Luminescence spectra of a quantum-dot cascade laser

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A quantum cascade laser in which the quantum wells in the active regions are replaced by quantum dots with their atom-like discrete energy levels is an interesting system with which to study novel features in optical spectroscopy. We study structures suitable for diagonal lasing transitions in coupled dots, and vertical transitions in a single dot. The luminescence spectra as a function of electron number and dot size show that for diagonal transitions a significant amount of blueshift in the emission spectra can be achieved by increasing the electron population in the quantum dots as well as by decreasing the size of the dots. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1357808]

Ever since the original work on quantum cascade laser (QCL) by Faist *et al.*[1] in 1994, the unipolar semiconductor laser based on intersubband transitions in coupled quantum wells has undergone rapid development. QCLs created in InGaAs/AlInAs systems have achieved record high power outputs in the midinfrared range that has the potential for wide-ranging applications.[2,3] QCLs in other material systems[4] have also shown promise of improved performance.

In this letter, we report on our study of the optical properties of quantum cascade structures in which the quantum wells in the active regions are replaced by quantum dots (QDs). The latter, popularly known as artificial atoms,[5] where electron motion is quantized in all three spatial directions, have been receiving wide attention. These zero-dimensional quantum confined systems are useful for investigating the fundamental concepts of nanostructures[5,6] as well as for its application potentials. In recent years, there has been considerable progress in quantum-dot laser research.[7] Because of their discrete atom-like states, quantum-dot lasers are expected to have better performance than quantum-well lasers.[8] Development of self-organizing growth techniques that allows formation of high-density of quantum dots with nanometer dimensions rapidly enhanced the development of QD-laser research, where the performance is now comparable to that of quantum-well lasers.[9] Researchers have also found important applications of quantum dots in storage devices[10] and fluorescence markers.[11]

Here we combine the properties of these two very interesting nanostructures, the QCLs and the QDs, to explore the luminescnece spectra of a quantum-dot cascade structure. There have already been some suggestions in the literature that quantum-dot cascade lasers will significantly reduce the threshold current density by eliminating single phonon decay.[12] This prediction was based on the fact that quantization of electron motion in the plane would greatly inhibit single phonon decay, provided the dots are sufficiently small. There is however no report in the literature (theoretical or experimental) as yet on the physical properties of a quantum-dot cascade laser structure. In this work, we have explored the luminescnece spectra of quantum cascade structures, both for vertical as well as diagonal lasing transitions, for various values of the dot size and the number of electrons in the quantum dots. One advantage of the quantum-dot cascade laser structure for theoretical studies over the quantum-well cascade laser is that, for few electrons in the QD, most of the physical properties can be calculated exactly, albeit numerically.[5]

The single-electron Hamiltonian for our system is

$$\mathcal{H}' = \frac{p_x^2}{2m^*} + \frac{p_y^2}{2m^*} + V_{\text{plane}}(x,y) + \frac{p_z^2}{2m^*} + V_{\text{conf}}(z),$$

where the confinement potential in the z direction (Fig. 1) is

$$V_{\text{conf}}(z) = -eFz + \begin{cases} 0 & \text{for wells,} \\ U_0 & \text{for barriers,} \end{cases}$$

with F being the electric field in the z direction. The confinement potential in the xy plane is

$$V_{\text{plane}}(x,y) = \begin{cases} 0 & |x| < L/2 \text{ and } |y| < L/2, \\ U_0 & \text{otherwise.} \end{cases}$$

In our calculations that follow, we have used the separation of $V_{\text{plane}}(x,y)$ for x and y motion which is not correct in the regions $|x|>L/2, |y|>L/2$. However, in these regions the wave function is exponentially small. In fact, our estimates indicate that this approximation will modify the results only slightly (e.g., corrections to all energies are less than 1%). We consider only two subbands in the z direction ($k=1,2$) and for a given subband index k, all possible states in the xy plane with the condition, $E_{nmk} < U_0$, where E_{nmk} is the single-electron energy. The integers n and m correspond to the level indices in the x and y directions, respectively. Solutions of the Schrödinger equation in the z direction are obtained numerically for the two lowest states (subbands) shown in Fig. 1. Due to the $x \leftrightarrow y$ symmetry, some of the levels are twofold degenerate (for example, $E_{122}=E_{212}$). From the single-electron basis functions, we construct the N-electron basis, and the Hamiltonian matrix with Coulomb interaction among the electrons is then calculated on that basis. The eigenvalues and eigenfunctions were calculated by exact (numerical) diagonalization of the Hamiltonian ma-

