Design of GaAs/Al$_x$Ga$_{1-x}$As asymmetric quantum wells for THz-wave by difference frequency generation*

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The energy levels, wave functions and the second-order nonlinear susceptibilities are calculated in GaAs/Al$_{0.2}$Ga$_{0.8}$As/Al$_{0.5}$Ga$_{0.5}$As asymmetric quantum well (AQW) by using an asymmetric model based on the parabolic and non-parabolic band. The influence of non-parabolicity can not be neglected when analyzing the phenomena in narrow quantum wells and in higher lying subband edges in wider wells. The numerical results show that under double resonance (DR) conditions, the second-order difference frequency generation (DFG) and optical rectification (OR) generation susceptibilities in the AQW reach 2.5019 μm/V and 13.208 μm/V, respectively, which are much larger than those of the bulk GaAs. Besides, we calculate the absorption coefficient of AQW and find out the two pump wavelengths correspond to the maximum absorption, so appropriate pump beams must be selected to generate terahertz (THz) radiation by DFG.

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Terahertz (THz)-wave technology has attracted a great deal of scientific and industrial interest in recent years[1-5]. Though suitable THz detectors and optical components have been developed in the last few years, the lack of efficient source is the key problem in the THz applications. Among all kinds of THz radiators, THz quantum cascade laser (QCL) is a promising one due to its higher output power and monochromaticity[6], while making QCL needs complicated technique procedure and high cost[7,8]. In order to overcome the shortcomings, an alternative way to produce THz radiation at room temperature (RT) is the difference frequency generation (DFG) in a nonlinear optical crystal using infrared (IR) or visible pump lasers[9,10]. Asymmetric quantum well (AQW) is suited for the design of THz devices because its energy and transition can be tailored by changing the geometry of the well in the THz field. Besides, large $\chi^{(2)}$ in quantum well structure also can be achieved by tailoring, so that all interacting fields are in resonance with intersubband transitions[11].

In this paper, the AQW is designed for optically exited intersubband THz emission. Because the optical nonlinearity for DFG is not expected, this approach can lead to an electrically pumped continuous wave (CW) RT semiconductor THz source. It is obvious that the AQW structure is a promising candidate for THz lasing due to its advantages[12].

The use of asymmetric or stepped quantum well structures for producing terahertz radiation from intersubband transitions was first suggested by Berger[13]. To enhance the nonlinear susceptibility, we design the structure to take advantage of resonant effects ($\hbar\omega_1 = E_3 - E_1, \hbar\omega_2 = E_2 - E_1$), leading to the maximum second-order DFG and optical rectification (OR) susceptibilities.

The structure of well is designed to be asymmetric and with intersubband transitions that can be excited by mixing two laser beams, as shown in Fig.1.

Under the condition of parabolicity, the Schrödinger equation is needed to solve the energy levels and the wave functions. The equation for each semiconductor layer can be written as follows

$$\left[-\frac{\hbar^2}{2m_1}\frac{\partial^2}{\partial z^2}+V_2(z)\right]\Psi_1(z)=E\Psi_1(z)\quad z<0$$

$$-\frac{\hbar^2}{2m_2}\frac{\partial^2}{\partial z^2}\Psi_2(z)=E\Psi_2(z)\qquad 0<z<L_1$$

$$\left[-\frac{\hbar^2}{2m_3}\frac{\partial^2}{\partial z^2}+V_1(z)\right]\Psi_3(z)=E\Psi_3(z)\quad L_1<z<L_2$$

$$\left[-\frac{\hbar^2}{2m_4}\frac{\partial^2}{\partial z^2}+V_2(z)\right]\Psi_4(z)=E\Psi_4(z)\quad z>L_2 \quad . \quad (1)$$
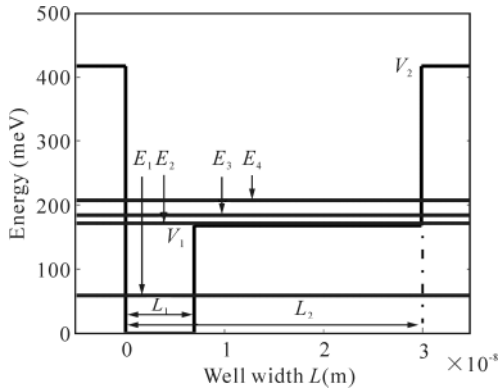

