Quantum Box Fabrication Tolerance and Size Limits in Semiconductors and Their Effect on Optical Gain

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Abstract—Lower and upper limits on size are established for quantum boxes. The lower limit is shown to result from a critical size below which bound electronic states no longer exist. This critical size is different for electrons and holes. The optical gain of arrays of quantum boxes is computed taking into account the inhomogeneous broadening of the gain spectrum resulting from fabrication variations in quantum box size and shape. The dependence of maximum possible gain on an rms quantum box roughness amplitude is determined. For high gain operation a medium composed of quantum boxes does not offer significant advantages over a conventional bulk semiconductor unless quantum box fabrication tolerances are tightly controlled. For low gain operation, however, arrays of quantum boxes may offer the unique advantage of optical transparency at zero excitation. This property does not require excellent fabrication control and may make possible ultralow threshold semiconductor lasers and low noise optical amplifiers.

INTRODUCTION

DIRECT write lithography technology is now capable of fabricating structures having nanometer scale feature sizes. If combined with epitaxially-grown quantum well material in systems like GaAs(AlGaAs) or InGaAsP(InP) structures exhibiting quantum size effects in two or three dimensions can be contemplated. Quantum wells and quantum boxes are examples of such structures [1]–[4].

It has been proposed that arrays of quantum wires or quantum boxes might be used as the active layer of a semiconductor laser [5]. In these devices the quantum wires or quantum boxes, composed of a low band-gap material like GaAs, would be imbedded in a higher band-gap material like AlGaAs (see Fig. 1). Theoretical studies indicate the potential advantages of these structures are reduced threshold current densities [5], [6], improved phase noise characteristics (i.e., reduced field spectrum line-widths) [7], [8], and improved $T_{2e}$'s [5]. Some of these effects have been observed by simulating quantum boxes and quantum wires using high magnetic fields [7], [8].

These improvements result from the narrower optical gain spectrum provided by these structures. In essence, the importance of Fermi statistics in shaping the gain pro-

Fig. 1. A quantum box semiconductor laser in the GaAs(AlGaAs) system. The quantum boxes are not drawn to scale.
only the natural linewidth of the optical transition (homogeneous broadening). This paper will focus on the quantum boxes as a source of optical gain. It differs from previous treatments of this subject in that the practical problems and limitations affecting the optical gain attainable with an array of quantum boxes are addressed. There are a host of problems associated with fabricating quantum box active layers which we must assume to be surmountable. There remain, however, basic theoretical questions concerning the quantum box volumetric density required to furnish a given optical gain, the limitations imposed on quantum box size, and the tolerances which must be maintained during fabrication. Fabrication tolerance will be of central importance since it determines the degree to which the optical gain spectrum is inhomogeneously broadened (see Fig. 3). It may be the case that a quantum box laser is theoretically feasible, but highly unrealistic in terms of the required fabrication tolerances. For example, since two dimensions of the quantum box are defined by a combination of beam writing and subsequent processing, it would be highly unrealistic to assume that these dimensions could be held to the same tolerance that can be achieved in the other direction by epitaxial means (e.g., held to a monolayer or less with molecular beam epitaxy).

In this paper we attempt to answer these questions. We will calculate the optical gain achievable with a given density of quantum boxes and study the effect of fabrication tolerance on the optical gain. In addition to studying optical gain we will address the issue of upper and lower limits on quantum box size, establishing allowable ranges and introducing a critical quantum box radius below which there are no bound electronic states in a quantum box. Two regimes of quantum box size will be discussed below. The behavior of optical gain with pumping will be seen to depend on the particular quantum box regime selected.

**Quantum Box Size Limitations**

It is necessary to establish what the characteristic sizes of quantum boxes must be and to determine if there are limits imposed on the range of allowable sizes. The model of quantum boxes employed here will be based on the effective mass approximation and will assume the carriers in the box behave as particles in a spherical well. The spherical well potential, unlike the three-dimensional square well potential employed elsewhere is separable and thus lends itself to simplified analytical solutions. Electronic states in these structures are given by the kets \(|k, j, m\rangle\), where \(j\) is the quantum number of total angular momentum, \(m\) is the \(z\)-component of angular momentum, \(k\) is a quantum number associated with radial motion and the subscript \(n\) is an energy band index. An optical dipole transition from the conduction band to the valence band conserves \(j\) and \(m\) in the single electron approximation. This rule differs from its free-space counterpart because the \(j\) and \(m\) employed here refer to envelope functions in the effective mass approximation. \(k\) is also conserved provided that the wave function does not extend significantly outside of the well. A transition from the lowest energy eigenstate of the conduction band will therefore be to the highest energy eigenstate in one of the respective valence bands. This rule will be assumed to hold rigorously throughout this analysis.

