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INTRINSIC RADIATIVE RECOMBINATION FROM QUANTUM STATES IN GaAs-$A\ell_xGa_{1-x}As$ MULTI-QUANTUM WELL STRUCTURES

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From absorption, emission, luminescence excitation and electron spin orientation studies of undoped GaAs-$A\ell_xGa_{1-x}As$ superlattices we demonstrate the intrinsic nature of the radiative recombination process. This is in direct contrast to recombination observed in similar purity thick GaAs material. Moreover, our results do not support a recent suggestion that enhanced LO phonon-electron coupling should occur in such superlattice structures.

Quantum effects associated with the confinement of carriers in ultrathin $A\ell_xGa_{1-x}As$-GaAs heterostructures and superlattices grown by molecular beam epitaxy (MBE) have been strikingly evidenced by interband absorption[1] and intersubband electronic Raman scattering[2] experiments. Even though lasing action under optical[3] and injection[4] pumping has been achieved and a short communication[5] concerning other luminescence properties has been published, a detailed understanding of the emission from such MBE grown structures has not yet emerged. More recently, laser related studies of equivalent structures (termed multi-quantum wells - MQW) grown by metallo-organic chemical vapor deposition (MO-CVD) have been interpreted[6] such that LO-phonon assisted radiative recombination is considered to provide a major radiative recombination mechanism. Moreover, it has been suggested[7] that the electron-phonon coupling constant is quite enhanced in such confined carrier MQW structures over that found in bulk GaAs. This implies that all MQW structures (MBE grown as well as MO-CVD) should exhibit enhanced electron-phonon coupling. The purpose of this work is to demonstrate that (i) in high-quality samples (i.e., low impurity content and good interface properties) intrinsic free-exciton recombination dominates the emission spectrum even at very low temperatures (in surprising contrast to similar thick GaAs where bound exciton and impurity processes dominate), (ii) the electron-phonon coupling does not appear significantly altered from that found in thick GaAs.

Experiments were performed over a wide range of temperatures (1.8 K - 100 K) with no qualitative changes of the luminescence spectrum, using a cw tunable dye laser or a tunable optical parametric oscillator for optical excitation. Absorption, photoluminescence and excitation spectra are shown at 1.8 K in Fig. 1 for the sample which presents some of the sharpest features among the ∿ 100 samples grown and examined in this laboratory.[8] This sample has 25 GaAs wells 188 Å thick, each separated from the next by a 19-Å $A\ell_{0.3}Ga_{0.7}As$ barrier.[9] The observation of sharp peaks in the absorption and excitation spectra is typical of recent high-quality material grown in this laboratory. It also indicates that free exciton effects dominate the absorption phenomena, as band-to-band transitions would lead to a step-like absorption lineshape.[1] Further evidence for this excitonic behavior comes from absorption experiments in high magnetic fields[10] where these features exhibit the expected non-linear diamagnetic contributions to the transition energy.

The observation of two luminescence lines (1.525 and 1.530 eV) at the same energies as the fundamental absorption peaks indicates that they have the same physical origin, i.e. that the photoluminescence is due to free-exciton recombination process.[11] Unfortunately, because of the complexity of the free exciton recombination process,[12] the lineshape cannot be analysed in a simple manner to support this assertion. If the k-conservation rule were strictly valid, the luminescence line due to free exciton recombination would be infinitely sharp and weak. However, a number of higher-order processes relax the k-selection rule, among which are impurity, electron-exciton and exciton-exciton scattering, and acoustic-phonon-assisted recombination. These processes lead to the observation of broader lines, the detailed shape of which depends in a non-trivial manner on impurity content, exciting light intensity and lattice temperature.[12] However, on increasing the lattice temperature or excitation intensity we observed a decrease of the high-energy slope of the lines and an increase of the intensity ratio of the e-ℓh to the e-hh

