OBSERVATIONS OF FORBIDDEN EXCITONIC TRANSITIONS IN GaAs/Al$_x$Ga$_{1-x}$As QUANTUM WELLS IN AN ELECTRIC FIELD

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Abstract

Photocurrent spectroscopy has been used to study the unallowed exciton associated with the n=1 conduction subband and the n=2 heavy hole valence subband (h_{12}) in a quantum well in an electric field. For well widths greater than 120 Å two excitons are observed in the energy range where only h_{12} should be seen. Based on the electric field, polarization and uniaxial stress dependence of the spectra we conclude that the extra peak arises from mixing between the first light hole and second heavy hole valence subbands.

The excitonic transitions with the largest oscillator strengths in square GaAs/Al$_x$Ga$_{1-x}$As quantum wells are those between conduction and valence subbands which have envelope functions with the same quantum number ($\Delta n=0$). When an electric field is applied perpendicular to the quantum wells, the symmetry is broken and optical matrix elements for "forbidden" transitions ($\Delta n\neq0$) increase.[1] In particular the h_{12} (n=1 conduction subband and n=2 heavy hole valence subband) exciton should become visible in absorption and related optical measurements. The energy of this exciton will be slightly greater than or less than that of the first light hole exciton (l_{11}) depending on the well width. We have made photocurrent measurements on quantum wells with widths between 80 and 160 Å. In samples with 80 Å wide wells we observe an electric field enhancement of the h_{12} exciton, but, for samples with well widths larger than approximately 120 Å, two peaks become visible in the energy range where only the h_{12} exciton is predicted to occur. Polarization dependent measurements show that the oscillator strength of the additional peak arises from the heavy hole valence band, while under uniaxial stress the energy of this peak shifts as if it is associated with the light hole valence band. We conclude that the ex-

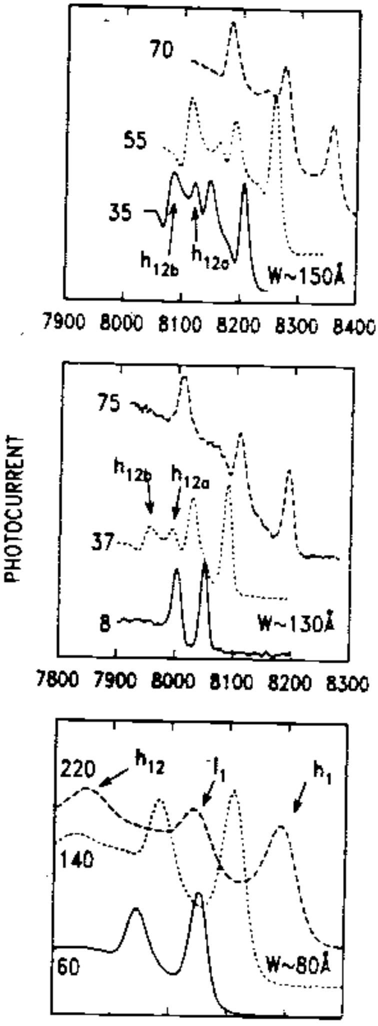

Fig. 1: Electric field dependent photocurrent spectra showing an extra exciton peak in the energy range of h_{12} in wider quantum well (130 and 150 Å) samples. The approximate field is given to the left of each spectrum in kV/cm. Some spectra at lower fields have been multiplied by a constant to allow the spectra to be seen on the same scale. Measurements were made at 10K.

tra peak arises from a mixing between the first light hole and second heavy hole subbands, and speculate that it is due to mixing between an excited state of the l_1 exciton and the ground state of the h_{12} exciton.

The samples used in this study were GaAs/$Al_xGa_{1-x}As$ p-i-n and Schottky barrier photodiodes with two to ten quantum wells imbedded in the depletion region of the diodes. They were grown by molecular beam epitaxy. The top p^+ (p-i-n) or metal (Schottky) layer were sufficiently thin to allow light to be transmitted to the quantum wells. The widths of the $Al_xGa_{1-x}As$ intrinsic regions were between 0.2 and 1.5 μm.

Figure 1 shows photocurrent spectra for 80, 130 and 150 Å wide quantum wells as a function of electric field. The light is polarized perpendicular to the [100] growth axis. In the 80 Å sample, the h_{12} exciton becomes visible as the field is increased. It has been identified by comparing its energy as a function of field to the results of envelope function calculations. In the 130 and 150 Å wells, two peaks become visible with increasing electric field (h_{12a} and h_{12b}). In the 150 Å sample for fields of 35 and 55 kV/cm, steps are also visible in the spectrum at energies slightly greater than the h_1 (lowest energy heavy hole exciton) and h_{12b} excitons. Such steps have previously been observed and were identified as a superposition of the excited states of an exciton and the continuum edge of the subband to subband transition associated with the exciton.[2] It should be noted that, as

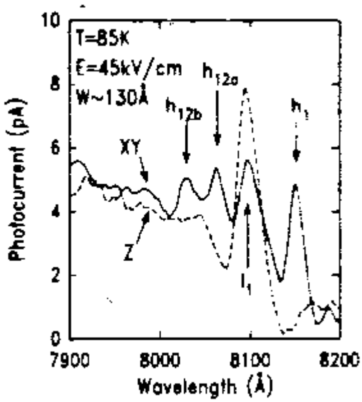

Fig. 2: Polarization dependent photocurrent spectra for 130 Å wide quantum wells.

