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SPECTROSCOPIC STUDIES OF EXCITONIC FINE STRUCTURE UNDER ELECTRIC FIELDS

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The application of an external electric field to GaAs/$Ga_{1-x}Al_xAs$ quantum wells has allowed us to resolve the 2p excited state of the heavy-hole exciton. A sharing of the oscillator strength between this excited state and the ground state of the light-hole exciton, together with an anticrossing behavior, has been observed in low-temperature photoluminescence excitation spectra. A rare structure for well thickness $\gtrsim 120$ Å, in the energy range of the forbidden exciton associated with the first conduction subband and the second heavy-hole subband, has been attributed to valence-band mixing between the first light-hole and the second heavy-hole subbands.

1. Introduction

The use of high-resolution spectroscopic techniques has led to a deeper understanding of many different physical phenomena, and to a refinement of the models which describe them [1]. The optical spectra of GaAs/$Ga_{1-x}Al_xAs$ quantum wells (QWs) are characterized by sharp excitonic structure [2]. The first observation of excited states in these spectra was reported by Miller et al. [3]. A direct measurement of the heavy- and light-hole excitonic term value $B_{1s} - B_{2s}$, where $B_{1s(2s)}$ represents the two-dimensional exciton 1s (2s) binding energy, was obtained and a determination of the binding energies was performed [3]. Magneto-optical studies offer an alternative way to obtain this information from the difference in the magnetic field dependence of the ground and excited states of the excitons [4]. More recently, Dawson et al. [5] have observed, as a consequence of the improvement in sample quality, better-resolved excited-state peaks in photoluminescence excitation (PLE) spectra.

Here, we review some of the effects of an electric field on the excitonic fine structure of the optical spectra of GaAs/$Ga_{1-x}Al_xAs$ QWs, measured by PLE and photocurrent (PC) spectroscopies. In the first part of the paper coupling between the ground state of the light-hole exciton (ℓ_1) and the excited states ($h_1^{(2x)}$, where x

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stands for excited) of the heavy-hole exciton (h_1) will be discussed. As a result of this interaction h_1^{2p} is resolved for the first time. In the second part of the paper we shall deal with forbidden excitonic transitions, namely the h_{12} exciton (associated with the first conduction- and the second heavy-hole valence-subband), and their dependence on the applied electric field.

2. Experimental details

The samples used in our study have been grown by molecular beam epitaxy on (100)-oriented n^+-GaAs substrates [6,7]. One or several GaAs QWs were grown on a n^+-GaAs buffer, followed by a $Ga_{0.65}Al_{0.35}As$ layer. The wells, separated by ~250 Å thick barriers, were capped by 800 Å $Ga_{0.65}Al_{0.35}As$ and 1.5 μm p^+-GaAs to result in a p^+–i–n^+ configuration. The magnitude of the electric field, perpendicular to the layers, was estimated from growth parameters and from the bias corresponding to flat band condition, and it is believed to be accurate within 10% [8]. Flat band corresponds to a certain positive bias (~1.7–1.75 V) and a decrease of the external voltage implies an increase in the electric field.

Excitation spectra were recorded at 4.8 K with a resolution of 0.2 meV. The samples were excited with an LD700 dye, pumped by a Kr^+ laser, with power densities below 0.5 W/cm^2, while a double spectrometer was set at the heavy-hole emission wavelength, which increases with increasing field. Photocurrent measurements were performed illuminating the sample with the light of a tungsten lamp dispersed from a grating spectrometer. The light-induced PC was detected using standard lock-in techniques [9]. A specially designed sample, where the thickness of the alloy layers and the number of QWs were chosen to form a leaky waveguide structure [10], was used for polarization-dependent PC measurements [6].

3. Fine structure of the heavy-hole exciton

Fig. 1 shows a low-field PLE spectrum of a sample with 160 Å thick QWs. Also shown in the figure is the heavy-hole photoluminescence (PL). The exceptional quality of the sample is reflected in the sharpness of the peaks in the spectrum, which have a full width at half maximum of 0.5 meV. Based on the agreement with previously published experimental [3,5] and theoretical [3,5,11,12] data, and with recent PLE experiments in the presence of a magnetic field [13], we assign the shaded structures to the excited states of the heavy- and light-hole excitons, $h_1^{(2x)}$ and $\ell_1^{(2x)}$, respectively. Calculations of the oscillator strength give a ratio of the excited to the ground states of 0.11 [12,14] and 0.078 [15], in excellent agreement with the experimental value of 0.07 ± 0.01. The peak labelled h_{12a} is a forbidden exciton associated with the $n=2$ heavy-hole and $n=1$ conduction subbands [6]

