Observation of the excited level of excitons in GaAs quantum wells

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The light- and heavy-hole two-dimensional exciton term values are determined directly from the excitation spectra of GaAs-$Al_xGa_{1-x}As$ heterostructures with GaAs well widths L from 42 to 145 Å. These values are in excellent agreement with a theoretical model which contains no adjustable parameters. This theory also gives integrated strengths for the excitons which agree with experiment, and ground-state binding energies as a function of L.

Early work[1] on the absorption spectra of GaAs-$Al_xGa_{1-x}As$ heterostructures grown by molecular-beam epitaxy (MBE) noted the excitonic nature of the absorption peaks, and an analysis of the spectra led to an estimate of $B_{1S} \sim 9$ meV for the two-dimensional (2D) exciton $1S$ ground-state binding energy for GaAs wells of width $L < 100$ Å. A more recent paper[2] emphasizes the role of the intrinsic free 2D exciton in the recombination radiation from high-quality quantum-well structures. Another recent publication[3] reports considerably larger estimates, 20 and 13 meV for heavy- and light-hole excitons, respectively, for $L \leq 120$ Å. In the present work excitation spectra of the photoluminescence for both *single* and multiple quantum wells with $42 \leq L \leq 145$ Å have yielded for the first time a direct measurement of the heavy- and light-hole 2D exciton term value $B_{1S} - B_{2S}$. The results confirm the validity of a theoretical model containing no adjustable parameters, and the theory is then used to obtain B_{1S} as a function of L.

The multi-quantum-well samples studied were grown on (001) planes by MBE under optimum conditions which resulted in sharp lines in both the emission and excitation spectra, as reported earlier.[4] In addition, single-quantum-well samples also grown by MBE, but in a different apparatus than those mentioned above, were also studied. We have been able to study single-quantum wells optically without having to resort to high excitation levels for L as small as 31 Å. Excitation was with a tunable cw dye laser, and the photoluminescence was detected 24° off normal incidence in the reflection direction. Circular-polarization techniques were also used to assist in identifying the origin of the luminescence.[5]

Figure 1 shows the excitation spectrum at 6 K for a single GaAs well with $L = 42$ Å and clad on both sides with 0.65 μm of $Al_{0.37}Ga_{0.63}As$. The detection system was set on the long-wavelength side of the peak of the photoluminescent intensity $I_{PL}(\lambda)$ and then the excitation photon energy E scanned at ~ 1.1 W/cm^2. The two large peaks correspond to $n = 1$ heavy- and light-hole excitons, denoted E_{1H} and E_{1L}, respectively. Of the most interest are the shoulders on the high-energy side of each exciton peak. Electron-spin-polarization measurements[5] verify the above assignments and also show that the shoulder at the lower energy is heavy hole in character and the shoulder at the higher energy exhibits approximately the same polarization, i.e., the same mix of light and heavy holes, as the region beyond at still higher energies. These properties lead us to conclude that the shoulders represent the onset of the heavy- and light-hole excited states and that the discrete excited states are not resolved from the continuum. It is also important to note in Fig. 1 that the level of photo-

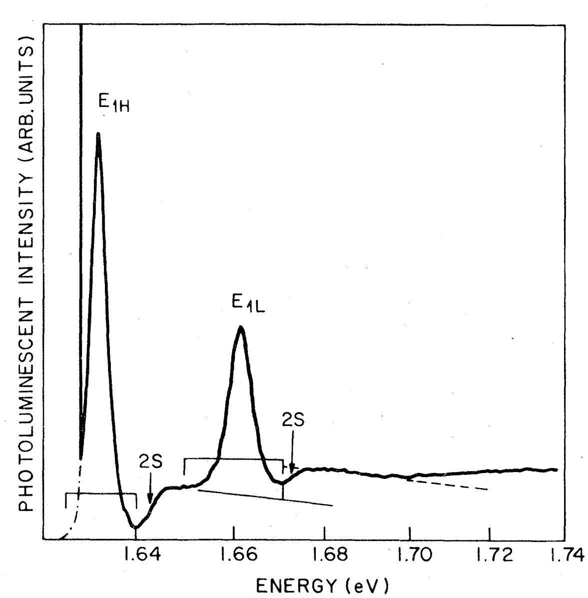

FIG. 1. Excitation spectrum (photoluminescence detected at 1.6288 eV as a function of excitation pump energy at ~ 0.3 W/cm^2) for a single-well sample with $L = 42$ Å. Arrows show the energies of the $2S$ exciton as explained in the text. Lines show constructions for obtaining the integrated intensities of the heavy-hole (E_{1H}) and light-hole (E_{1L}) exciton peaks.

luminescence at the light-hole continuum edge is about $\frac{4}{3}$ that at the heavy-hole continuum edge as would be expected since the heavy and light-hole bands have relative strengths[6] of 3 and 1, respectively.