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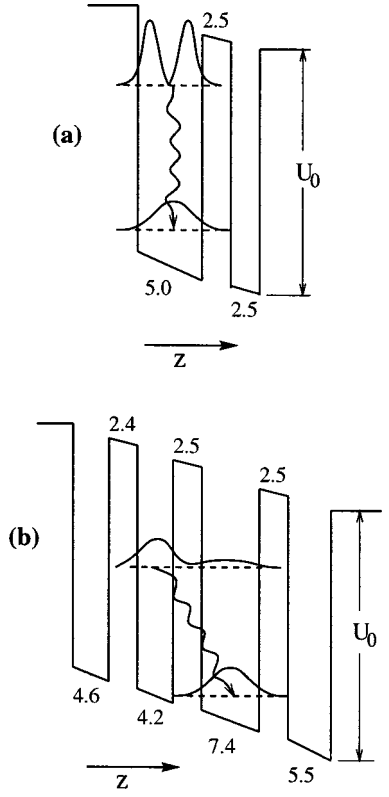

FIG. 1. Energy band diagram (schematic) of the active region of a quantum cascade structure and (a) vertical lasing transition under an average applied electric field of 85 kV/cm and (b) diagonal transition under a field of 55 kV/cm. The relevant wave functions (moduli squared) as well as the transition corresponding to the laser action are also shown schematically. The numbers (in nm) are the well ($Ga_{0.47}In_{0.53}As$) and barrier ($Al_{0.48}In_{0.52}As$) widths. The material parameters considered in this work are electron effective mass m_e^* ($Ga_{0.47}In_{0.53}As$) $=0.043\,m_0$, m_e^* ($Al_{0.48}In_{0.52}As$) $=0.078\,m_0$, the conduction band discontinuity, $U_0=520$ meV, and the nonparabolicity coefficients, $\gamma_w=1.3\times10^{-18}\,\mathrm{m}^2$ for the well and $\gamma_b=0.39\times10^{-18}\,\mathrm{m}^2$ for the barrier.

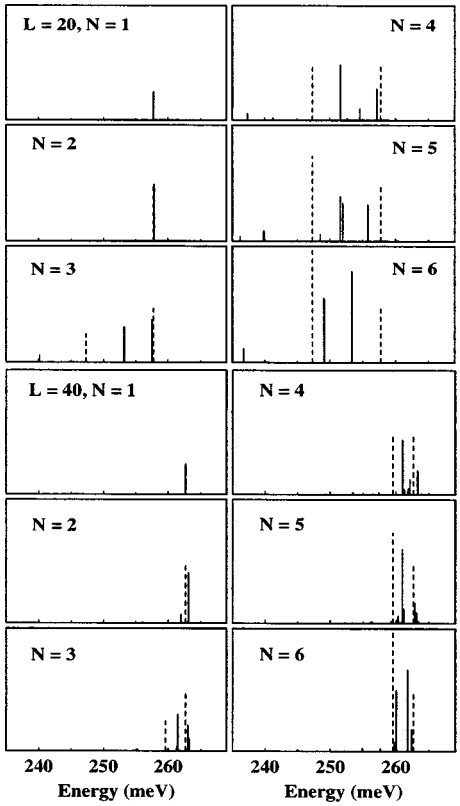

FIG. 2. Luminescence spectra of a quantum-dot cascade structure and *vertical* optical transitions for various values of the dot size (L in nm) and number of electrons (N) in the dot. The dashed lines correspond to the luminescence of noninteracting electron systems.

trix. As the manybody Hamiltonian also has the $x\leftrightarrow y$ symmetry some of the states are twofold degenerate.

In the initial state (before optical emission) all electrons are in the second subband, $k=2$. In the final state (after optical emission) one electron is in the first subband, $k=1$, and all other electrons are in the second subband, $k=2$. The intensity of optical transitions is calculated from

$$\mathcal{I}_{if}(\omega)=\frac{1}{Z}\sum_{if}\ \delta(\omega-E_i+E_f)\Big|\int\ \chi_1(z)z\chi_2(z)dz \times\int\ \Phi_i^*(x_1y_1,\cdots,x_Ny_N) \times\Phi_f(x_1y_1,\cdots,x_Ny_N)dx_1dy_1\cdots dx_Ndy_N\Big|^2 \times\exp(-\beta E_i),$$

where $Z=\Sigma_i\, e^{-\beta E_i}$ is the partition function and $\beta=1/kT$. In all our computations, we take $T=20$ K.