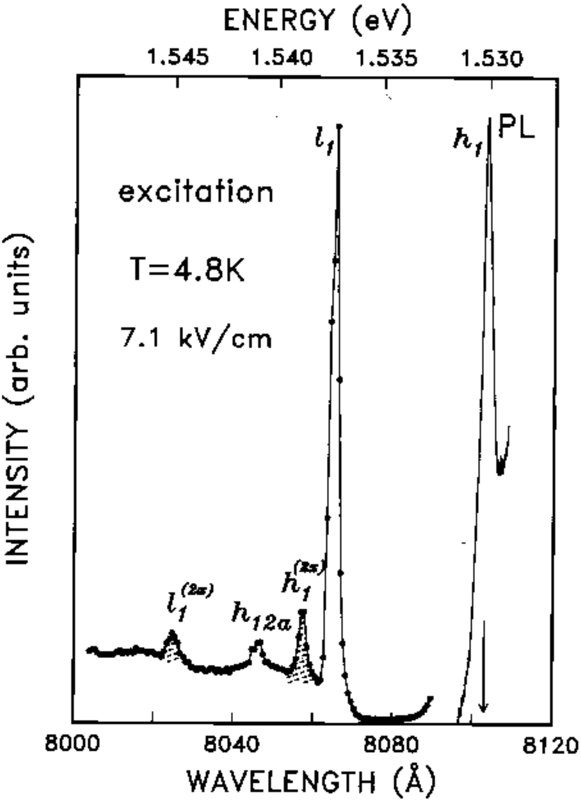

Fig. 1. Low-field excitation spectra of a p^+–i–n^+ structure with 160 Å wide wells. The shaded structures $h_1^{(2x)}$ and $\ell_1^{(2x)}$, correspond to excited states of the heavy- and light-hole excitons, respectively. The photoluminescence (PL) spectrum of the heavy-hole exciton (h_1) is also shown. The arrow indicates the detection energy for the PLE spectrum.

and will be discussed later. The small electric field has already shifted the structures by ~0.1 meV and it is responsible for the appearance of h_{12a}.

We shall now concentrate on the effects of the electric field on the ℓ_1 and the $h_1^{(2x)}$ excitons. PLE spectra for different electric fields are shown in fig. 2. At flat band condition, $h_1^{(2x)}$ is 1.5 meV higher than ℓ_1 and the ratio of their oscillator strengths is ~1/4. With increasing field, $h_1^{(2x)}$ shifts more than ℓ_1, because of its heavy-hole character; their relative energy separation decreases, and their intensities become comparable.

Fig. 3 shows a more detailed spectrum of $h_1^{(2x)}$. As a consequence of the $h_1^{(2x)}$–ℓ_1 interaction, fine structure is resolved in the excited states of the heavy-hole exciton. A shoulder, which is already hinted at zero field at the low energy-side of the structure, becomes comparable with the original peak at 5.7 kV/cm, and