According to our interpretation, the apparent continuum edge actually corresponds to the $2S$ excited state of the exciton. The true continuum corresponding to the ionization threshold is not directly seen in the spectra. The midpoint of the rising portion of the shoulder is taken as the $2S$ level, as indicated by arrows in Fig. 1; the $1S$ levels are the peaks E_{1H} and E_{1L}. In this way we have directly obtained from the spectra the term value $B_{1S}-B_{2S}$ for a number of samples with different L. The results, believed accurate to about 0.5 meV, are plotted (points) in Fig. 2.

If the well width L is sufficiently small, the heavy and light holes moving along the layer normal direction $\hat{z}$ (masses m_H, m_L, respectively) are uncoupled. The Hamiltonian for an electron and hole can then be written

$$H = H_e + H_h + H_x \ , \tag{1}$$

$$H_x = \frac{P^2}{2M\pm} + \frac{p^2}{2\mu\pm} - \left(\frac{e^2}{\epsilon}\right)\Big/(r^2+z^2)^{1/2} \ , \tag{2}$$

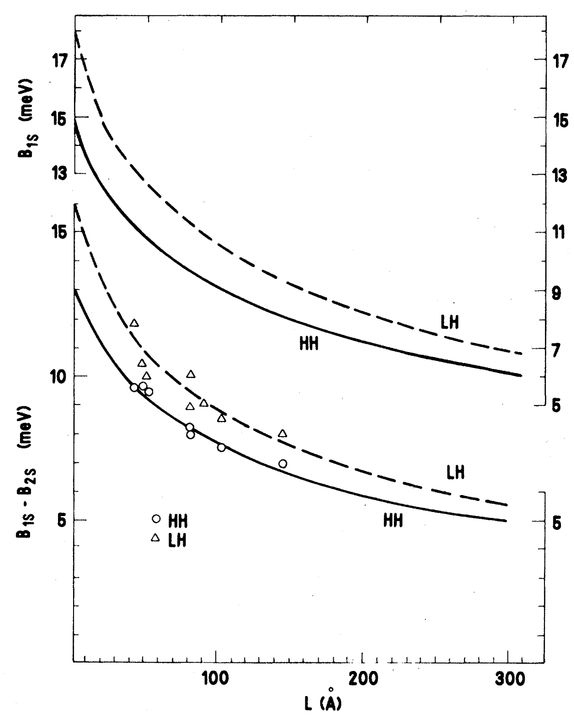

FIG. 2. 2D exciton term value $B_{1S}-B_{2S}$ as a function of GaAs layer width L for the heavy hole (HH) and light hole (LH). Points are measured values and curves are calculated with no adjustable parameters. Also shown is the calculated exciton $1S$-binding energy B_{1S}.

where H_e (H_h) describe the motion of the electron (hole) along $\hat{z}$ in the quantum well, and H_x describes the 2D exciton in the xy plane with total masses $M\pm$ and reduced masses $\mu\pm$ corresponding to the heavy (+) and light (−) holes. In Eq. (2), $z = z_e - z_h$, $\vec{r} = \vec{r}_e - \vec{r}_h$ is the relative coordinate in the xy plane, $\vec{p}$ is the momentum conjugate to $\vec{r}$, and we take the total momentum in the xy plane $\vec{P} = 0$. It is assumed that H is dominated by $H_e + H_h$ so that the wave function can be written in factored form

$$\psi = \phi(r,z)\,\omega_e(Z_e)\,\omega_h(Z_h) \ , \tag{3}$$

where $\omega_e(Z_e)$ $[\omega_h(Z_h)]$ is an eigenfunction of H_e (H_h), and $\phi(r,z)$ describes the 2D exciton. The appropriate reduced masses $\mu\pm$ appearing in Eq. (2) are given by

$$\mu_{\pm}^{-1} = m_e^{-1} + (1 \mp \tfrac{1}{2})(2m_H)^{-1} + (1 \pm \tfrac{1}{2})(2m_L)^{-1} \ . \tag{4}$$

The appropriate dielectric constant ϵ we take to be $\epsilon = \sqrt{\epsilon_\omega \epsilon_b}$, where ϵ_ω, ϵ_b are the dielectric constants of the well and barrier material, respectively. This comes from a consideration of the equivalent dielectric continuum for thin wells and barriers of equal width.