Fig.1 Conduction-band diagram of the AQW structure

Because the wave functions and their first derivatives are continuous on boundary conditions, we can acquire the determining equations

$$\begin{cases}\varphi_1(z)=A\exp(k_1 z), & k_1=\sqrt{2m_1^*(V_2-E)/\hbar^2} & z<0\\ \varphi_2(z)=B\cos(k_2 z+\alpha), & k_2=\sqrt{2m_2^* E/\hbar^2} & 0<z<L_1\\ \varphi_3(z)=C\exp(-k_3 x), & k_3=\sqrt{2m_3^*(V_1-E)/\hbar^2} & L_1<z<L_2\,\text{and}\,E<V_1\\ \varphi_3(z)=C\cos(k_3 x+\beta), & k_3=\sqrt{2m_3^*(E-V_1)/\hbar^2} & L_1<z<L_2\,\text{and}\,E>V_1\\ \varphi_4(z)=D\exp(-k_4 x), & k_4=\sqrt{2m_4^*(V_2-E)/\hbar^2} & L_2<z\end{cases} \quad . \quad (2)$$

Non-parabolicity in the neighborhood of energyband extreme in bulk semiconductors can be described by the dispersion relation[14]

$$E=\frac{\hbar^2 k^2}{2m^*}(1-\gamma k^2), \tag{3}$$

$$\gamma=\frac{\hbar^2}{2m^*}(1-\frac{m^*}{m_0})^2\frac{3+4y+2y^2}{3+5y+2y^2}\frac{1}{E_g}\quad , \tag{4}$$

where k is the wave number, $\tilde{a}$ is the non-parabolicity parameter, $y=\ddot{A}/E_g$ (we take the zero of energy at the bottom of the conduction band), $\ddot{A}$ is the spin-orbit splitting, E_g is the energy gap, and m_0 is the free-electron mass. When an AQW is formed, for example，by an $Al_{0.2}Ga_{0.8}As$ step layer, $Al_{0.5}Ga_{0.5}As$ barrier layer and GaAs well structure, generally, we assume that the conduction bands of the well and step layer are non-parabolic, while the conduction band of bar-

rier layer is parabolic. So the Schrödinger equation for the wave functions can be obtained after inserting $k\rightarrow d/dz$,

$$\hat{H}\psi(z)=-\frac{\hbar^2\gamma(z)}{2m^*(z)}\frac{d^4}{dz^4}\psi(z)-$$
$$\frac{\hbar^2}{2m^*(z)}\frac{d^2}{dz^2}\psi(z)+V(z)\psi(z)=E\psi(z)\quad . \tag{5}$$

The nonlinear susceptibility can be expressed as[15]

$$\chi_{DFG}^{(2)}=\frac{Ne^3}{\hbar^2}Z_{12}Z_{13}Z_{23}\times[\frac{-(\rho_{11}^{(0)}-\rho_{22}^{(0)})}{(\Omega-\omega_{32}+j\Gamma_{32})(\omega_{21}-\omega_2+j\Gamma_{21})}+$$
$$\frac{-(\rho_{11}^{(0)}-\rho_{33}^{(0)})}{(\Omega-\omega_{32}+j\Gamma_{32})(\omega_1-\omega_{31}+j\Gamma_{31})}]\approx$$
$$-\rho_{11}^{(0)}\frac{Ne^3}{\varepsilon_0\hbar^2}Z_{12}Z_{13}Z_{23}\frac{\omega_T-\omega_{32}+j(\Gamma_{21}+\Gamma_{31})}{\omega_T-\omega_{32}+j\Gamma_{32}}\times$$
$$\frac{1}{(\omega_1-\omega_{31}+j\Gamma_{31})(\omega_{21}-\omega_2+j\Gamma_{21})}\quad , \tag{6}$$