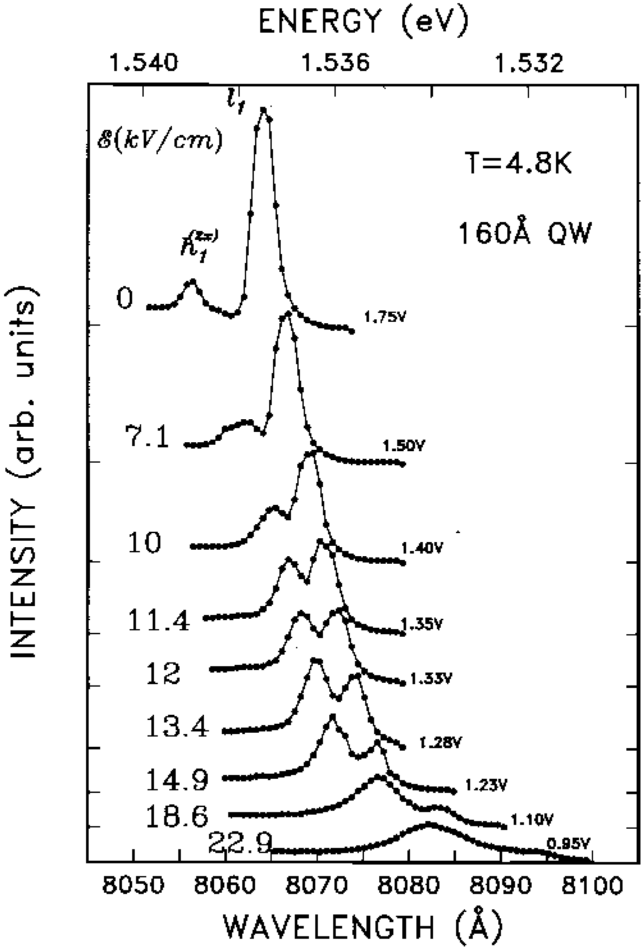

Fig. 2. Excitation spectra, recorded at the h_1 exciton, for several electric fields in the spectral range of the light-hole exciton (ℓ_1) and the excited states of the heavy-hole exciton ($h_1^{(2s)}$).

it is the dominant feature at higher fields. We have attributed the high- (low-)energy component of the doublet to the 2s (2p) state of the h_1 exciton [7]. This assignment, which was based on the agreement of the experimental splitting between the $h_1^{(2s)}$ and $h_1^{(2p)}$ states (~0.45 meV) and the results of calculations of their binding energies [11], has been confirmed in recent calculations by Bauer and Ando [15]. These calculations also show that the interaction between the ℓ_1 exciton and $h_1^{(3d)}$ becomes important at higher fields. Although we are not able to resolve $h_1^{(3d)}$, a shoulder in the low-energy side of ℓ_1 (see, for example, the spec-

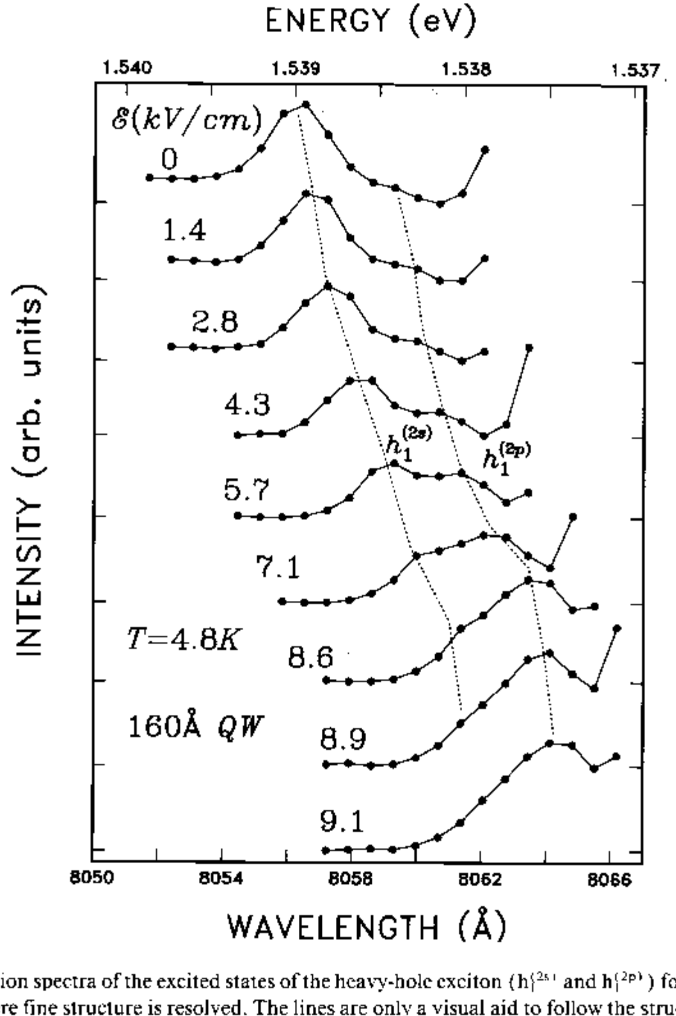

Fig. 3. Excitation spectra of the excited states of the heavy-hole exciton ($h_1^{(2s)}$ and $h_1^{(2p)}$) for different fields where fine structure is resolved. The lines are only a visual aid to follow the structures.

trum at 14.9 kV/cm in fig. 2) and its larger broadening with increasing field, compared to that of $h_1^{(2x)}$, could be an indication of the presence of $h_1^{(3d)}$ in the spectra.

