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Interdiffusion as a means of fabricating parabolic quantum wells for the enhancement of the nonlinear third-order susceptibility by triple resonance

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Interdiffusion of a standard AlGaAs/GaAs quantum well is proposed as a viable alternative to the complex techniques necessary to fabricate parabolic quantum wells. The extent of the linear diffusion is optimized in order to produce an energy-level ladder if four almost equally spaced eigenstates. The calculated third-order susceptibility of 2700 $(\mathrm{nm/V})^2$ is comparable with that of the parabolic quantum well, which is more than six orders of magnitude compared to that of the bulk GaAs. © *1996 American Institute of Physics.* [S0003-6951(96)01230-2]

Nonlinear optics[1] is the study of phenomena that occurs as a consequence of the modification of the optical properties of a material system by the presence of light. Typically, only higher-power laser is sufficiently intense to modify the optical properties of a material system. Conventionally, nonlinear optics is often taken to be the study of intensive light generated second-harmonic frequency, as it is first demonstrated by Franken *et al.* in 1961.[2] The nonlinear optical response can often be described by expressing the polarization *P(t)* as a power series in terms of the field strength, *E(t)*, and susceptibility, χ, as $P(t)=\Sigma_{k=1}^{N}\chi^{(k)}E^{k}(t)$. The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are known as the second- and third-order nonlinear optical susceptibilities, respectively. In the past decade, interests have been focused on the quantum confinement effect of carriers in a semiconductor QW,[3] including its nonlinear optical properties. Strong infrared absorption features, such as extremely large oscillator strength and relatively narrow linewidth, associated with intraband transitions between subbands of the QW have been observed.[4] This gives a very large optical nonlinearlity and for a conventional square, QW, there are about six orders of magnitude larger than that of the bulk GaAs. However, this cannot be stretched too far due to the limitation of the single pair of resonant subbands.

Recently, very strong third-harmonic susceptibility has been achieved in superlattice structures,[5] and, in the parabolic QW structure[6] due to the fourth power dependence on the dipole moment. The equally spaced eigenstate energy in a finite parabolic quantum well provides multiple pairs of resonant subbands so that the enhancement in the third-order susceptibility is much more enhanced than that in the case of square QWs. Two fabrication techniques can be used to realize a parabolic QW, one is by changing the material composition of the well gradually[7] and the other is by using alternate deposition of thin undoped layers of GaAs and AlGaAs of varying thickness, which increases quadratically with position.[8] However, whether using smooth or abrupt heterointerfaces, there still remain difficulties in accurately generating the two types of parabolic structure, thus making it not cost effective for mass production. On the other hand, there is some recent interest[9] in using the technique of interdiffusion to modify the confinement profile of a QW including its intersubband optical properties[10] and detector.[11] By adjusting carefully the parameters such as composition, well width and diffusion length of the QW, the interdiffused QW, (DFQW) with a parabolic-like confinement profile can be obtained. This is much simpler and easier than the methods for the fabricating a parabolic QW suggested previously. In this letter, a DFQW structure with a parabolic-like confinement profile is proposed to produce an enhanced third-order susceptibility similar to the case of a parabolic QW.

Thermal processing of GaAs-$\mathrm{Al}_w\mathrm{Ga}_{1-w}\mathrm{As}$ QW, such as rapid thermal annealing around 1000 °C, produces interdiffusion across the heterointerfaces of an as-grown square QW which results in a modification of the composition and confinement profiles of the QW structure. Assuming linear diffusion, the interdiffusion process can be characterized by a diffusion length L_d defined as $L_d=(Dt)^{1/2}$, where D and t are the diffusion coefficient and annealing time, respectively. A small value of L_d corresponds to a lightly interdiffused QW while a large value of L_d corresponds to an extensively interdiffused well. The diffused Al composition profile, $w(z)$, across the QW structure is given by[12]

$$w(z)=w_0\left\{1-\frac{1}{2}\left[\mathrm{erf}\left(\frac{L_z+2z}{4L_d}\right)+\mathrm{erf}\left(\frac{L_z-2z}{4L_d}\right)\right]\right\},\qquad(1)$$

where w_0 is the as-grown Al mole fraction in the barrier, L_z is the as-grown width of the QW, z is both the quantization and the growth axis (QW centered at $z=0$), and erf denotes the error function. The DFQW confinement profile for the conduction band, $U_C(z)$, is defined by $U_C(z)=Q_C[E_g(z)-E_g(z=0)]$, where Q_C is the offset ratio which is set to 0.7 and $E_g(w)=1.424+1.594w+w(1-w)(0.127-1.31w)$ is the bulk band gap. The Schrödinger equation is solved using this potential numerically by a finite difference method consistently with the Poisson equation. Once the eigenstates have been obtained, the third-harmonic susceptibilities can now be readily computed via the density matrix formalism as follows:[13]

