Dependence of third-order nonlinear susceptibility on strain induced piezoelectric field in $In_xGa_{1-x}N/GaN$ quantum well

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The third-order susceptibility of $In_xGa_{1-x}N/GaN$ quantum well (QW) has been investigated by taking into account the strain-induced piezoelectric (PZ) field, and the effective-mass Schrödinger equation is solved numerically. It is shown that the third-order susceptibility for third harmonic generation (THG) of $In_xGa_{1-x}N/GaN$ QW is related to indium content in QW and the intensity of the PZ field. The characteristics of $\chi^{(3)}_{\mathrm{THG}}(-3\omega, \omega, \omega, \omega)$ as the function of the wavelength of incident beam, well width and indium content, have been analyzed.

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Since the first demonstration of intersubband transition (ISBT) in semiconductor quantum wells (QWs)[1], there have been many reports on this topic. ISBT in QWs is considered to be an excellent device mechanism for ultrafast optical switches because of the ultrashort intersubband relaxation time (τ_{ISB})[2]. The near-infrared ISBT in GaAs and InGaAs QWs has been extensively investigated[3,4] and its relaxation time of 0.65 − 2.7 ps has been reported at the wavelengths (λ) of 9.4 − 2.5 μm[5,6]. For ISBT to be applied to 1.55-μm optical telecommunications, a large conduction band discontinuity is required. Nitride semiconductor systems, such as $In_xGa_{1-x}N/GaN$ multiple quantum wells (MQWs), are shown to be superior to conventional materials in conduction band discontinuity for that kind of devices: 1) the conduction band discontinuity is large enough to achieve ISBT at 1.55 μm with a simple MQWs structure; 2) the relaxation time of the ISBT is as short as about 200 fs[7,8]; 3) nitride material is robust under high-power excitation. ISBT is also a very useful phenomenon for applications in far-infrared detectors and cascade lasers[9,10], the latter is based on electronic transitions between energy subbands within the conduction band of a MQWs GaN-based structure.

Recently there has been considerable interest in the nonlinear properties of GaN/AlGaN QW associated with intersubband transition. But many authors neglected the intrinsic properties such as piezoelectricity by using so-called free carrier method in order to grant a guideline on the device designing issues. Actually, the strain-induced piezoelectric (PZ) field is very large. Piezoelectricity occurs in a wurtzite-type QW due to a strain-induced polarization in the longitudinal (growth) direction. The PZ field for a low indium content in $In_xGa_{1-x}N/GaN$ QWs has a strength of 2 MV/cm[11,12], which is more than ten times larger than that of the strained AlGaAs/GaAs QWs structure. The third-order susceptibility of $In_xGa_{1-x}N/GaN$ for third harmonic generation (THG) of $In_xGa_{1-x}N/GaN$ QWs has been calculated in this letter by taking into account the PZ field. It has been found that the quantum wells with different indium contents have different PZ fields, which obviously affects subband energies of conduction band and the profile of conduction band. It is presented that third-order susceptibility is related to indium content and the intensity of the PZ field.

From the density matrix formalism, the analytic formulas of the third-order susceptibility for THG can be expressed as[13]

$$\begin{aligned}\chi^{(3)}(3\omega) = \frac{N}{\hbar^3} \times \sum_{nmvl} \Bigg[& \frac{(\rho^{(0)}_{mm} - \rho^{(0)}_{ll}) M_{mn} M_{nv} M_{vl} M_{lm}}{(\omega_{nm} - 3\omega - i\Gamma_{nm})(\omega_{vm} - 2\omega - i\Gamma_{vm})(\omega_{lm} - \omega - i\Gamma_{lm})} \\ & - \frac{(\rho^{(0)}_{ll} - \rho^{(0)}_{vv}) M_{mn} M_{nv} M_{lm} M_{vl}}{(\omega_{nm} - 3\omega - i\Gamma_{nm})(\omega_{vm} - 2\omega - i\Gamma_{vm})(\omega_{vl} - \omega - i\Gamma_{vl})} \\ & - \frac{(\rho^{(0)}_{vv} - \rho^{(0)}_{ll}) M_{mn} M_{vm} M_{nl} M_{lv}}{(\omega_{nm} - 3\omega - i\Gamma_{nm})(\omega_{nv} - 2\omega - i\Gamma_{nv})(\omega_{lv} - \omega - i\Gamma_{lv})} \\ & + \frac{(\rho^{(0)}_{ll} - \rho^{(0)}_{nn}) M_{mn} M_{vm} M_{lv} M_{nl}}{(\omega_{nm} - 3\omega - i\Gamma_{nm})(\omega_{nv} - 2\omega - i\Gamma_{nv})(\omega_{nl} - w - i\Gamma_{nl})} \Bigg], \end{aligned} \tag{1}$$