Fig. 4, where the energies of $h_1^{(2x)}$ and ℓ_1 are plotted as a function of field, shows clearly the coupling between these excitons, that manifest itself as an anticrossing. The dashed and solid lines represent calculations of the Stark shifts of uncoupled heavy- and light-hole excitons, respectively [7,8]. As seen in the figure the excitons follow closely the theory outside of the range of strong coupling. The calculation of ref. [15], where hole–band hybridization and 1s, 2s, 2p and 3d excitons are included, reproduce also the data in the whole field region. The inset in the figure depicts the integrated intensity of $h_1^{(2x)}$, normalized to that of ℓ_1, as a function of

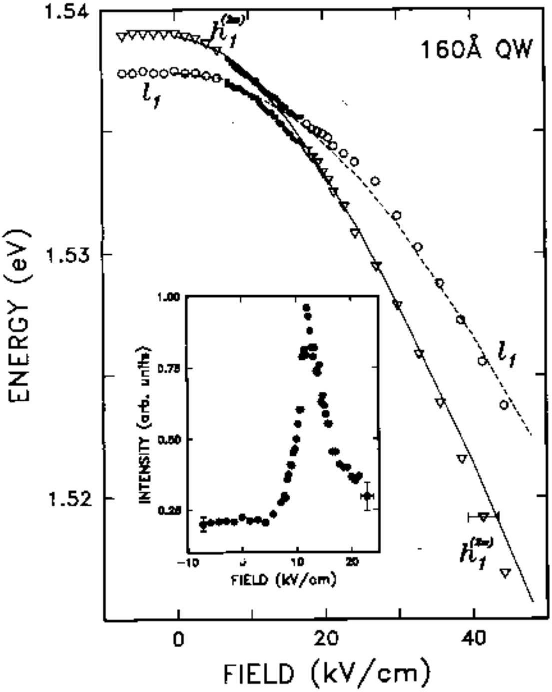

Fig. 4. Stark shifts of the light-hole exciton (open circles) and excited states of the heavy-hole exciton (open triangles). In the region of strong coupling the data are shown as solid dots. The lines correspond to calculations in the envelope-function approximation, neglecting changes in binding energy and coupling between excitons. The inset depicts the integrated intensity of $h_1^{(2s)}$, normalized to that of ℓ_1, as a function of field.

field. A pronounced peak is observed at fields where their relative energy separation is minimum. This separation, which amounts to 0.7 ± 0.2 meV, indicates an enhancement in the coupling between these excitons of ~2 with respect to bulk GaAs [16].

4. Forbidden excitonic transitions

Fig. 5 shows several PLE spectra, from zero field up to 14 kV/cm, including the region of the first forbidden ($\Delta n \neq 0$) exciton, h_{12}. The observation of h_{12} in the optical spectra of GaAs/$Ga_{1-x}Al_xAs$ quantum wells has been attributed to valence-band mixing effects [17]. However, the presence of a *small* electric field causes a lifting of the inversion symmetry in the QWs and this transition becomes allowed (see the disappearance of h_{12} in the figure as the field is reduced). Therefore, this mechanism needs to be considered in interpreting the presence of h_{12} in the spectra.

