Enhancement in excitonic absorption due to overlap in heavy-hole and light-hole excitons in GaAs/InAlGaAs quantum well structures

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In this letter we present experimental and theoretical results for excitonic transitions in coherently strained GaAs/InGaAlAs multiquantum well structures grown on a GaAs substrate. Absorption spectra of the structure with the substrate removed show an extremely sharp exciton peak with an absorption constant corresponding to nearly twice the value of that in the lattice-matched GaAs/AlGaAs system. Theoretical calculations suggest that the biaxial tensile strain in the well region, occurring after the substrate is removed, causes the heavy-hole and light-hole exciton states to coincide for a specific composition of the quaternary alloy. A comparison between the experiments and theory is made and the potential for devices based on this phenomenon is discussed.

Optical absorption studies of excitonic features in multiquantum well structures have become extremely important because of their applications in high-speed optical modulators and switches.[1-5] An important issue in these studies is the value of the excitonic absorption coefficient since modulator action depends upon the absorption strength. The peak height of the absorption coefficient (α_p) depends among other parameters upon the excitonic linewidth σ, since $\alpha_p \propto 1/\sigma$. Therefore, an obvious approach to increasing α_p is to have a small σ. This requires growth of high-quality heterostructures and low-temperature operation. As will be immediately evident, it is also possible to increase α_p by a careful incorporation of biaxial tensile strain in the well region of a quantum well. The approach is schematically described in Fig. 1. In bulk semiconductors, the heavy-hole (HH) and light-hole (LH) states are degenerate, as shown. In the presence of a biaxial tensile strain (applicable if the barrier is of a larger lattice constant than the well), the light-hole state (which has a light mass in the z direction and heavy mass in the x-y plane as shown) has a higher energy than the heavy-hole state.[6,7] If the material is now confined by a quantum well, a proper choice of the well size can cause the heavy-hole and light-hole bands to coincide at the zone center. If the strain and well size are such that

$$E_e + E_{hh} - E_{ex}^{hh} = E_e + E_{lh} - E_{ex}^{lh}, \quad (1)$$

one can have coincident light- and heavy-hole exciton resonances and a high absorption coefficient. In Eq. (1), E_e, E_{hh}, and E_{lh} are the subband levels for the electron, heavy hole, and light hole and E_{ex}^{hh} and E_{ex}^{lh} are the exciton binding energies for the heavy- and light-hole excitons.

In this letter we explore experimentally and theoretically the feasibility of obtaining coincidence between light- and heavy-hole excitons to enhance the exciton absorption. There are several potential candidates for quantum well structures which can provide biaxial tensile strain in the quantum well, the important GaAs-based systems being GaAs/$In_xAl_yGa_{1-x-y}As$ and $GaAs_{1-x}P_x$/$Al_yGa_{1-y}As$. The latter would be ideal since the lattice constant of GaAsP is smaller than that of the GaAs substrate. However, this structure cannot usually be grown by the conventional molecular beam epitaxy (MBE) technique other than using solid P sources and requires growth techniques using gaseous sources such as metalorganic chemical vapor deposition (MOCVD) or metalorganic MBE. In the former structure, the $In_xAl_yGa_{1-x-y}As$ barrier has a larger lattice constant than the substrate. Normally the well region may not be expected to be under strain, but if the substrate is removed, one can expect the strain to be shared between the well and the barrier. A candidate compatible with InP-based technology is $In_{0.53-x}Ga_{0.47+x}As/In_{0.52}Al_{0.48}As$ with x positive and of the order of 3–5%. However, in this combination the well is an alloy and alloy broadening severely limits the peak absorption constant in this system. Here we have chosen the binary well system, i.e., GaAs/$In_xAl_yGa_{1-x-y}As$ to study the absorption enhancement phenomenon.

In order to understand the absorption constant data of the strained system of interest, we note that the effect of the biaxial strain for (001) direction growth is to cause a splitting in the heavy hole ($|3/2,3/2\rangle$ state) and light hole ($|3/2,1/2\rangle$ state). If ϵ is the strain in the direction parallel to the quantum well interfaces, the effects on the heavy- and the light-hole band gaps are

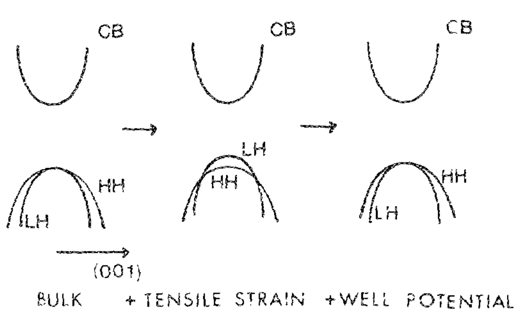

FIG. 1. Schematic of the energy-band structure of a quantum well showing the effects of biaxial tensile strain and quantum confinement.