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where N is the electron density in the QW, $\omega_{nm} \equiv (E_n - E_m)/\hbar$, and $\Gamma_{nm}^{-1} = \tau_{nm}$ is the dephasing time between the states $|\Psi_n\rangle$ and $|\Psi_m\rangle$, and M_{nm} is the dipole moment matrix element. The effective-mass approximation has been used to calculate the subband structure of a single $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N/GaN}$ QW grown at the z-axis direction. The energy levels and wave functions for particles in the system can be obtained by solving the Schrödinger equation with a proper Hamiltonian. The one-dimensional Schrödinger equation is given by

$$\left[-\frac{\hbar^2}{2m^*}\frac{\mathrm{d}^2}{\mathrm{d}z^2}+\nu(z)\right]\Psi_n(z)=E_n(z)\Psi_n(z), \qquad (2)$$

where m^* is effective mass equals m_w^* (in QW) or m_b^* (in barrier layer) and $\nu(z)$ is defined as

$$\nu(z)=\nu_0(z)+eF_\mathrm{PZ}z, \qquad (3)$$

where $\nu_0(z)$ is the confinement potential due to band offset between the well and the barrier and $eF_\mathrm{PZ}z$ is the potential due to PZ field. F_PZ is the sum of PZ fields and expressed by

$$F_\mathrm{PZ}=-\frac{2\varepsilon_{xx}}{\varepsilon_0\varepsilon}\left[\frac{c_{13}e_{33}}{c_{33}}-e_{31}\right], \qquad (4)$$

where $\varepsilon_{xx} = \varepsilon_{yy}$ is the in-plane strain, c_{ij} is the elastic constants, e_{ij} is the PZ constant (referred to hexagonal principal axes), ε is the z-axis static dielectric constant, and ε_0 is the permittivity of free space. For GaAs QW, the potential due to PZ field is small and can be considered as a perturbation. But in GaN QW, PZ field is large enough and the consequent potential cannot be treated as a perturbation. We have to solve Eq. (2) numerically following a simple but highly accurate technique given in Ref. [14]. Wave functions and conduction band profile are shown in Fig. 1.

We consider a (0001)-grown $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N/GaN}$ QW structure, an undoped strained InGaN epilayer sandwiched between two GaN layers, and the GaN barriers are assumed unstrained and have no PZ field. The following parameters were adopted for these calculations: c_{13} = 106 GPa, c_{33} = 398 GPa, e_{33} = 0.79 C/m^2, e_{31} = 0.49 C/m^2, and ε = 10.3 for GaN; c_{13} = 94 GPa, c_{33} = 200 GPa, e_{33} = 0.97 C/m^2, e_{31} = 0.57 C/m^2, and ε = 14.6 for InN[15]. We take the conduction discontinuity as 2 eV and the dephasing time $\tau_{12} = \tau_{23} = \tau_{34}$ = 100 fs. Selecting two different well widths of 7 and 6.5 nm, we show the $\chi^{(3)}_\mathrm{THG}(-3\omega,\omega,\omega,\omega)$ as a function of incident wavelength with low indium content x = 0.22, 0.28 and 0.34. The results are shown in Figs. 2 and 3.

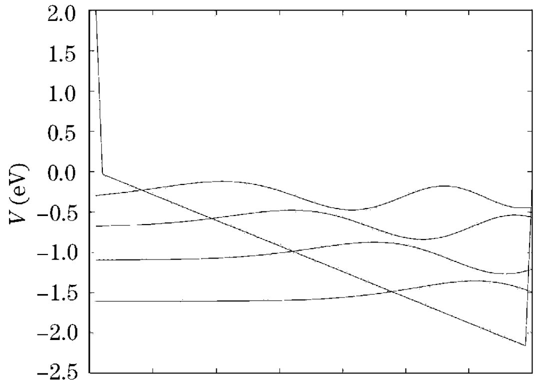

Fig. 1. Conduction-band profile of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N/GaN}$ QW taking into account PZ field. The lowest four wave functions are shown.

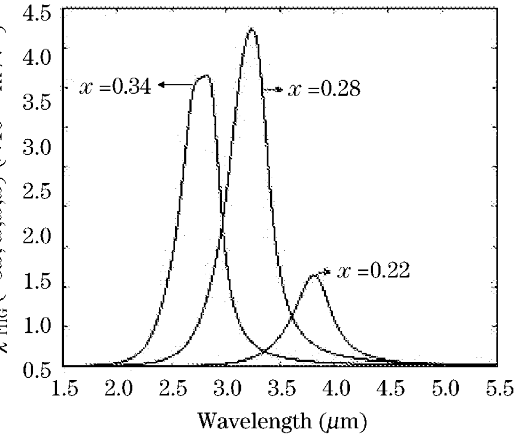

Fig. 2. The variation of $\chi^{(3)}_\mathrm{THG}(-3\omega,\omega,\omega,\omega)$ of the QW with the incident wavelength with different indium contents, the well width is 7 nm.