The two lowest energy eigenstates occur for \(j = 1/2, m = \pm 1/2\). These correspond to the two spin states having zero orbital angular momentum. The radial quantum number for these states obeys a particularly simple eigenvalue equation which upon solution of the Schrodinger equation in spherical coordinates is easily shown to be [11]

\[
\sqrt{\frac{2m_e V_0}{\hbar^2} - k^2} = -k \cot (kR)
\]  

where \(m_e\) is the effective mass, \(V_0\) is the depth of the well, and \(R\) is the radius of the quantum box. The eigenenergy
$E$ is related to $k$ through the familiar expression,

$$E = \frac{\hbar^2 k^2}{2m_e}. \quad (2)$$

Equations (1) and (2) are similar to the eigenvalue equations for a one-dimensional potential well of width $w$ and depth $V_e$ if we make the correspondence $R \rightarrow w$. In fact, the eigenvalues of (1) correspond to the odd parity solutions in the one-dimensional well. Consequently, whereas the one-dimensional problem has a bound state for all $w$ (i.e., the lowest energy even parity solution), there exists a critical radius $R_c$ in the three-dimensional problem below which there are no bound states. For the spherical well this critical radius is given by

$$R_c = \frac{\pi}{2} \frac{\hbar^2}{2m_e V_e}. \quad (3)$$

Three-dimensional wells of arbitrary shapes should also have such a critical size. This fact seems to have gone unnoticed thus far in the study of quantum boxes. It has important consequences, one of which is illustrated in Fig. 4. There, (3) has been used to plot the critical quantum box diameter against the energy-gap difference between GaAs and AlGaAs. We have assumed effective masses of 0.067 $m_e$ for electrons and 0.47 $m_e$ for heavy holes in GaAs with $m_e$ the free electronic mass. Furthermore, a 60-40 percent apportioning of the band-gap offset energy is assumed. Note that because of the large difference in the electron and hole effective masses there is a range of diameters for which there remain bound holes, but no bound electrons.

$R_c$ for the electrons sets a lower limit on quantum box size. The upper limit is set by the need to create a "discrete" state space in at least one of the energy bands. A state space will appear discrete at a given temperature $T$ if the energy spacing between states is larger than $k_B T$ (26 meV at room temperature). In a quantum box laser it is actually better to have the energy spacing still larger, since ideally only the two lowest energy states in a given band should contribute to lasing. As already noted, in the conduction band the lowest energy eigenstates are states of zero orbital angular momentum (i.e., $l = 0$). The next highest energy eigenstates, however, occur for $l = 1$. For the infinite potential well their energy separation from the lowest energy states is given by the expression

$$\Delta E_{1,0} = 10.3 \frac{\hbar^2}{2m_e R^2}. \quad (4)$$

For conduction band electrons in GaAs if $\Delta E_{1,0} = 3k_B T_{room}$ then $R = 87 \text{ Å}$ (i.e., a quantum box diameter $d$ of 174 Å). At this energy separation the sixfold degenerate $l = 1$ states will have an electron Fermi occupancy of 0.047 when the Fermi level resides at the energy of the two lowest energy states. Consequently, for practical considerations involving computation of threshold gain, etc., the conduction band in a structure this small can be treated as one doubly-degenerate energy level.