The binding energy corresponding to ψ is

$$B = -\int\int\int \psi H \psi r \, dr \, dz_e \, dz_h \ . \tag{5}$$

We assume that both particles are in the lowest quantum-well state adequately described by[1]

$$\omega(z) = \sqrt{2/L}\,\cos(\pi z/L), \quad |z| < L/2 \ . \tag{6}$$

We calculate B_{1S} using the exciton function

$$\phi_{1S}(r,z) = \frac{2}{b}\,\frac{\exp(|z|/b)}{(1+2|z|/b)^{1/2}} \times \exp\left[\frac{-(r^2+z^2)^{1/2}}{b}\right] \ , \tag{7}$$

and varying b to maximize Eq. (5). The result in general lies below the true B_{1S}, but should be very close in cases of interest here because Eq. (7) is exact in the thin-layer limit $z^2 \to 0$ (i.e., $L \to 0$). When $L > b$ calculations show that Eq. (7) gives significantly greater binding than the simpler function $\phi_{1S}(r,0)$ used previously.[7]

Our calculation of B_{2S} depends upon the fact that the $2S$ wave function has relatively little density in the region where z^2 is important in the potential. Therefore, we use

$$\phi_{2S}(r) = (2/c\sqrt{3})[1-(2r/c)]e^{-r/c} \ , \tag{8}$$

and vary c to maximize Eq. (5). The result can, in general, lie above or below the true B_{2S}, but should be very close because it is exact for $L \to 0$.

The results of these calculations are shown by the solid (heavy hole) and dashed (light hole) curves in Fig. 2 giving B_{1S} (meV) and $B_{1S}-B_{2S}$ as a function of L (Å). The calculated term values $B_{1S}-B_{2S}$ with no adjustable parameters are in very good agreement with the data. With this confirmation of the theory the calculated B_{1S} may be considered reliable. The calculations are based on the following physical constants: $m_H=0.35$,[8] $m_L=0.080$,[8] $m_e=0.067$,[8] $\epsilon_\omega=13.1$,[9] and $\epsilon_b=11.4$.[9]

According to the theory[10] of the limiting 2D exciton ($L\rightarrow 0$) the spectrum $I_{\rm PL}(E)$ is given by

$$I_{\rm PL}(E)\propto\frac{2\Theta(E)}{1+\exp(-2\pi\sqrt{\mathcal{R}/E})}+\sum_n\frac{4\mathcal{R}}{(n-\frac{1}{2})^3}\delta(E-En)\ , \tag{9}$$

where the sum is over discrete levels, $1S$, $2S$, . . . ,nS, E is measured from the true continuum threshold, $\mathcal{R}=e^4\mu/2\epsilon^2\hbar^2$ is the Rydberg ($\mathcal{R}_+=3.7$ meV, $\mathcal{R}_-=4.5$ meV), and $\Theta(E<0)=0$, $\Theta(E>0)=1$. It is permissible to keep only $n=1$ in the sum and include $n>1$ with the continuum, which is essentially constant over the energy range accessible here. We define an integrated strength for the $1S$ exciton peak.

$$S=[I_{\rm PL}(c)]^{-1}\int_{1S}I_{\rm PL}(E)\,dE\ , \tag{10}$$

where $I_{\rm PL}(c)$ refers to the continuum $I_{\rm PL}$. According to Eq. (9), $S(L\rightarrow 0)=16\mathcal{R}$. To generalize this result to finite L we note that the main effect of finite L is to spread the $1S$ state (i.e., increase b) while the $2S$ and higher discrete states [and therefore $I_{\rm PL}(c)$] are only slightly affected. The contribution of the $1S$ state in Eq. (9) is proportional to $|\phi(0,0)|^2\propto b^{-2}$, and $b(L\rightarrow 0)=a/2$, where $a=\epsilon\hbar^2/e^2\mu$ is the Bohr radius ($a_+=160$ Å, $a_-=130$ Å). Therefore, the theory predicts a strength

$$S=4\mathcal{R}(a/b)^2\ . \tag{11}$$

We have measured S for the three samples ($L=42$, 48.5, and 51 Å) in which the heavy- and light-hole $1S$ peaks and continuum thresholds were sufficiently well resolved. We illustrate in Fig. 1 the necessary constructions. Brackets indicate the $1S$ integration range. The base line for integrating E_{1L} is the sloping line drawn through the heavy-hole continuum starting point and having the slope (dashed line) of the light-hole continuum. The low-energy side of E_{1H} (partially obscured because of the nearness to the detection energy) was drawn in by hand to produce a symmetrical peak of nearly the same width as E_{1L}. We obtain the following results [theoretical values from Eq. (11) in parentheses]:

$$\begin{aligned} &L=42\text{ Å}:\ I_+=29(35),\ \ I_-=32(40)\text{ meV}\ ,\\ &L=48.5:\ I_+=41(34),\ \ I_-=44(38)\ ,\\ &L=51:\ I_+=20(33),\ \ I_-=31(37)\ . \end{aligned} \tag{12}$$

Since the various constructions make the measured values uncertain to about ±25%, a significant discrepancy between measurement and theory exists in only one case [I_+(51 Å)]. The general agreement is satisfactory and strongly confirms the excitonic interpretation of the peaks.

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