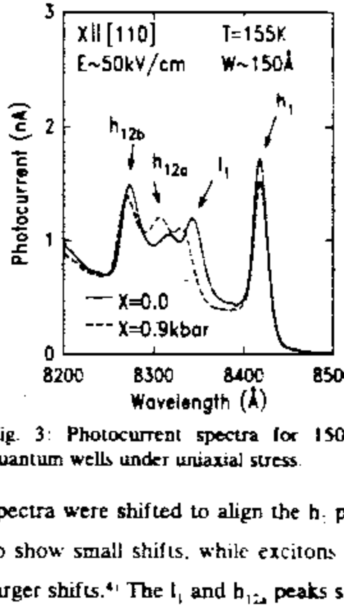

Fig. 3: Photocurrent spectra for 150 Å quantum wells under uniaxial stress.

the field increases, the h_{12a} exciton amplitude decreases until it becomes a step on the high energy side of l_1 and only h_1, l_1, and h_{12b} are seen.

The dependence of the photocurrent spectra on the polarization of the incident light and on uniaxial stress were also investigated. Figure 2 gives spectra for a sample with 130 Å quantum wells imbedded in an $Al_{0.3}Ga_{0.7}As$ waveguide.[3] The sample was illuminated from a cleaved [110] edge with light polarized parallel (Z) or perpendicular (XY) to the [100] growth axis. In the XY polarization h_1, l_1, h_{12a}, and h_{12b} are observed as in Fig. 1., but in the Z polarization only l_1 and the continuum step associated with l_1 are seen. Since Z polarized light should selectively excite optical transitions associated with the light hole valence band,[3] the oscillator strengths of h_{12a} and h_{12b} are predominantly heavy hole in character. Figure 3 gives the dependence of the exciton energies upon uniaxial stress (X) for 150 Å wide quantum wells. The stress was applied along a [110] crystal axis. Since only a small change in the energy of the h_1 exciton was observed, the spectra were shifted to align the h_1 peak. We, therefore, expect the heavy hole excitons to show small shifts, while excitons associated with the light hole valence band exhibit larger shifts.[4] The l_1 and h_{12a} peaks show a shift of approximately 2.0 meV, while the energy of h_{12b} shifts 0.6 meV. From this we conclude that the energy of h_{12a} is predominantly determined by the light hole valence band.

The above results, which show that h_{12a} has heavy hole oscillator strength, but is energetically related to the light hole valence band (for the electric field of Fig. 3) strongly suggest that the peak arises from mixing between the first light hole and second heavy hole subbands. This would explain the absence of extra peaks in the 80 Å wells of Fig. 1, since, as the well width is decreased, the subbands separate and the mixing is reduced. Also, as the electric field is increased, the first heavy and light hole subbands exhibit a larger Stark shift than the second heavy hole subband[1], and the mixing should again be reduced. This explains the decrease in h_{12a} as electric field is increased in Fig 1 and also serves to identify h_{12b} as the true h_{12} exciton in the limit of large field. Since h_{12a} appears to merge with the continuum of the l_1 exciton, we speculate that it arises from a mixing between the ground state of the h_{12} exciton and an excited state of l_1. That such a mixing may occur has been predicted by Chan[5]. Another possible model for h_{12b} is that it is associated with points of low dispersion away from the center of the Brillouin zone in the first light hole or second heavy hole subbands as predicted in calculations of superlattice band structure which include the effects of valence band mixing.[6]

The observation of this mixed state exciton is significant for a number of reasons. The energies of the excitonic levels in quantum wells can generally be fit quite accurately with simple envelope function calculations which neglect valence band mixing effects. The strongest evidence for mixing has been the observation in optical measurements on symmetric quantum wells of peaks associated with forbidden transitions.[7] In the present study, we observe a peak which cannot be identified energetically using the simple envelope function model, thereby providing a better opportunity to test the various methods of calculating the electronic properties of quantum wells. In addition, a number of important physical parameters in heterojunction systems, such as band offsets, have been inferred from the energies of forbidden excitonic transitions.[8] The present study shows that care must be taken in interpreting the energies and identifying the transitions.

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