At this same diameter, however, the heavy hole system will remain a quasi-continuum owing to the heavy hole effective mass. This range of sizes will be referred to as the large quantum box regime. To achieve a discrete spectrum of states in both the conduction and valence bands the quantum box size must be reduced still further to what we call the small quantum box regime. For the purposes of demonstration we will neglect problems associated with valence band mixing effects and consider only the heavy hole band. To achieve a $\Delta E_{1,0} = 3k_B T_{room}$ for the heavy hole band in GaAs would require a quantum box diameter of 66 Å. If we tolerate an increased occupancy of lower energy hole states by reducing $\Delta E_{1,0}$ to $\Delta E_{1,0} = k_B T_{room}$ then a diameter of 114 Å is possible. In view of Fig. 4 it is clear that selection of such small diameters must be accompanied by an adequately deep well to guarantee the existence of bound electron states. The results of this section are summarized in Fig. 5. In particular, the small and large quantum box regimes are given at several temperatures for the material systems GaAs/AlGaAs and GaInAs/InP. The latter system has been chosen because of its very light electronic effective mass (0.04$m_e$).

There are important differences between active layers composed of large quantum boxes and small quantum boxes. These will be addressed below.

**Fabrication Tolerance and the Gain Spectrum**

A quantum box semiconductor laser can be viewed as a gas laser in which the atoms are likened to the quantum boxes. The overall gain spectrum produced by this gas is broadened both homogeneously by the natural linewidth...
It is therefore reasonable to assume that the system is composed of several hundred thousand quantum boxes. Broadening resulting from the variations in quantum box size is even the equivalent of pressure broadening in this system, since when the quantum box density becomes too large the state space broadens as a result of interaction between electrons in different quantum boxes. In this section the gain spectrum resulting from a given volumetric density of quantum boxes will be calculated.

The optical transition from conduction band state to valence band state in each quantum box is assumed to be homogeneously broadened according to a broadening function \( g(E) \). In addition we must account for variations in transition energy resulting from the varying sizes of the quantum boxes. This produces an inhomogeneous broadening of the gain spectrum which we describe using a distribution function \( D(E) \) (in effect, a density of states function) that is related to the distribution of quantum box sizes. The gain coefficient per unit time at frequency \( \omega \) is related to these functions through the following expression [12]:

\[
G(N, \omega) = 2 \int dE (E - E_c) g(h\omega - E) \cdot \left| M \right|^2 \left( f_c(N, E) - f_v(N, E) \right)
\]

(5)

where \( E_c \) is the average transition energy, \( M \) is the matrix element for a quantum box transition, \( N \) is the carrier density, and \( f_c(f_v) \) is the Fermi factor for the conduction (valence) band. A typical quantum box active layer will be composed of several hundred thousand quantum boxes. It is therefore reasonable to assume that \( D(E) \) is a Gaussian distribution function of the form

\[
D(E) = \frac{2\eta}{\gamma \sqrt{2\pi}} \exp \left( -\frac{1}{2} \left( \frac{E - \bar{E}}{\gamma} \right)^2 \right)
\]

(6)

where \( \eta \) is the number of quantum boxes per unit volume (note: this quantity can have a spatial dependence) and where the normalization factor for \( D(E) \) includes a factor of two to account for spin degeneracy.

The quantity \( \gamma \) gives a measure of the inhomogeneous broadening resulting from the variations in quantum box sizes (see Fig. 3). It is important to relate this quantity to actual statistical variations in quantum box size. Smearing of the optical transition energy will result primarily from variations of the lowest conduction band eigenenergy, because of the small conduction band effective mass. To simplify the calculation we therefore consider only variations in the conduction band eigenenergy that result from quantum box size variation (i.e., variations of valence state energies are ignored). Obviously, fluctuations in size and shape of a quantum box can take on a very complicated form as illustrated in Fig. 3. Therefore, to proceed consider first a simple variation \( \delta R \) of a perfectly spherical well.

If the electron is tightly bound to the quantum box, as a result of either the well potential being deep or the quantum box being large, then it is adequate to approximate the well as infinitely deep. The lowest eigen-energy is then given by

\[
E_c^\infty = \frac{\hbar^2 \pi^2}{2m_R^2}
\]

(7)

and therefore the associated energy shift in the optical transition due to the variation \( \delta R \) is given by

\[
\delta E = -2E_c^\infty \frac{\delta R}{R}
\]

(8)

In general, the potential cannot be treated as infinite. It is straightforward to show using (1) that a more general expression is given by

\[
\delta E = -\xi E_c \frac{\delta R}{R}
\]

(9)

where \( \xi < 2 \) approaches zero as the electron becomes more weakly bound.

