was used as the matrix element $\langle g|r_i|\Psi_{ex}\rangle$ for $i=x$. The matrix element $\langle\Psi_{ex}|r_j|\Psi_{cv}(\mathbf{k}_\parallel)\rangle$ is given by

$$\begin{aligned}\langle\Psi_{ex}|r_j|\Psi_{cv}(\mathbf{k}_\parallel)\rangle&=\langle\Psi_{ex}|z_e-z_h|\Psi_{cv}(\mathbf{k}_\parallel)\rangle\\&=\int\int d^3\mathbf{r}_e d^3\mathbf{r}_h\left[\left\{\sum_{\mathbf{k}'_\parallel}A^*(\mathbf{k}'_\parallel)e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{r}_{e\parallel}-\mathbf{r}_{h\parallel})}\varphi_v^*(z_h)u_v^*(\mathbf{r}_h)\varphi_c^*(z_e)u_c^*(\mathbf{r}_e)\right\}\right.\\&\quad\left.\times(z_e-z_h)\varphi_v(z_h)u_v(\mathbf{r}_h)e^{i\mathbf{k}_{h\parallel}\cdot\mathbf{r}_{h\parallel}}\phi_c(z_e)u_c(\mathbf{r}_e)e^{i\mathbf{k}_{e\parallel}\cdot\mathbf{r}_{e\parallel}}\right] .\end{aligned} \tag{12}$$

Since the value is nonzero only when $\mathbf{k}'_\parallel=\mathbf{k}_\parallel$, it is written as

$$\langle\Psi_{ex}|z_e-z_h|\Psi_{cv}(\mathbf{k}_\parallel)\rangle=A(\mathbf{k}_\parallel)\{\langle\varphi_c|z_e|\varphi_c\rangle-\langle\varphi_v|z_h|\varphi_v\rangle\} . \tag{13}$$

The matrix element $\langle\Psi_{cv}(\mathbf{k}_\parallel)|\mathbf{r}_k|g\rangle$ in Eq. (10) is the same as the dipole transition for a nonexcitonic electron-hole pair:

$$\langle\Psi_{cv}(\mathbf{k}_\parallel)|\mathbf{r}_k|g\rangle=\frac{\hbar}{im_0[E_c(\mathbf{k}_\parallel)-E_v(\mathbf{k}_\parallel)]}\{\langle u_v|p_k|u_c\rangle_{cell}\langle\varphi_c(\mathbf{k}_\parallel)|\varphi_v(\mathbf{k}_\parallel)\rangle\} . \tag{14}$$

Ignoring the k dependence of $\omega_{cv\text{-}g}(\mathbf{k}_\parallel)$, $E_c(\mathbf{k}_\parallel)$, $E_v(\mathbf{k}_\parallel)$, $|\varphi_v(\mathbf{k}_\parallel)\rangle$, and $|\varphi_c(\mathbf{k}_\parallel)\rangle$, the summation in Eq. (10) may be performed only on Eq. (14) and $A(\mathbf{k}_\parallel)$ in Eq. (13). The result of these operations is rewritten as $\langle g|r_k|\Psi_{ex}\rangle$ and shown in Eq. (8). The results, Eq. (10), can be simplified to

$$\chi^{(2)}_{xzx}(\omega;\omega_s,\omega_i)_{\text{ex-band}} = -\frac{N_z q^3}{\epsilon_0 \hbar^2} \frac{|\langle g|r_x|\Psi_{ex}\rangle|^2\{\langle\varphi_c|z_e|\varphi_c\rangle - \langle\varphi_v|z_h|\varphi_v\rangle\}}{(\omega-\omega_{\text{ex1-}g}+i\Gamma)(\omega_i-\omega_{cv\text{-}g}+i\Gamma)} \ . \tag{15}$$

This simplification is justified by the fact that $A(\mathbf{k}_\parallel)$ decreases rapidly for large k vector as shown by Eqs. (5) and (6). It is considered that Eq. (4) in Ref. 2 is a result modified from Eq. (15) under the assumption of $\Gamma=0$, $\omega_{cv\text{-ex1}}=0$, and $\omega=2\omega_s=2\omega_i$. As $|\langle g|r_i|\Psi_{ex}\rangle|^2$ is proportional to the oscillator strength, large values of $\chi^{(2)}$ are expected. We can ignore the contribution of higher-order heavy-hole Wannier exciton states or light-hole Wannier exciton states, whose resonant peaks are several tens of meV's larger than that of the lowest heavy-hole exciton.

From the above discussion, $\chi^{(2)}$ is dominated by the third type of transition. In the next section, we calculate $\chi^{(2)}_{xzx}(\omega;\omega_s,\omega_i)_{\text{ex-band}}$ using Eq. (15).