where N is the three-dimensional (3-D) free carrier density in the AQW, $\hbar$ is the Planck's constant, Z_{ij} is the matrix element of the intersubband dipole moment of the electron, $\omega_{ij}=(E_i-E_j)/\hbar$ is the Bohr's frequency, $\rho_{ij}^{(0)}$ is the diagonal element of the matrix density taken at thermal equilibrium, Γ_{ij} is the damping rate of the coherence ρ_{ij} when $i\neq j$, and $\Omega=\omega_3-\omega_2$ is the difference frequency of the pump beams. For simplicity, the diagonal terms of the relaxation rate matrix Γ are assumed to be equal to $1/T_1$. T_1 can be considered as an averaged intersubband relaxation time among the three states. The last equality in the above equation is based on the fact that the population $\rho_{ii}^{(0)}$ is much larger than $\rho_{22}^{(0)}$ and $\rho_{33}^{(0)}$.

According to the double resonance (DR) conditions of $\hbar\omega_1=E_3-E_1$ and $\hbar\omega_2=E_2-E_1$, the maximum $\chi_{DFG}^{(2)}$ can be obtained as

$$\chi_{DFG}^{(2)}\approx-\rho_{11}^{(0)}\frac{Ne^3}{\varepsilon_0\hbar^2}Z_{12}Z_{13}Z_{23}\frac{\Gamma_{21}+\Gamma_{31}}{\Gamma_{32}\Gamma_{31}\Gamma_{21}}=$$
$$-\rho_{11}^{(0)}\frac{Ne^3}{\varepsilon_0\hbar^2}Z_{12}Z_{13}Z_{23}T_2^2. \tag{7}$$

When the difference frequency is much smaller than ω_{32}, the OR process takes over. Normally, we can discuss only two energy levels, so the nonlinear susceptibility can be expressed as

$$\chi_o^{(2)}\approx\frac{Ne^3}{\hbar^2}(\rho_{11}^{(0)}-\rho_{22}^{(0)})Z_{12}^2(Z_{11}-Z_{22})\times\frac{\omega_T+j2\Gamma_{31}}{\omega_T+j\Gamma}\times$$
$$\frac{1}{(\omega_{21}-\omega_2+j\Gamma_{21})(\omega_1-\omega_{21}+j\Gamma_{21})}\quad . \tag{8}$$

According to the DR conditions, the maximum $\chi_0^{(2)}$ can be obtained

$$\chi_{o,max}^{(2)}\approx2\frac{Ne^3 T_1 T_2}{\varepsilon_0\hbar^2}\rho_{11}^{(0)}Z_{12}^2(Z_{22}-Z_{11})\quad . \tag{9}$$

Since the second-order optical susceptibilities have a cubic dependence relative to the dipole matrix elements, stronger second-order optical nonlinearities are expected in AQW's insofar as the inversion symmetry is broken.

Solving Eq.(2), we can get energy levels and wave functions of all states under the condition of parabolicity.