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$$\chi^{(3)}=\frac{N}{\hbar^3}\times\sum_{abcd}\left[(\Delta\rho_{bd}^{(0)})\times\frac{M_{ba}M_{ac}M_{cd}M_{db}}{(\Delta\omega_{ab}-3\omega-i\Gamma_{ab})(\Delta\omega_{cb}-2\omega-i\Gamma_{cb})(\Delta\omega_{db}-\omega-i\Gamma_{db})}-(\Delta\rho_{dc}^{(0)})\right.$$
$$\times\frac{M_{ba}M_{ac}M_{db}M_{cd}}{(\Delta\omega_{ab}-3\omega-i\Gamma_{ab})(\Delta\omega_{cb}-2\omega-i\Gamma_{cb})(\Delta\omega_{cd}-\omega-i\Gamma_{cd})}-(\Delta\rho_{cd}^{(0)})$$
$$\times\frac{M_{ba}M_{cb}M_{ad}M_{dc}}{(\Delta\omega_{ab}-3\omega-i\Gamma_{ab})(\Delta\omega_{ac}-2\omega-i\Gamma_{ac})(\Delta\omega_{dc}-\omega-i\Gamma_{dc})}+\Delta\rho_{da}^{(0)}$$
$$\left.\times\frac{M_{ba}M_{cb}M_{cd}M_{ad}}{(\Delta\omega_{ab}-3\omega-i\Gamma_{ab})(\Delta\omega_{ac}-2\omega-i\Gamma_{ac})(\Delta\omega_{nl}-\omega-i\Gamma_{ad})}\right], \tag{2}$$

where N is the total electron density in the DFQW which is defined as $N_{2\mathrm{DEG}}/L_z$ and the total surface 2D electron gas is determined to be 1.2×10^{12} cm^{-2} in this calculation, $\Delta\omega_{nm}=(E_n-E_m)/\hbar$, and $\Gamma_{nm}^{-1}=\tau_{nm}$ is the dephasing time between the DFQW states $|\phi_n\rangle$ and $|\phi_m\rangle$. The dipole moment matrix element M_{nm} is given by $M_{nm}=\int_{-5L_z}^{5L_z}\phi_n^*(z)qz\phi_m(z)dz$, where q is the electronic charge; $\Delta\rho_{mn}^{(0)}=\rho_{mm}^{(0)}-\rho_{nn}^{(0)}$ is the difference of unperturbed diagonal density matrix $\rho_{nn}^{(0)}$ which is equal to the thermal equilibrium occupation probability of the corresponding state and can be expressed as[1] $\rho_{nn}^{(0)}=\{1+\exp[(E_n-E_F)/(K_BT)]\}^{-1}$, where E_F is the Fermi level, T is the temperature which is taken to be 300 K, and K_B is the Boltzmann constant. In Eq. (2), a,b,c,d are numbers representing the four lowest electron eigenstates and they are permutated in cyclic order in the calculation. Four iterations are required in the summation and their respective combinations are $(a,b,c,d)=(1,2,3,4),(2,3,4,1),(3,4,1,2)$, and $(4,1,2,3)$. The Fermi level is determined by a method briefly described as follows. The electrons in each subband are considered as a quasi-2D electron gas, and their corresponding occupy states can be expressed as $N_n=(m_e^*/\pi\hbar^2)k_BT\ \ln\{1+\exp[(E_f-E_n)/k_BT]\}$, where $m_e^*=(0.0632+0.0856w+0.0231w^2)m_0$ is the effective mass of electron, and m_0 is the free-electron mass. Since $N_{2\mathrm{DEG}}=\Sigma N_n$ which is already known and hence the quasi-Fermi level can be obtained.

In order to tailor design the DFQW with an aim to achieve equally spaced eigenenergies, the structural parameters and interdiffusion extent of the DFQW is chosen very carefully so that its confinement profile is parabolic-like in shape. During the design, it is found that the eigenenergy spacing and thus the magnitude and position of the third harmonic susceptibility is a strong function of the diffusion length. An optimized result shows that if the structural parameters is set to $w=0.37$, $L_z=170$ Å, and $L_d=22$ Å in the AlGaAs/GaAs material system, this will give a good parabolic shaped profile. This is shown in Fig. 1 together with the parabolic QW ($L_z=220$ Å, $w=0.3$, spring constant $K=0.049$ meV/Å^2) for comparison. As can be seen, the DFQW profile matches the parabolic profile exactly at the barrier but only with a slight mismatch in the center of the well. This should have caused a minor effect on the enhanced $\chi^{(3)}$ generated by the DFQW. The calculated eigenenergy differences for the DFQW are, $\Delta E_{21}=E2-E1=74$ meV, $\Delta E_{31}=E_3-E_1=153$ meV, and $\Delta E_{41}=E_4-E_1=226$ meV. Thus it can be seen from these values that the four energy levels are nearly equally spaced.