III. CALCULATION

The envelope functions were calculated by the transfer-matrix method[12] with parameters summarized in Table I. The band discontinuity was divided 6:4 between the conduction band and the valence band. The band structure and calculated envelope functions are shown in Fig. 1. For simplicity, the electric field at the barrier was assumed to be 0, and the well was divided into 20 flat steps. AlAs barriers were selected. The Al composition of the well was selected to be 0.15, so the Wannier exciton will resonate with a pumping photon which has about twice the energy of 1.55-μm-wavelength signal light. As shown in Fig. 1, the envelope functions of the conduction and valence bands move to opposite sides. $\langle\varphi_c|z_e|\varphi_c\rangle - \langle\varphi_v|z_h|\varphi_v\rangle$ increases with the applied electric field and becomes as large as 20 Å.

The transition-matrix element for the interband optical transition $|\langle u_c|p_j|u_v\rangle_{\text{cell}}|^2$ in Eq. (8) was given as $2M^2 M_{\text{QW}}$, where M^2 is the transition-matrix element[13] in bulk materials,

$$M^2 = \frac{m_0^2}{12}\left[\frac{1}{M_{\Gamma_6}}-\frac{1}{m_0}\right]\frac{(E_g+\Delta)}{(E_g+2\Delta/3)} \ , \tag{16}$$

and M_{QW} is a polarization factor, for which we selected $\frac{3}{2}$ for the electron–heavy-hole transition.[14] The factor of 2 represents the degeneracy due to spin.

TABLE I. Parameters used in the calculation. E_{g_Γ} is the band-gap energy at the Γ point, Δ is the spin-orbital splitting energy, and m_e is the effective mass of electrons.

	E_{g_Γ} (eV)	Δ (eV)	m_e (m_0)	Luttinger-Kohn parameters γ_1	γ_2	γ_3
Well	1.59	0.34	0.079	6.34	3.99	2.66
Barrier	2.88	0.28	0.15	3.45	2.78	1.29

$\sum A(\mathbf{k}_\parallel)$ was approximated by $\Phi_{ex}(0)$ using the relationship of Eq. (6) in Ref. 10. $\hbar\Gamma$ was assumed to be equal to the linewidth of the Wannier exciton at room temperature, 3.7 meV.[15]

To bring the pump photon near resonance with the exciton, it was chosen to be 5 meV smaller than the exciton peak energy after taking into account the quantum-confined Stark effect redshift. In this situation, the detuning between the pump energy and the exciton energy is larger than the linewidth of the excitons, which means that the QW is almost transparent to the pump wavelength. The signal wavelength was fixed to 1.55 μm.

In the case of multiple quantum wells (MQW's), N_z is given as

$$N_z = \frac{1}{(W+L)} \ , \tag{17}$$

where W is the width of the wells and L is the width of the barriers. N_z increases with decreasing well width. We fixed the barriers of the MQW's to 40 Å. As shown in Fig. 1, 40-Å barriers were selected to prevent interaction between QW's.

IV. RESULTS AND DISCUSSION

The electric-field dependence of the absolute value of $\chi^{(2)}$ is shown in Fig. 2(a) for QW width less than 100 Å, and in Fig. 2(b) for widths more than 100 Å. For QW width greater than 50 Å, an optimum electric field exists

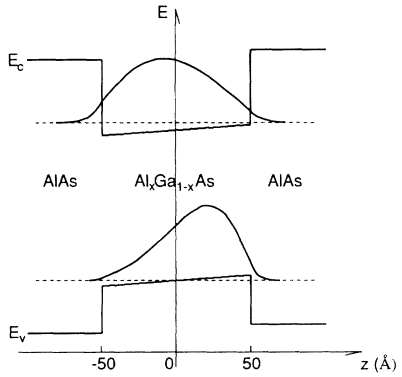

FIG. 1. Band structure and calculated envelope functions for the case of well width 100 Å and electric field 5×10^6 V/m. E_c is the energy at the Γ point in the conduction band in the bulk and E_v is that in the valence band. Broken lines show the energy of the lowest subband of electrons and holes. As the effective mass of holes is heavier than that of electrons, the subband energy of holes is smaller, and the movement of the envelope function from the center is larger than for electrons.