Fig.2 displays the energy levels as a function of well width. The four states have the same trend, because of the decline of quantum confinement effect. The gaps of energy levels are variable at different points. So we may design different well widths to acquire a variety of THz wavelengths through DFG. The corresponding wave functions are illustrated in Fig.3.
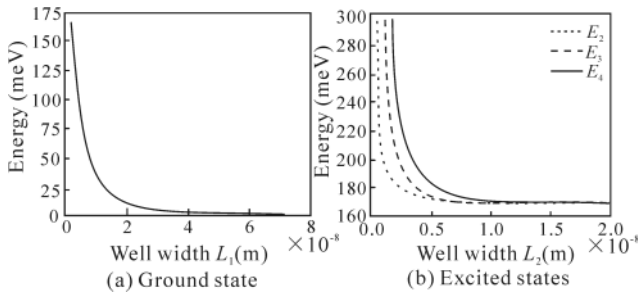

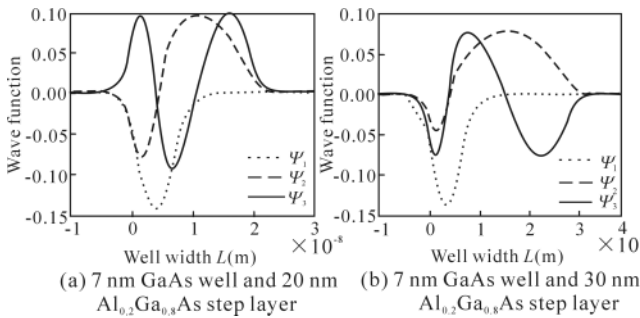**Fig.2 Energy levels in the AQW under the condition of parabolicity**

Fig.3 Wave functions of the first three energy levels under the condition of parabolicity

From Eqs.(3) and (4) we can get $\tilde{a}_1=\tilde{a}_4=0$ ($Al_{0.5}Ga_{0.5}As$), $\tilde{a}_2=3.288\times10^{-19}$ (GaAs), and $\tilde{a}_3=1.7031\times10^{-19}$ ($Al_{0.2}Ga_{0.8}As$). After substituting the value of $\tilde{a}$ into Eq.(3), the energy levels and wave functions under the condition of non-parabolicity are calculated.

Fig.4 displays the results of calculation for the lowest four energy states in the AQW. All the four states show the same monotonic behavior. When the gap of energy level is greater or the well width is narrow, the influence of non-parabolicity is obvious.
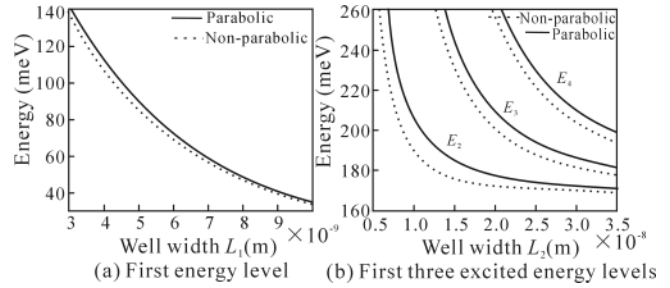

Fig.4 Calculation results of the first four energy levels in the AQW

It can be seen from Fig.5 that the wave functions are also strongly affected by the non-parabolicity. For the wave functions of higher levels, the curves are no longer smooth. However, when the value of energy is greater, the influence of non-parabolicity can not be neglected. The energy levels and wave functions of all states under the condition of non-parabolicity all illustrate the importance of accounting correctly for non-parabolicity when analyzing phenomena in narrow quantum wells and in higher lying subband edges in wider wells.
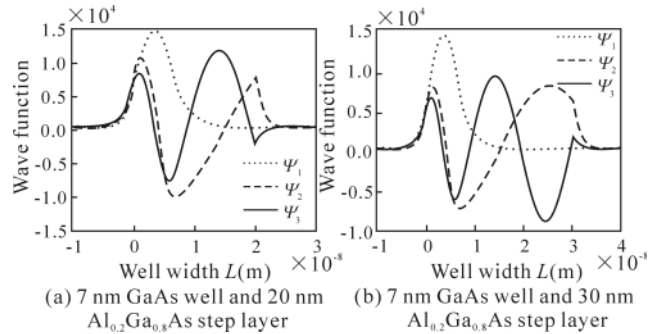

Fig.5 Wave functions of the first three energy levels under the condition of non-parabolicity

For a given concentration of carriers, the second-order DFG and OR generation susceptibilities are determined by the geometrical factor of $Z_{ij}=<i|z|j>$. From Figs.3 and 5, we can see that the wave function of ground state is mainly distributed in the low well (GaAs well), so it is completely defined when the low well width is constant. Under the condition of non-parabolicity, if the high well ($Al_{0.2}Ga_{0.8}As$ well) is narrow, the first and second excited wave functions can have larger distribution in low well.