To include the possibility of more complex shape and size variations we introduce an amplitude roughness function \( \rho(\theta, \phi) \) which characterizes deviations from the ideal spherical well (see Fig. 3). We will assume that the actual well potential has the following form as a result of imperfections in the fabrication process:

\[
V(r) = Y_\ell U(r - R - \rho(\theta, \phi))
\]

(10)

where \( U(r) \) is the Heaviside function. Upon application of first-order perturbation methods this yields the following expression for the resulting shift in the optical transition energy

\[
\delta E = -V_\rho \left| \psi(R) \right|^2 \frac{\delta \rho}{R}
\]

(11)

where \( \bar{\rho} \) is the average of the roughness function \( \rho(\theta, \phi) \) over \( 4\pi \) steradians and where \( \psi(R) \) is the eigenfunction of the lowest energy conduction band state. In the limit of an infinite potential well (i.e., \( V_\rho \to \infty \)) it can be shown that this expression reduces to the following form:

\[
\delta E = -2E_c^\infty \frac{\bar{\rho}}{R}
\]

(12)

where \( E_c^\infty \) is given by (7). This is similar to (8) with \( \bar{\rho} \) replacing \( \delta R \). We will assume this relation also holds in...
the finite well. Finally, it is helpful to relate $\bar{\rho}$ to a roughness amplitude $\rho_0$ (see Fig. 3) by assuming a particular form for $\rho(\theta, \phi)$. For the purposes of demonstration we will assume that $\rho(\theta, \phi) = \rho_0 \cos^a \theta \cos^b \phi$. For $a$ and $b$ large enough we obtain $\bar{\rho} = \rho_0 / 4$. Using this result and (9), the characteristic inhomogeneous linewidth, $\gamma$, can be expressed as

$$\gamma^2 = \langle \delta E^2 \rangle = \frac{\langle \delta E_x^2 \rangle}{\langle \delta E_y^2 \rangle} \frac{\langle \delta E_x^2 \rangle}{4R} \theta^2$$

where $\langle \cdot \rangle_x$ indicates an average over the ensemble of quantum boxes and where $\theta^2 \equiv \langle \rho_0^2 \rangle_x$. That is, $\theta$ is an rms roughness amplitude for the ensemble of quantum boxes.

To simplify (5) we will assume that the homogeneous broadening function $g(E)$ is also a Gaussian of the following form:

$$g(E) = \frac{\hbar}{\delta} \sqrt{\frac{\pi}{2}} \exp \left( -\frac{1}{2} \left( \frac{E}{\delta} \right)^2 \right).$$

The normalization here is chosen so as to recover Fermi's Golden Rule in (5) in the limit $\delta \to 0$. Using this form, the gain spectrum is given by

$$G(N, \omega) = \frac{4\pi |M|^2 \hbar}{\sqrt{2\pi}} \sqrt{\frac{\gamma^2 + \delta^2}{\gamma^2 + \delta^2}} \left( f_s(N, E_o) \right)$$

$$- f_s(N, E_o) \exp \left( -\frac{(\hbar \omega - E_o)^2}{2(\gamma^2 + \delta^2)} \right)$$

where we have assumed both the matrix element and the quasi-Fermi factors are approximately constant over the width of the quantum box gain spectrum. This expression will now be studied for the two size regimes discussed in the previous section.

Small Quantum Boxes

Small quantum boxes have dimensions that cause the energy spectrum in the both the conduction and valence bands to be discrete. Equation (15) can be written in terms of the carrier density $N$ by noting that $N = 2f_c \eta$ and by assuming that the quantum boxes are, on the average, quasi-neutral and undoped (i.e., $f_s = 1 - f_c$). Doing so yields

$$G_s = A_s(N - \eta) \exp \left( -\frac{(\hbar \omega - E_o)^2}{2(\gamma^2 + \delta^2)} \right)$$

$$A_s = \frac{4 \pi |M|^2 \hbar}{\sqrt{2\pi}} \sqrt{\frac{\gamma^2 + \delta^2}{\gamma^2 + \delta^2}}. \tag{17}$$

The gain is therefore linear in carrier density and abruptly saturates when all quantum boxes are filled (i.e., $N = 2\eta$). This is illustrated in Fig. 6 where $G_s$ is plotted versus $N$. Transparency of the active layer occurs at the excitation level $N = \eta$.