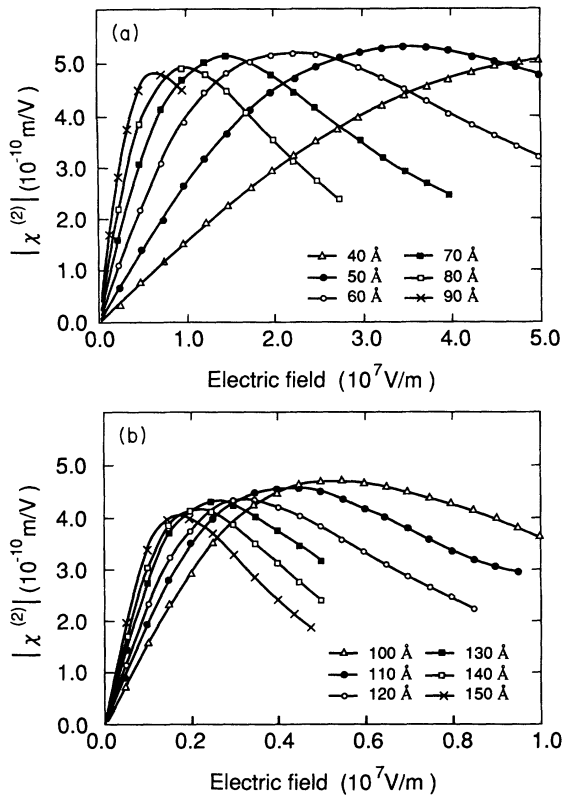

FIG. 2. Electric-field dependence of the absolute value of $\chi^{(2)}$ in biased $Al_xGa_{1-x}As$ QW's. The barriers are AlAs and the well is $Al_{0.15}Ga_{0.85}As$. (a) Well width between 40 and 90 Å. (b) Well width between 100 and 150 Å. The signal wavelength is 1.55 μm and the pump energy is 5 meV smaller than the exciton resonance energy. (a) is calculated in an electric field of less than 5×10^7 V/m, (b) less than 1×10^7 V/m.

for each well width. As the electric field increases, the dipole of the Wannier exciton increases due to the separation of the electron and hole overlap functions, while the oscillator strength weakens due to the decrease of the overlap integral of the electron and hole envelope functions.

As the well width decreases, the optimum electric field increases and the $\chi^{(2)}$ value at the optimum field point increases. A large $\chi^{(2)}$ of more than 5×10^{-10} m/V is obtained for a 70-Å well width with an electric field of 1.5×10^7 V/m. The reason for the large value is the strong resonance of the exciton energy and the pump photon energy, the strong oscillator strength of Wannier excitons, and the large dipole of the Wannier excitons. In spite of the increase of N_z with narrower QW width, the increase of the peak $\chi^{(2)}$ value is rather modest, because the effect of increasing N_z is diminished by the decrease of the $\chi^{(2)}$ of a single QW, caused by the decrease of the Wannier exciton dipole. The electric field required for the peak value increases rapidly because the shift of envelope functions of electrons and holes to opposite sides becomes difficult. For QW's less than 40 Å wide, a larger optimum $\chi^{(2)}$ may exist; however, it would require fields greater than 5×10^7 V/m, which is impractical.

The difference between Shimizu's estimation[2] (3×10^{-9} m/V at QW width of 120 Å and $E=1\times10^7$ V/m) and our calculations is mainly due to the difference in the detuning between the exciton resonant peak energy and the pumping-light energy. Shimizu selected a 1-meV detuning and assumed low-temperature conditions, while we selected the detuning as 5 meV considering the usage at room temperature.

A $\chi^{(2)}$ of more than 5×10^{-10} m/V is two orders of magnitude larger than that of $LiNbO_3$, $\chi^{(2)}=2.7\times10^{-12}$ m/V ($d_{13}=13\times10^{-9}$ esu),[3] and one order of magnitude larger than that of bulk GaAs, $\chi^{(2)}=4.6\times10^{-11}$ m/V ($d_{13}=2.2\times10^{-7}$ esu).[3]

The parametric signal gain is given as[8]

$$g=\left[\frac{\omega_s\omega_i\epsilon_0\mu_0}{n(\omega_s)n(\omega_i)}|\chi^{(2)}|^2|E_\omega|^2\right]^{1/2}, \tag{18}$$

where $n(\omega_s)$ and $n(\omega_i)$ are the refractive indices at ω_s and ω_i. The index of refraction of GaAs is about 3.0 at a wavelength of 1.55 μm. If we assume a pump-light electric field E_ω of 1.5×10^6 V/m corresponding to 1×10^6 W/cm^2 power density, which is smaller than the catastrophic optical damage level 5×10^6 W/cm^2 of a GaAs semiconductor laser,[16] we get a gain of more than 10 cm^{-1} using a $\chi^{(2)}$ of 5×10^{-10} m/V. The parametric gain can easily overcome the waveguide loss, which can be as low as 0.05 cm^{-1} at a wavelength of 1.55 μm in GaAs waveguides.[17] Therefore the $\chi^{(2)}$ of a QW with an optimized bias is large enough for nonlinear optical devices in the optical communication wavelength range.

V. SUMMARY

The second-order optical nonlinear susceptibilities $\chi^{(2)}$ of biased $Al_xGa_{1-x}As$ quantum wells have been estimated. The influence of Wannier excitonic states and nonexcitonic excited states on $\chi^{(2)}$ was examined. It was shown that the transitions between a Wannier excitonic state and nonexcitonic excited states dominate the $\chi^{(2)}$ of biased $Al_xGa_{1-x}As$ QW's. Values of $\chi^{(2)}$ were calculated for various QW widths and electric fields. A large $\chi^{(2)}$ of 5×10^{-10} m/V was predicted for QW's with widths of less than 70 Å. Such a large value is due to the strong resonance between the exciton and the pumping light, the strong oscillator strength of Wannier excitons, and the large dipole of Wannier excitons. This value is very large in the 1.55-μm wavelength range, making biased QW's good materials for nonlinear optical devices for telecommunications.

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