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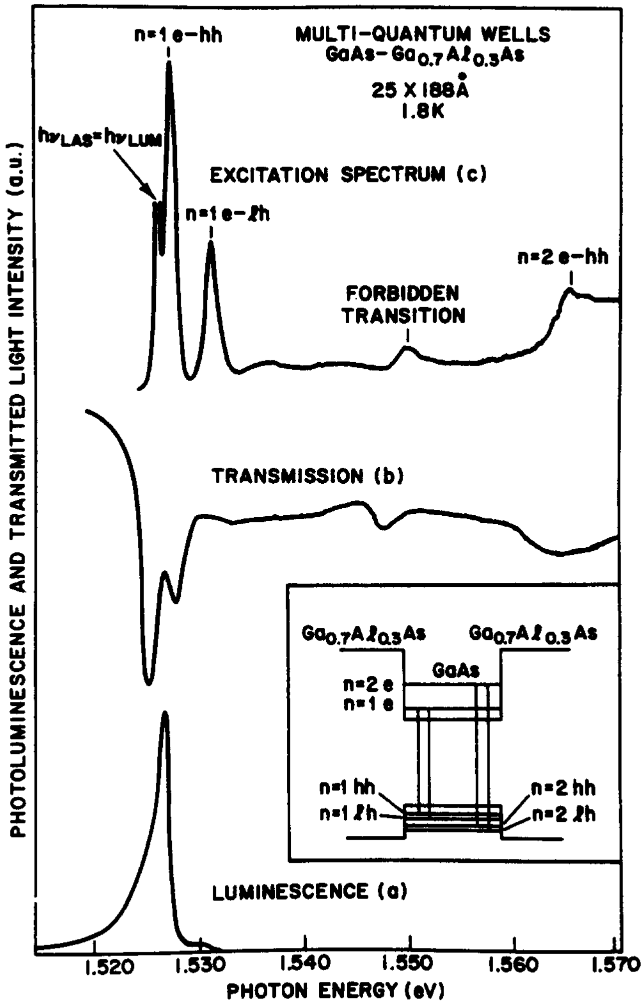

Fig. 1 Photoluminescence (a), absorption (b) and excitation spectra (c) of a MQW sample. All spectra were recorded at 1.8 K. (a) Photoluminescence under 1.62 eV excitation (power density $\sim$ 5 W/cm^2). (b) Transmission spectrum under white light excitation (c) Excitation spectrum of the luminescence at 1.525 eV: The luminescence-analysing spectrometer is set at 1.525 eV while the exciting light wavelength from a cw dye laser is scanned. Peaks occur at higher-lying exciton energies as a result of increased absorption and/or efficient relaxation to the luminescent level. The insert displays the first allowed transitions for a MQW structure (not drawn at scale). The slight shift ($\sim$ 1.5 meV) between the transmission spectrum and both the luminescence and excitation spectra is due to the strain present in the sample used for transmission, which had the GaAs substrate removed. The "forbidden" transition (parity allowed, $\Delta n = 2$) is observed in all samples which show sharp allowed transitions.

lines shown in Fig. 1, which is consistent with a luminescence originating from freely moving excitations of the crystal.

Direct verification of the assignment of the two peaks to free-exciton energy levels associated with the heavy and light hole bands can be had from measurements of transition polarizations. This can be achieved by optical spin orientation measurements[5,13] or by measuring the linear polarization of emission emanating from a cleaved edge of the superlattice structures. Here we will discuss only the more detailed spin orientation results, although both experiments give equivalent results. The luminescence and circular polarization spectra at 50 K for excitation at 1.647 eV with circularly polarized light ($\sim$ 5 W/cm^2) appear in Fig. 2a. The various transitions in absorption and luminescence under circular polarized light are shown in