It is also possible to calculate the optical gain when the quantum boxes have been doped. $f_c$ or $f_o$ will then be biased by the presence of donors or acceptors. In Fig. 6 we have plotted the case of doping at a level of two donors or acceptors per quantum box. The differential gain $A_s$ is reduced by doping. Notice, however, that a significant improvement in the transparency level has occurred under doping. In fact, a nearly zero transparency level is expected. This result has important consequences to the design of lasers and optical amplifiers. For lasers it means there is no transparency penalty in achieving threshold. Extremely low threshold semiconductor lasers are therefore possible by simply reducing loss. In amplifiers, this eliminates the spontaneous noise associated with achieving transparency and hence reduces the overall amplifier noise at a given gain.

In Fig. 7 $G_s$ as a function of carrier density has been plotted for the specific case of an array of quantum boxes having an average diameter $d = 100 \text{ Å}$, center to center spacing $200 \text{ Å}$ (i.e., $\eta = 1.25 \times 10^{17} \text{ cm}^{-3}$), and a fabrication tolerance $\delta = 20 \text{ Å}$. Gain has been plotted in both temporal and spatial rate units. In performing this calculation we have used [12]

$$|M|^2 = \frac{E_o}{2e \hbar^2} |\mu|^2 \tag{18}$$

where $\epsilon$ is the dielectric constant of the material and $\mu$ is the component of the dipole matrix element in the direction of the electric field vector. In accordance with the results contained in [6], $|\mu|/q = 4 \text{ Å}$, corresponding to the value in bulk GaAs, has been employed here. In addition $|\mu|^2$ has no field polarization dependence owing to the spherical symmetry of these lowest energy box states.

Large Quantum Boxes

A large quantum box falls at the other end of the range indicated in Fig. 5. It has a conduction band energy spectrum that is discrete, but its valence band remains a quasi-continuum. This makes it more difficult to invert the quantum box to achieve optical gain, since a large number of holes must be added to the structure to produce an ap-
preciable hole occupancy in the highest energy state in the valence band. By quasi-neutrality these holes must be balanced by an equal number of electrons. The net effect is thus to increase the required threshold carrier density. To see this consider an undoped quantum box of diameter \(d = 200 \, \text{Å}\). Suppose that this structure is quasi-neutral and contains a single electron and a single hole. Upon intraband thermalization the electron will occupy the lowest energy level of the conduction band. The hole occupancy, however, is smeared out over a larger number of valence band states owing to the narrow energy spacing of the heavy hole states.

The occupancy \(f_h\), at a given energy, is related to the hole occupancy \(f_e\) by the expression \(f_h = 1 - f_e\). In Table I \(f_h\) has been tabulated for various numbers of holes in this quantum box. The equivalent hole density is also given. The heavy hole effective mass in GaAs has been used for this calculation and we have assumed the hole state space to be a quasi-continuum. The optical gain of this structure will be proportional to \(f_e - f_h\), the difference in electronic occupancies between the lowest energy conduction band state and the highest energy valence band state. Ideally, to achieve maximum gain we would like \(f_e = 1\) and \(f_h = 0\). For the present case of one electron and one hole in the quantum box, however, \(f_e = 1\) and \(f_h = 0.97\), resulting in a greatly-reduced optical gain. For this size quantum box the large effective mass of the heavy holes therefore undermines the potential advantages offered by the structure. To circumvent this problem the quantum boxes could be p-doped to bias \(f_e\) towards zero.

The optical gain of an array of large quantum boxes can also be expressed in terms of the injected carrier density \(N\) by using \(N = 2nf_e\) and by treating \(f_e\) as a fixed quantity dependent upon the p-doping level. Using (15) we find

\[
G_L = A_L(N - 2nf_e) \exp \left( -\frac{(\hbar \omega - E_L)}{2(\gamma^2 + \delta^2)} \right) \tag{19}
\]

\[
A_L = \frac{2 |M|^2 \hbar}{\sqrt{2\pi \sqrt{\gamma^2 + \delta^2}}} \tag{20}
\]