The nonlinear $|\chi^{(3)}(3\omega)|$ of the optimized DFQW as a function of the photon wavelength λ is shown in Fig. 2 together with that of a parabolic QW ($L_z=170$ Å, $w=0.4$, $K=0.125$ meV Å^2) for comparison purposes. It can be clearly seen that both spectra consist of only one peak and with

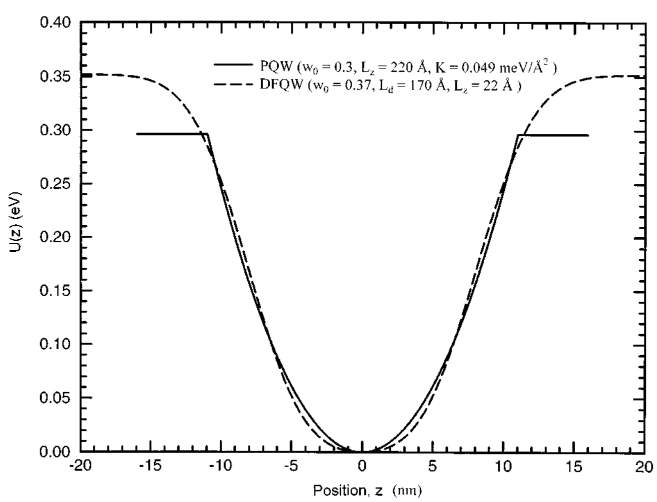

FIG. 1. Confinement profile of the DFQW ($w=0.37$, well width=170 Å, diffusion length=22 Å) and the PQW ($w=0.3$, well width=220 Å, $K=0.049$ meV/Å^2).

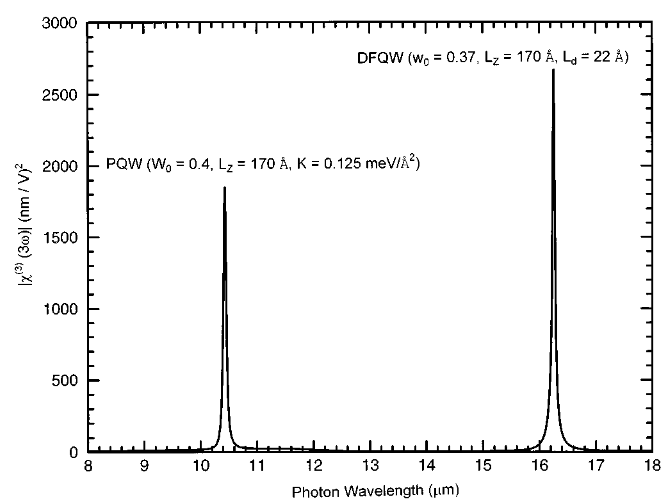

FIG. 2. Third-order susceptibility of the DFQW and that of the PQW ($w=0.4$, well width=170 Å, $K=0.125$ meV/Å^2).

about the same magnitude except in the case of the DFQW, the $\chi^{(3)}$ resonance is at a longer wavelength $\lambda=15.3$ μm. The magnitude of the DFQW peak is about 1000 $(\mathrm{nm/V})^2$ more than that of the parabolic QW. This is due to the fact that the nonlinear enhancement does not only depend on the energy ladder but also on the overlapping of the eigenstate wavefunctions, a phenomena also observed in the case of a hyperbolic QW.[12] If both the as-grown QW structure and diffusion length are adjusted, the wavelength position of the enhanced $\chi^{(3)}$ can be varied around $\lambda=15.3$ μm, thus providing a tuning range. This demonstrates the advantage of using DFQW to achieve an enhanced $\chi^{(3)}$ while at the same time simplifies the fabrication technology.

To conclude, a DFQW has been successfully employed to enhance the third-harmonic susceptibility to an extent that it is six times better than the case of square QW. The obtained $\chi^{(3)}$ enhancement is as high as 2700 $(\mathrm{nm/V})^2$. However, the operating wavelength is at around 15.3 μm and not at the 10.6 μm common pumped by CO_2 lasers. Although this enhancement can also be achieved by utilizing the parabolic QWs, technical difficulties in their fabrication with a high yield and low price still exist. In addition, by carefully adjusting the DFQW structures, the operating wavelength for an enhanced $\chi^{(3)}$ can be varied to produce a tuning range. The technology of using interdiffusion is thus useful, simple, and effective for producing large nonlinearlity.

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