For the calculation of GaAs/Al_xGa_{1-x}As AQW, the parameters are adopted as $N=5.0\times10^{16}$ cm^{-3}, $\rho_{11}=1$, $T_1=1$ ps, $T_2=0.14$ ps, $L_1=7$ nm and $L_2=30$ nm. Assuming most of the electrons are in the state of E_3, we calculate the value of the nonlinear susceptibility for DFG process as $\chi_{DFG,max}^{(2)} \approx 2.5019$

μm/V, and for OR process as $\chi_{o,max}^{(2)} \approx 13.208$ μm/V, which are both more than four orders of magnitude higher than those in bulk GaAs.

Under the condition of non-parabolicity, the THz absorption coefficient in GaAs AQW is discussed. Through Eq.(5), we can figure out the energy levels as E_1=56.7078 meV, E_2=170.0292 meV and E_3=181.4327 meV, respectively. Assuming most of the electrons are in the state of E_3, the dipolar matrix element in intersubband transitions of Z_{23}=<2|z|3> is given as -2.6251×10⁻⁹.

In optical device applications, a large variation of absorption with pump beam could be affected or applied to modulate the confinement in optical devices[16]. Such modulation effects are of special interest in the 9−11 μm region, where the refractive index change corresponds to the CO_2 laser lines. Considering the AQW with optical intensity of 10.0 MW/cm², the absorption coefficient of THz wave in AQW as a function of pump wavelength is illustrated in Fig.6. We can see that with the wavelengths of two pump beams, the absorption coefficient first decreases and then increases. The maximum value is 1.553×10^7 m⁻¹ when the two pump wavelengths are 9.69 μm and 10.64 μm, respectively.

(a) As a function of the pump wave λ_{P1} (b) As a function of the pump wave λ_{P2}
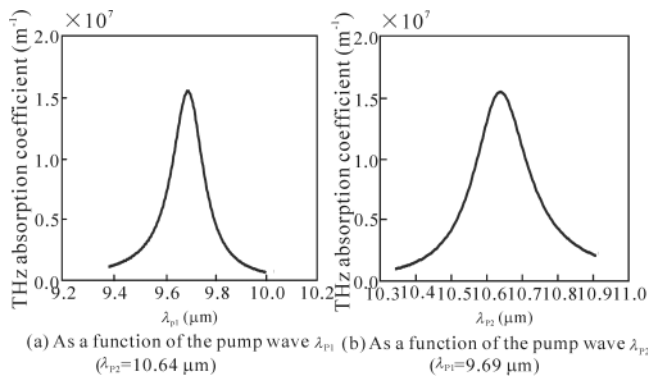(λ_{P2}=10.64 μm)   (λ_{P1}=9.69 μm)

Fig.6 THz-wave absorption coefficient

In summary, electron quantized energy levels, wave functions and dipolar matrix elements in the GaAs/Al$_{0.2}$Ga$_{0.8}$As/Al$_{0.5}$Ga$_{0.5}$As AQW are calculated taking the conduction-band non-parabolicity into account through the original Schrödinger equation. The results indicate that the non-parabolicity significantly affects the quantized level positions, particularly in narrow wells and higher excited levels. The non-

parabolicity and quantum confinement can determine the matrix element of the intersubband dipole moment. Because the absorption coefficient may have a maximum value, we must select appropriate pump wavelengths to avoid the maximum absorption.

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