This function, like \(G_L\), is linear in carrier density and saturates when \(N = 2\gamma\). It is plotted in Fig. 8 for the various p-doping levels given in Table I. The effect of p-doping on the transparency level is apparent. In Fig. 9 a specific case is plotted in which the array contains quantum boxes having a diameter \(d = 200 \, \text{Å}\), center to center spacing of 400 Å (i.e., \(\eta = 1.56 \times 10^{16} \, \text{cm}^{-2}\)), and a fabrication tolerance \(\theta = 20 \, \text{Å}\). It should be noted that the introduction of heavy p-doping will produce strong Coulomb effects which will in turn alter the electronic statespace of the quantum box [13]. Such heavily-doped structures would become a variation of the so-called superatom [14]. These effects will not be addressed here.
boxes having a given fabricational tolerance. For the large old is possible it is necessary to estimate the maximum optical gain possible with a given density of quantum box array the maximum possible gain is those possible with the assumed quantum box diameters how these quantities vary with fabrication tolerance tum boxes. In general, large quantum boxes are less susceptible to the effects of fabrication as compared to small quantum boxes. As a result higher gains for given quantum box densities are possible as compared to the typical for an optical gain of 50 cm^{-1} arrays of quantum boxes do not offer significant advantages unless there is very good control of fabrication tolerance. For example, Fig. 9 indicates improvements in threshold carrier density exceeding two orders of magnitude (bulk carrier densities of 3 \times 10^{19} \text{cm}^{-3} are typical for an optical gain of 50 cm^{-1}) provided the rms roughness amplitude is held to 20 \text{Å} on an array of 200 \text{Å} diameter quantum boxes. Since 200 \text{Å} diameter quantum boxes have not yet been fabricated it is impossible to assign a difficulty factor to this specification. It most probably exceeds the state-of-the-art in direct-write lithography, however.

For low optical gains, however, a quantum box medium may offer a unique advantage that does not require extremely good fabrication control. This is a nearly-zero transparency level made possible by p-doping the quantum boxes. This property may make possible very low threshold semiconductor lasers provided that cavity losses are substantially reduced. It may also lead to optical amplifiers with improved noise characteristics. We will investigate these applications elsewhere.

**CONCLUSION**

Arrays of quantum boxes may one day be employed as sources of optical gain in lasers or amplifiers. We have established size limitations on the quantum boxes in these arrays and have studied the dependence of optical gain on quantum box fabrication tolerance. The lower limit on quantum box size is set by the disappearance of bound electronic states at a critical diameter. We have calculated this critical diameter for the spherical quantum box and have shown that it differs for electrons and holes owing to the difference in their effective masses. The upper limit on quantum box size is set by the need to generate a discrete spectrum of states in one or both of the principal energy bands. When both the conduction band and valence band spectra are discrete, the quantum box is in the small regime. When only the conduction band spectrum is discrete the quantum box is in the large regime. The optical properties of quantum box arrays is regime dependent. Each regime has its own advantages and disadvantages. For example, it would appear unlikely that an array of quantum boxes would provide the necessary gain for lasing action (even under high excitation) unless it is heavily p-doped. An array of small quantum boxes, on the other hand, can provide gain without being doped. Large quantum boxes, however, are easier to fabricate and once p-doped should attain higher optical gains for a given quantum box density at a given excitation level. This is a direct result of their gain spectra being less sensitive to fabrication induced broadening.

To compare quantum box arrays and bulk semiconductor material in terms of their optical gain at a given excitation level, it is useful to distinguish between the cases of high and low gain. To achieve the very high gains typical of a conventional semiconductor laser (e.g., tens of cm^{-1}), arrays of quantum boxes do not offer significant advantages unless there is very good control of fabrication tolerance. For example, Fig. 9 indicates improvements in threshold carrier density exceeding two orders of magnitude (bulk carrier densities of 3 \times 10^{19} \text{cm}^{-3} are typical for an optical gain of 50 cm^{-1}) provided the rms roughness amplitude is held to 20 \text{Å} on an array of 200 \text{Å} diameter quantum boxes. Since 200 \text{Å} diameter quantum boxes have not yet been fabricated it is impossible to assign a difficulty factor to this specification. It most probably exceeds the state-of-the-art in direct-write lithography, however.

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Kerry J. Vahala (S'82–M'84–S'84–M'85), photograph and biography not available at the time of publication.