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$$E_{g\frac{3}{2}} = E_g + \tfrac{1}{2}\delta E_{sh} - \delta E_{hy}\,, \tag{2}$$

$$E_{g\frac{1}{2}} = E_g - \tfrac{1}{2}\delta E_{sh} - \delta E_{hy}\,, \tag{3}$$

where

$$\delta E_{sh} = -2b\,[(C_{11} + 2C_{12})/C_{11}]\epsilon\,, \tag{4}$$

$$\delta E_{hy} = -2a[(C_{11} - C_{12})/C_{11}]\epsilon\,. \tag{5}$$

Here C_{11},C_{12} are the force constants for the well material, a and b are the deformation potentials, and ϵ indicates strain. For GaAs the shifts for the heavy- and light-hole gaps are -5.96ϵ eV and -12.4ϵ eV, respectively. In the presence of a biaxial tensile strain, the light hole is expected to be above the heavy-hole state.

In a quantum well, the particle states are confined and since the light-hole mass is lighter in the z direction (perpendicular to the interface), one can expect the LH and HH states to move closer to each other because of their difference in effective masses. The subband positions for the electron and hole states are determined by solving the one-dimensional Schrödinger equation.

$$-\frac{\hbar^2}{2m}\nabla^2\Psi(k,z) + V(z)\Psi(k,z) = E\Psi(k,z)\,, \tag{6}$$

where $V(z)$ represents the potential barrier seen by the electrons.

To calculate the exciton binding energy and absorption constant it is important to include the interaction between the light-hole and heavy-hole states especially for the system we are considering here, since the states are very close. This is done via the Kohn–Luttinger Hamiltonian with inclusion of strain.[8,9] The equation to be solved is

$$H\Psi = \begin{pmatrix} H_{\rm hh} + \frac{\delta}{2} & c & b & 0 \\ c^* & H_{\rm lh} - \frac{\delta}{2} & 0 & -b \\ b^* & 0 & H_{\rm lh} - \frac{\delta}{2} & c \\ 0 & -b^* & c^* & H_{\rm hh} + \frac{\delta}{2} \end{pmatrix} \begin{pmatrix} \Phi_{\frac{3}{2}\frac{3}{2}} \\ \Phi_{\frac{3}{2}\frac{1}{2}} \\ \Phi_{\frac{3}{2}-\frac{1}{2}} \\ \Phi_{\frac{3}{2}-\frac{3}{2}} \end{pmatrix}, \tag{7}$$

$$H_{\rm hh} = -\frac{\hbar^2}{2m_0}\left((k_x^2 + k_y^2)(\gamma_1 + \gamma_2) - (\gamma_1 - 2\gamma_2)\frac{\partial^2}{\partial^2 z}\right) + V_p(z)\,, \tag{8}$$

$$H_{\rm lh} = -\frac{\hbar^2}{2m_0}\left((k_x^2 + k_y^2)(\gamma_1 - \gamma_2) - (\gamma_1 + 2\gamma_2)\frac{\partial^2}{\partial^2 z}\right) + V_p(z)\,, \tag{9}$$

$$c = \frac{\sqrt{3}\hbar^2}{2m_0}\,[\gamma_2(k_x^2 - k_y^2) - 2i\gamma_3 k_x k_y]\,, \tag{10}$$

$$b = \frac{\sqrt{3}\hbar^2}{m_0}(k_y - ik_x)\gamma_3\frac{\partial}{\partial z}\,. \tag{11}$$

Here γ_1, γ_2, and γ_3 are Kohn–Luttinger parameters and δ is heavy-hole and light-hole state splitting in bulk material due to strain. The general hole solutions can be written as

$$\Psi_h^m(k,z) = \sum_\nu g_m^\nu(z)\,U_0^\nu(r)\,e^{ik_\parallel\rho}\,, \tag{12}$$

where $g_m^\nu(z)$ is the z-dependent function arising from the confinement of the potential, ν is the index representing the

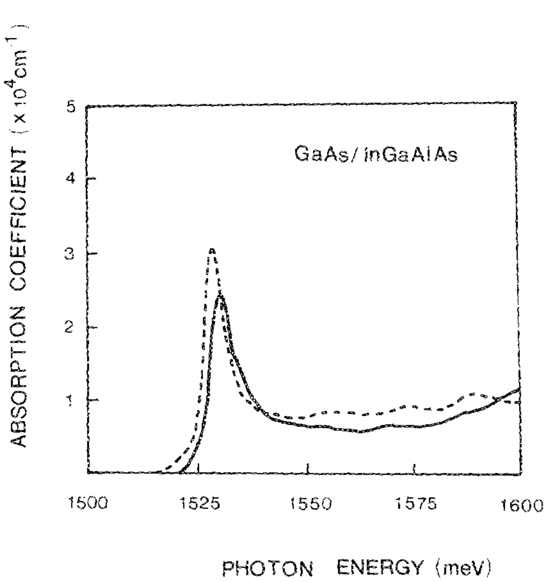

FIG. 2. Absorption spectra of GaAs/InGaAlAs strained MQW at low temperature. Solid line is used for experimental data; dotted line is for theoretical fit based on the formalism discussed in the text.