In Fig. 2, the optical spectra are shown for vertical transitions [Fig. 1(a)] for two sizes of quantum dots: $L=20$ and 40 nm, and for different numbers of electrons in the quantum dot. In all these cases the first moment of the emission spectra for the interacting system is almost the same as that for the noninteracting system. This is bacause in the case of vertical transitions Coulomb interaction between the electrons in the second subband and those in the second and the first subbands is almost the same (electrons are localized in the same quantum well in the z direction). The Coulomb interaction between electrons is about half the energy separation between the one-electron states in the xy plane for $L=20$ nm and is of the same order as the energy separation between xy levels in $L=40$ nm. That is why the interaction is more important for $L=40$ nm. For the two-electron system there is a small blueshift of the emission line due to the interaction which increases with an increase of the size of the quantum well. In addition, there is also a small red satellite at $L=40$ nm. For the six-electron system, the interaction results in redistribution of the intensities between peaks: the higher energy peak becomes more intense than that for the lower energy. For three, four and five electrons in a noninteracting system, we have a degenerate initial state. The degeneracy is lifted by the interaction and for the four-electon system the initial ground state is partially polarized as expected from Hund's rules. The interaction also results in the appearance of satellites and at the same time the separation between the main peaks becomes smaller for the interacting system than for the noninteracting case.

In Fig. 3 the optical spectra are presented for diagonal transitions [Fig. 1(b)] and for $L=10$ and 20 nm, and for different numbers of electrons. Interestingly, we notice the behavior characteristics of fully filled shells for two and six electrons. For the two-electron system, we have a single line for both noninteracting and interacting systems. For the six-electron system and $L=10$ nm, the emission spectra have the same two-peak structure as that of the noninteraction system.

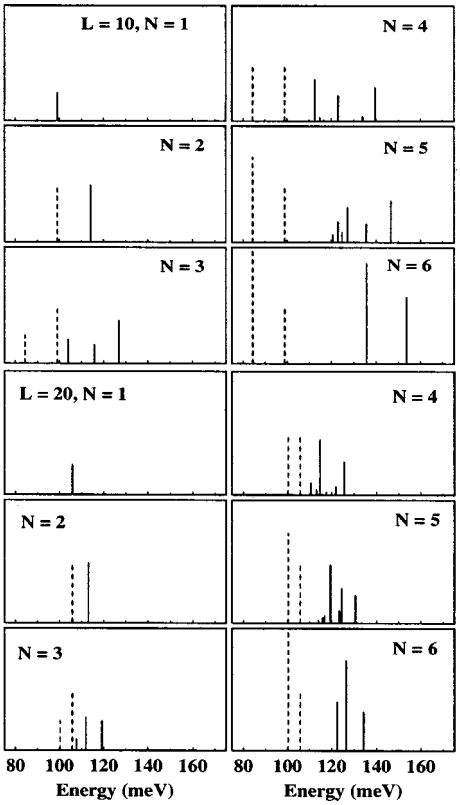

FIG. 3. Luminescence spectra of a quantum-dot cascade structure and *diagonal* optical transitions for various values of the dot size (L in nm) and number of electrons (N) in the dot. The dashed lines correspond to the luminescence of noninteracting electron systems.

For $L=10$ nm there is a small redistribution of intensities between the peaks while there is an additional line for $L=20$ nm. For two, three and four electrons in the dot, the interaction results in splitting of lines of the corresponding noninteracting systems. With increasing size of the quantum dots the lower energy lines become more intense.

As the Coulomb interaction between electrons in the second subband is about two times larger than the Coulomb interaction between electrons in the second and in the first subbands we have a highly nonsymmetric system and as a result there is the *large blueshift* in all cases, compared to in the noninteracting system. This blueshift decreases with increasing size of the quantum dots. The blueshift however increases with an increasing number of electrons. For $L=10$ nm, there is a blueshift of the emission line of about 55 meV when the electron number is increased from 1 to 6. These results open up the possibility of tuning the laser emission frequency for diagonal transitions by changing the number of electrons in quantum dots and/or decreasing the size of the dots.

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