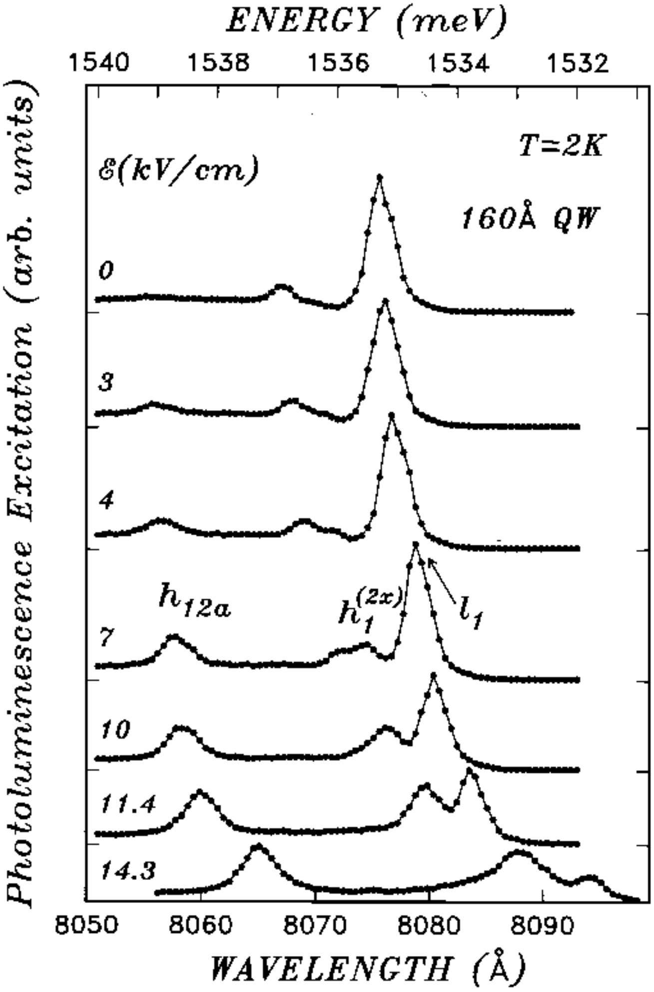

Fig. 5. Excitation spectra for several electric fields showing the disappearance of the forbidden ($\Delta n \neq 0$) exciton, h_{12a}, as the electric field is reduced. Notice that even at very small, but finite, electric fields this structure is present in the spectra.

When the energy range of the spectra is increased towards the high-energy side, a striking effect is observed. Fig. 6 shows PLE (solid lines) and PC (dashed lines) spectra for several electric fields (note the good agreement between the two techniques in the spectra at 25 kV/cm). According to envelope function calculations [18], the exciton corresponding to the $n=1$ conduction subband and $n=2$ heavy-hole subband (h_{12}) and only this exciton, should appear in the energy range where the two excitons, labelled h_{12a} and h_{12b}, are observed. For fields larger than ~50

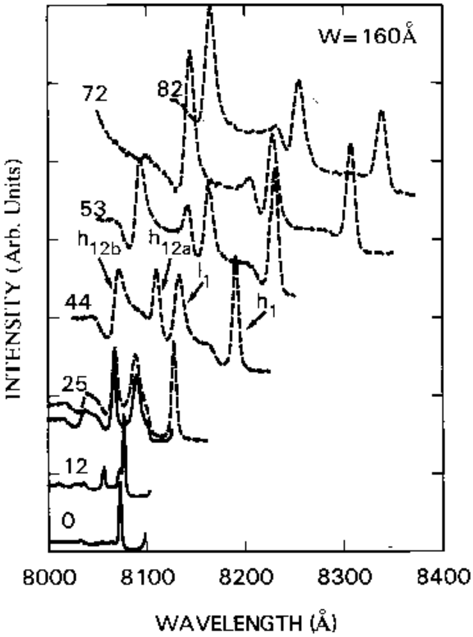

Fig. 6. Photocurrent (dashed) and PLE (solid) spectra as a function of electric field. The heavy-hole exciton (h_1) is not shown in the PLE spectra. h_{12a} and h_{12b} are excitons arising from mixing between the $n=1$ light-hole and the $n=2$ heavy-hole subbands.

kV/cm, h_{12b} dominates h_{12a}, and its Stark shift agrees with the results of the calculations for h_{12} [19], indicating that this is the "true" h_{12} exciton. The fact that the excited states of the light-hole exciton, $\ell_1^{(2x)}$, run in between the two transitions [19], suggests a possible contribution of light-hole mixing to the observation of the extra peak. Furthermore, this would explain the absence of extra peaks for well widths narrower than ~120 Å, since, as the well width is decreased, the subbands separate, and the mixing is reduced.

Polarization and uniaxial-stress dependent PC measurements have helped to establish the heavy-light mixed character of h_{12a} and h_{12b} [6]. For fields where the two excitons are comparable, the polarization dependent results show that both structures have *oscillator strengths* arising primarily from the second *heavy-hole* subband, while the uniaxial-stress measurements establish the light-hole character of h_{12a} [6]. The apparently contradictory conclusions obtained from the two measurements can be understood by realizing that the uniaxial-stress measurements provide information of the entire wavefunction, while the polarization dependent measurements involve only the part of the wavefunction which contributes to the absorption. Based on the predicted mixing, for well widths in the range of our studies, between the $n=1$ light-hole and the $n=2$ heavy-hole subbands [20] and on recent results by Chan [21], we have attributed the presence of the two peaks to an interaction between $\ell_1^{(2x)}$ and the ground state of h_{12}.

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