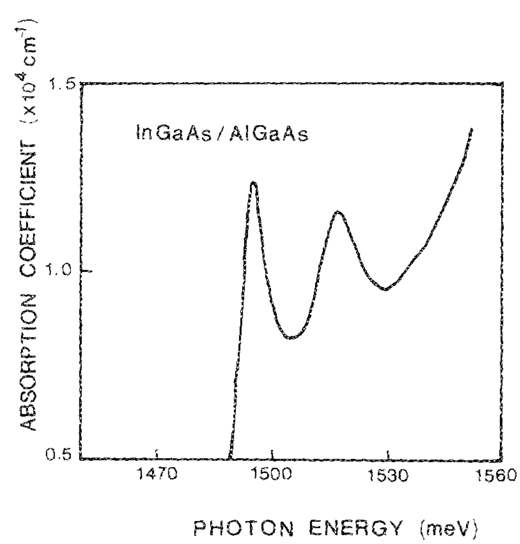

FIG. 3. Absorption spectra of a biaxially strained $In_{0.02}Ga_{0.98}As$/$Al_{0.3}Ga_{0.7}As$ MQW at low temperature.

total angular momentum of the state, m is index for each subband in the well, and $U_0^\nu(r)$ is the zone-center valence-band Bloch state for the ν spin component in the bulk material. In the absence of the off-diagonal mixing terms in the Kohn–Luttinger Hamiltonian (the so-called diagonal approximation), the hole problem is as simple to solve as the electron problem. However, in real semiconductors the off-diagonal mixing is quite strong and must be included for quantitative comparison with experiments. The off-diagonal terms are also responsible for the normally "forbidden" transitions to occur.

Once the subband levels are known, the exciton problem is solved variationally and the absorption constant is determined by[10]

$$\alpha_{nm} = \frac{4\pi^2 e^2 \hbar}{\eta m_0 c W} \frac{1}{\hbar\omega} \left| \sum_{k_\parallel} G_{nm}(k_\parallel) \hat{\epsilon} p_{nm}(k_\parallel) \right|^2 \delta(\hbar\omega - E_{nm}), \quad (13)$$

where η is the refractive index of the semiconductor, ω is the photon frequency, $\hat{\epsilon}$ is the polarization vector of the radiation, and m,n are the hole and electron subband indices. The $p_{nm}(k_\parallel)$ are optical matrix elements and the $G_{nm}(k_\parallel)$ are the Fourier components of the exciton envelope function and are determined by the solution of the exciton problem.[11] The Dirac-delta function has to be replaced by the broadening function since there is always a certain amount of linewidth in the exciton transition.

The experimental samples were grown in a Riber 2300 system. Growth was initiated on an n^+ GaAs substrate with a 0.2-μm GaAs buffer layer. Next a 0.5-μm undoped $Al_{0.3}Ga_{0.7}As$ region was grown primarily to act as an etch stop layer for substrate removal. Finally, a 58 period multiquantum well (MQW) consisting of 220 Å $In_{0.06}Ga_{0.57}Al_{0.37}$ barriers and 150 Å GaAs wells is grown. The growth temperature was maintained at 610 °C during the growth of the entire structure.

In Fig. 2 we show the 11 K absorption results for the MQW structure described above. This can be contrasted with results from a MQW structure in which the biaxial strain in the well region is compressive, as shown in Fig. 3. The structure of Fig. 3 has a 2.1-μm $In_xGa_{1-x}As$/$Al_{0.3}Ga_{0.7}As$ MQW region (with $x = 0.02$–0.03) with 150 Å wells and 220 Å barriers. Note that the sample with biaxial compression has a 20-meV splitting between the HH and LH exciton resonances. Theoretical calculations show that out of this 20 meV, 8 meV is due to quantum confinement and 12 meV to strain. However, in the sample with biaxial tensile strain in the wells the effect of the strain is such that it is cancelling the quantum confinement splitting and causing a merger of the HH and LH states. In Fig. 2 we have also shown the results for the absorption spectra calculated using the formalism discussed above. We have included a broadening for the excitonic transition to fit the experimental linewidth.

In summary, we have reported in this letter the demonstration of enhanced optical absorption using the tensile strain to merge the light-hole and heavy-hole transitions. This phenomenon, which can be achieved in a number of material systems, can be of significant importance in artificially enhancing the quantum efficiency of high-frequency photodiodes or in modulators with high on-off ratios.

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