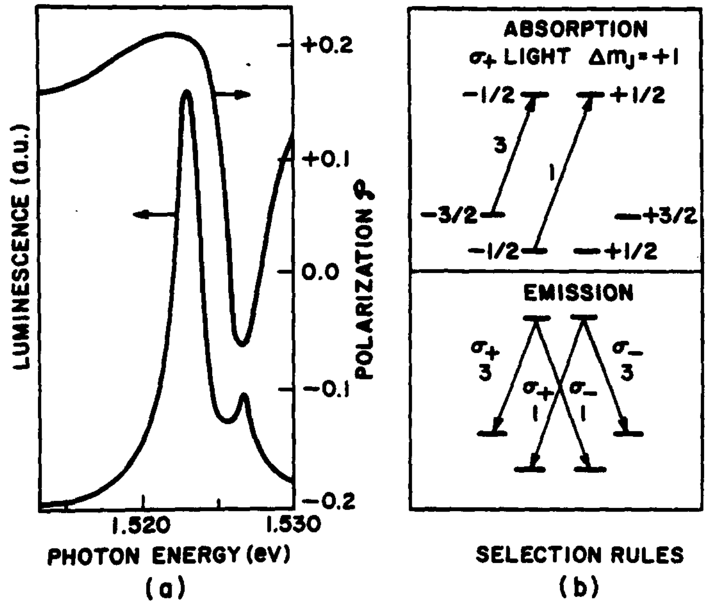

Fig. 2 (a) Photoluminescence intensity and its circular polarization at 50 K for excitation at 1.647 eV with circularly polarized light. (b) Absorption and emission transitions upon excitation with circularly polarized light (relative oscillator strengths are indicated). The axis of quantization is along the direction of propagation of light (perpendicular to the GaAs-$Ga_{0.7}A\ell_{0.3}As$ layers). Under nonresonant conditions (such as in Fig. 2a), electrons with m_j = -1/2 states are predominantly created. Assuming a completely spin relaxed hole population,[14] this leads to a positive polarization (σ+) of the heavy-hole recombination light and a negative polarization (σ-) of the light-hole recombination light, as observed in Fig. 2a.

Fig. 2b. The main luminescence peak has a circular polarization $\rho \gtrsim +0.2$ (signs are relative to the pump) and can be associated with the n = 1 heavy-hole band with m_j predominantly -3/2, while the smaller peak with an opposite polarization involves predominantly light-holes with m_j = +1/2. This level scheme is fully corroborated by resonant pumping experiments. Resonant excitation of the e-hh transition excites only m_j = -1/2 electrons and gives a negative polarization ρ at the e-ℓh line while resonant excitation of the e-ℓh transition excites m_j = +1/2 electrons and gives a negative polarization at the e-hh emission. These polarization characteristics, provide the first definite evidence for the heavy and light-hole level assignments which were heretofore mainly supported by the good agreement between the measured absorption spectra and calculated energy level schemes. They also support the free-exciton nature of the luminescence, as bound exciton levels would show different, symmetry determined spin orientation properties.

The most striking difference between the luminescence of this MQW material and that of thick GaAs is the weakness of transitions due to bound excitations; bound excitons (BE), electron-to-neutral acceptor (e-A°), hole-to-neutral donor (D°-h), donor-acceptor pairs (D°-A°) - which usually dominate the free-exciton luminescence, especially in thick material of comparable purity.[15] Only a small structureless luminescence (∿ 2-3 orders of magnitude smaller than the peak free-exciton luminescence of our samples) is observed in the corresponding spectral range (hν > 1.45 eV). However, in structures with thicker layers L_z > 1000 Å, some of the usual luminescence lines of thick GaAs can be observed. This indicates that confinement of the wavefunction down to ∿ 200 Å in MQW plays a major role in the luminescence process. A first effect of confinement is to spread the impurity-binding energy when the impurity-interface distance becomes of the order of or smaller than the impurity Bohr radius[16] (in GaAs, a_B ≈ 105 Å for donors and 25 Å for acceptors). This leads to a smearing of the luminescence bands. A second effect comes from the symmetry of the impurity ground-state wavefunction, which gradually changes from a 1s hydrogenic symmetry in the bulk to a 2p symmetry at the interface. Depending on the exact position of impurities, the oscillator strength of the transition is therefore decreased compared to that of bulk materials. These two effects can account for the nonobservation of strong e-A° and D°-A° bands. The same explanation should be valid for bound exciton emission, with the additional possibility that excitons bound to neutral impurities, which usually dominate in thick GaAs, might not even exist in MQW because the confinement should increase electron-electron interaction, thereby diminishing the binding energy.

Finally, in no case has emission corresponding to enhanced LO phonon coupling been seen in spontaneous or stimulated[3,4] emission from these high purity, MBE-grown MQW structures. The companion excitation and absorption studies reported here leave no doubt as to the relative energy of PL and absorption and we must conclude, along with recent studies[17] of the magneto phonon effect in similar MBE layers, that enhanced electron-LO phonon coupling due to 2D confinement effects is not a fundamental property of MQW structures. In contrast, the intrinsic nature of the dominant (free-exciton) emission and the absence of strong impurity induced features present an intriguingly different situation from that found in similar quality thick GaAs as well as in other direct gap III-V semiconductors.

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