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The enhancement of optical third harmonic susceptibility in a parabolic quantum well by triple resonance

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The third harmonic susceptibility for the finite parabolic quantum well (PQW) is studied theoretically. An important feature of the finite PQW is that the subband eigenenergies \( E_n \) form an equally spaced energy-level ladder. Since the eigenenergy spacing \( \hbar \omega_0 \) can be designed to resonate with the pumping source, the third harmonic susceptibility can be greatly enhanced through the triple resonance. A third harmonic susceptibility as high as 2000 \((\text{nm/V})^2\) could be achieved for the finite parabolic quantum well structure. This is a more than six orders of magnitude enhancement as compared to that of the bulk GaAs.

Quantum confinement of carriers in a semiconductor quantum well leads to the formation of discrete subbands.\(^1\) Transitions between these subbands of an isolated quantum well have extremely large oscillator strength and relatively narrow line width. Strong infrared absorption features associated with transitions between subbands of the quantum well have been observed.\(^2-5\) This large dipole moment for the intersubband transition suggests a very large optical nonlinearity may exist for the semiconductor quantum well. Recently, very strong second order nonlinear effects have been observed in various asymmetric quantum-well structures.\(^6-10\) Those observed second harmonic susceptibilities are several orders of magnitude larger than that of a bulk semiconductor. Similarly, a very large third order nonlinear optical susceptibility can be expected in a quantum-well structure due to its fourth power dependence on the dipole moment. The observed third order intersubband nonlinearity at \( \lambda \approx 10 \mu\text{m} \) measured by S'aar et al.\(^11\) in a doped \( \text{Al}_0.4\text{Ga}_{0.6}\text{As}/\text{GaAs} \) quantum-well structure is about six orders of magnitude larger than that of the bulk GaAs.

Although the conventional square quantum well does give a large enhancement on the third order nonlinearities, such enhancement cannot be stretched too far due to the limitation of the single pair of resonant subbands. To have multiple pairs of resonant subbands, a finite parabolic quantum well (PQW) is employed in this article. The potential profile of a finite parabolic quantum well is shown in Fig. 1 and the potential energy \( U(z) \) can be expressed as:

\[
U(z) = \begin{cases} 
\frac{1}{2} Kz^2 & |z| < W/2 \\
U_b - \frac{1}{2} K(W/2)^2 & |z| > W/2.
\end{cases}
\]

\(^1\)To whom correspondence should be addressed.

The eigenenergy \( E_n \) of the \( n \)th subband is roughly equal to \((n+\frac{1}{2})\hbar \omega_0\), where \( q \) is the elementary charge, \( \hbar \) is the reduced Plank's constant, \( \omega_0 = \sqrt{(K/m^*)} \), and \( m^* \) is the effective mass of the electron. An important feature of this finite parabolic quantum well, as compared to the square quantum well, is that all subbands are equally spaced with eigenenergy spacing \( E_{n+1} - E_n = \hbar \omega_0 \). In this way, by a suitable choice of the spring constant \( K \), i.e., the composition gradient, the subband eigenenergy spacing \( \hbar \omega_0 \) of this finite PQW structure can be chosen to resonate with the pumping source and give a very large enhancement of the third harmonic susceptibilities \( \chi^{(3)}(3\omega) \). The value of spring constant \( K \) is chosen to be about 0.125 meV/Å\(^2\) to give eigenenergy spacing \( \hbar \omega_0 \approx 117 \text{ meV} \) in resonance with the CO\(_2\) 10.6 \( \mu\text{m} \) laser line.

From the density matrix formalism, the analytic formulas of the third harmonic susceptibilities can be expressed as:\(^12\)

![FIG. 1. Schematic diagram of a finite parabolic quantum well. The well width is 170 Å and \( K=0.125 \text{ meV/Å}^2 \). The barrier height \( U_b=451 \text{ meV} \). Subband energy levels and their associated envelope wave functions are also displayed.](image-url)
where $N$ is the electron density in the quantum well, $W_{nm}=(E_n-E_m)/\hbar$, and $\Gamma_{mn}=\tau_{nm}$ is the dephasing time between the state $|\psi_n\rangle$ and the state $|\psi_m\rangle$. The dipole moment matrix element $M_{nm}$ is given by

$$M_{nm}=\int_{-W/2}^{W/2} \psi_n^*(z)qz\psi_m(z)dz,$$

where $\psi_n(z)$ is the $n$th subband envelope wave function of the finite PQW solved by the transfer matrix method. The unperturbed density matrix $\rho_{nn}(0)$ is equal to the thermal equilibrium occupation probability of the corresponding state. From Eq. (2), if the relevant energy levels are equally spaced, then the $\chi^{(3)}(3\omega)$ can be greatly enhanced through the multiple resonance.