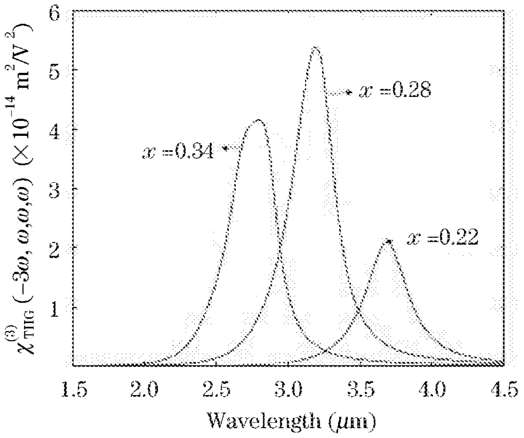

Fig. 3. The variation of $\chi^{(3)}_\mathrm{THG}(-3\omega,\omega,\omega,\omega)$ of the QW with the incident wavelength with different indium contents, the well width is 6.5 nm.

From the above figures, we can see indium content is directly related to PZ field, which affects electron energy levels, wave functions and dipole moment matrix element. The QWs with different indium content have different values of $\chi^{(3)}_\mathrm{THG}(-3\omega,\omega,\omega,\omega)$. It is found that $\chi^{(3)}_\mathrm{THG}(-3\omega,\omega,\omega,\omega)$ has the maximum value when indium content is 0.28 and reaches minimum if indium content drops to 0.22. While indium content decreasing, the peaks of spectrum of $\chi^{(3)}_\mathrm{THG}(-3\omega,\omega,\omega,\omega)$ cause red shift. Another important factor affecting $\chi^{(3)}_\mathrm{THG}(-3\omega,\omega,\omega,\omega)$ is QW width. For the same indium content, the value of $\chi^{(3)}_\mathrm{THG}(-3\omega,\omega,\omega,\omega)$ with 6.5-nm well width is larger than that with 7-nm well width. Indium content and well width can be selected to control PZ field and optimize the third-order susceptibility. Therefore, we also select

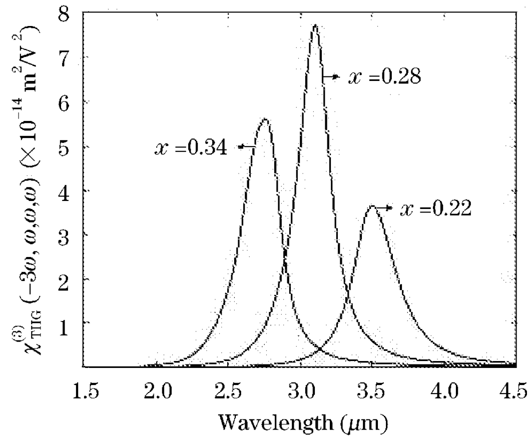

Fig. 4. The variation of $\chi^{(3)}_{\mathrm{THG}}(-3\omega,\omega,\omega,\omega)$ of the QW with the incident wavelength with different indium contents, the well width is 6 nm.

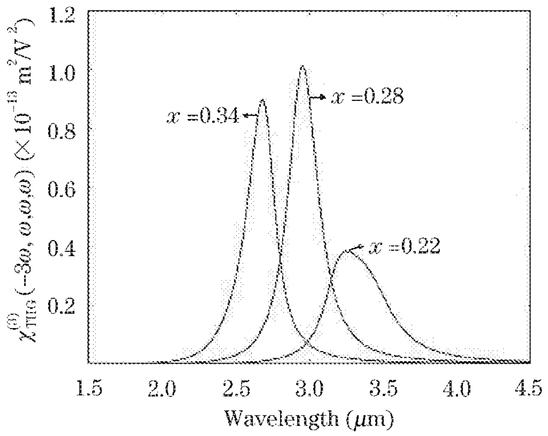

Fig. 5. The variation of $\chi^{(3)}_{\mathrm{THG}}(-3\omega,\omega,\omega,\omega)$ of the QW with the incident wavelength with different indium contents, the well width is 5.5 nm.

well widths of 6 and 5.5 nm and give the spectra of $\chi^{(3)}_{\mathrm{THG}}(-3\omega,\omega,\omega,\omega)$ in Figs. 4 and 5.

Although the well widths are different from those in Figs. 2 and 3, there is still a red shift when the indium content decreases. The lower the indium content is, the narrower the electron energy gap becomes. Assaying these four figures from Figs. 2 to 5, we find that when the well width becomes narrower, the value of third-order susceptibility becomes larger. The reason is that QW with narrower well width has larger dipole moment matrix element. In order to select proper indium content, we also calculate third-order susceptibility with indium content $x = 0.25$, 0.31 and 0.37 with the same condition. Among these six different indium compositions, third-order susceptibility reaches maximum at $x = 0.28$ with the same well width. The reason is that indium content affects PZ field, wave functions, electron energy structure and dipole moment matrix element. These factors directly affect $\chi^{(3)}_{\mathrm{THG}}(-3\omega,\omega,\omega,\omega)$.

In conclusion, the third-order susceptibility for THG of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N/GaN}$ QW has been studied by taking into account PZ field. Indium content is directly related to PZ field, which intensitively affects the value and the position of third-order susceptibility. When indium content decreases, the peak value of $\chi^{(3)}_{\mathrm{THG}}(-3\omega,\omega,\omega,\omega)$ has a red shift. We can select proper indium content and well width to obtain larger third-order susceptibility.

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