The calculated $|\chi^{(2)}(3\omega)|$ as a function of the well width $W$ is shown in Fig. 2. All the numerical calculations done in this article are based on the following parameters unless otherwise stated: the pumping source is 10.6 $\mu$m CO$_2$ laser line, $T=77$ K, $m^*=0.067 m_0$, and all of the dephasing time $\tau_{nm}$ is assumed to have the same value of 0.14 ps. The AlGaAs/GaAs material system is assumed for the finite PQW structure and the density of the two dimensional electron gas $N_{2DEG}$ in the PQW is assumed to be $1.2 \times 10^{12}$ cm$^{-2}$. In Fig. 2, two steps located at $W_1=127$ Å and $W_2=155$ Å could be observed. This is an expected result and could be understood from the following argument. As the well width $W$ is broadened, the barrier height $U_b$ will be raised. As the barrier height $U_b$ raised over a step of $\hbar\omega_0$, one more subband will be accommodated in the PQW. Since all these subbands of the PQW are equally spaced, this additional subband will contribute one more resonant level and the $|\chi^{(3)}(3\omega)|$ will be enhanced. For the quantum-well width $W<127$ Å, only two subbands are allowed in the PQW (two-level system), the $|\chi^{(3)}(3\omega)|$ is enhanced through the single resonance. For $127$ Å $< W < 155$ Å, three subbands can be accommodated in the PQW (three-level system), and the $|\chi^{(3)}(3\omega)|$ moderately enhanced through the double resonance. The step located at $W_1$ is a consequence of the transition from the single resonance to the double resonance. As for the quantum-well width $W>155$ Å, four subbands can be resided in the PQW (four-level system). The $|\chi^{(3)}(3\omega)|$ is greatly enhanced through the triple resonance. The step located at $W_2$ is a consequence of the transition from the double resonance to the triple resonance. A third order nonlinear optical susceptibility $|\chi^{(3)}(3\omega)|$ as high as 2000 (nm/V)$^2$ can be achieved through the triple resonance for the finite PQW structure with $W=170$ Å and $K=0.125$ meV/A$^2$.

This is a more than six orders of magnitude improvement as compared to the $|\chi^{(3)}(3\omega)|$ of the bulk GaAs. For a finite PQW, the subband levels may not be exactly equally spaced, but the necessary resonance can still be achieved by tuning the spring constant $K$ for a fixed well width $W$. In order to have a fair comparison of the effect of well width $W$ on the $|\chi^{(3)}(3\omega)|$, three kinds of structures with well widths $W=100,146,$ and 155 Å are chosen. The $|\chi^{(3)}(3\omega)|$ is individually maximized by adjusting the spring constant $K$ for each PQW structure. These optimized results are summarized in Table I. It is evident from Table I that the $|\chi^{(3)}(3\omega)|$ depends slightly upon the

<table>
<thead>
<tr>
<th>Well Width W (Å)</th>
<th>100</th>
<th>146</th>
<th>155</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\chi^{(3)}(3\omega)$ (nm/V)$^2$</td>
<td>200</td>
<td>584.6</td>
<td>270.1</td>
</tr>
<tr>
<td>$\Delta E_{31}$ (meV)</td>
<td>0.125</td>
<td>0.129</td>
<td>0.169</td>
</tr>
<tr>
<td>$\Delta E_{13}$ (meV)</td>
<td>119.0</td>
<td>119.7</td>
<td>116.9</td>
</tr>
<tr>
<td>Resonant status</td>
<td>Triple</td>
<td>Double</td>
<td>Single</td>
</tr>
</tbody>
</table>

TABLE I. Summary of the calculated results.
spring constant $K$ and the triply resonant structure gives the largest $|\chi^{(3)}(3\omega)|$.

The $|\chi^{(3)}(3\omega)|$ as a function of the pumping wavelength $\lambda$ for the finite PQW structure with $W=170$ Å and the Al$_{0.4}$Ga$_{0.6}$As/GaAs square quantum-well structure with a well width of 85 Å are plotted in Fig. 3. For the square quantum-well structure, $\Delta E_{21} = E_2 - E_1 \approx 124$ meV and $\Delta E_{31} = E_3 - E_1 \approx 147$ meV, the corresponding two resonant peak of the $|\chi^{(3)}(3\omega)|$ located at $\lambda \approx 8.5$ and 10 $\mu$m can be observed in this figure. Since $\Delta E_{31} \neq 2\Delta E_{21}$, this is thus a single resonant process. As a result, both peaks of the $|\chi^{(3)}(3\omega)|$ for the square quantum-well structure are less than one sixth of the peak value of $|\chi^{(3)}(3\omega)|$ of the PQW structure. As for the finite PQW with $W=170$ Å, $\Delta E_{41} \approx 3\hbar \omega$, $\Delta E_{31} \approx 2\hbar \omega$, and $\Delta E_{21} \approx 2\hbar \omega$ (Table I), only one resonance peak of the $|\chi^{(3)}(3\omega)|$ located at $\lambda \approx 10.6$ $\mu$m is observed and the peak value of the $|\chi^{(3)}(3\omega)|$ of this PQW structure is improved by sixfold as compared to that of the square quantum well. With the appropriate choice of the spring constant $K$, the $|\chi^{(3)}(3\omega)|$ of the PQW could also be resonantly enhanced for a pumping source other than the 10.6 $\mu$m CO$_2$ laser.

In conclusion, the parabolic quantum well has been employed to greatly enhance the third harmonic susceptibility at 10.6 $\mu$m by triple resonance. When the triple resonant condition is achieved, the maximum $|\chi^{(3)}(3\omega)|$ of the finite PQW can reach a value as high as 2000 (nm/V)$^2$. This triply resonant method used to enhance the third order nonlinear susceptibility is also valid for other quantum well structures that possess equal subband spacing. This indicated a wide possibility of synthesizing new third order optical nonlinear materials. In addition, the assumption of the AlGaAs/GaAs material system is not essential in this calculation; any quantum well system that can be designed to meet the resonant enhancement condition will give a large third harmonic generation susceptibility. The GaInAs/AlInAs system is of particular interest due to the larger conduction band edge offset.

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