Biexcitons in GaAs quantum wells

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Careful examination of the exciton edge of the photoluminescence from a number of high-quality multiple-GaAs-quantum-well samples grown by molecular-beam epitaxy reveals at low temperatures a double peak whose splitting of approximately 1 meV decreases somewhat with increasing GaAs well width L. The higher-energy peak is due to the $n=1$ free-heavy-hole-exciton transition while the excitation intensity, temperature, and polarization dependences of the lower-energy peak suggest that it is due to biexcitons with a binding energy B of about 1 meV. In support of the biexciton interpretation a theoretical calculation of $B(L)$ is presented. This calculation gives two-dimensional biexciton binding energies more than an order of magnitude larger than the three-dimensional calculated values.

The existence of free excitons in high-quality GaAs-$Al_xGa_{1-x}As$ quantum-well samples grown by molecular-beam epitaxy (MBE) has been well documented.[1,2] Also, the experimental observation of the $2S$ excited state of these excitons as a function of the GaAs quantum-well width L has been shown to be in good agreement with a theory which gives the L dependence of the $2S$ excited state and the $1S$ ground state.[3] Careful examination of the photoluminescent spectra $I_{PL}(E)$ of a number of high-quality multiple-quantum-well (MQW) samples, $50 \lesssim L \lesssim 1000$ Å, shows a split peak in the region of the exciton luminescence. The dependences of these double peaks on the excitation intensity and sample temperature, coupled with the polarization of the luminescence, suggests that, in contrast to bulk GaAs, biexcitons are present in GaAs quantum wells at low temperatures for values of L ranging from 81 to at least 327 Å. The separation of the two peaks, ≈ 1 meV, is found to decrease with increasing L. A theoretical calculation of the biexciton binding energy $B(L)$ supports the biexciton interpretation.

The samples discussed herein were grown by MBE under conditions which led to intense, sharp, photoluminescence peaks and excitation spectra which show that the quantum wells are extremely uniform in thickness.[4] $Al_{0.3}Ga_{0.7}As$ was employed for the ~ 200-Å barriers, and the substrates were (100). Almost all samples grown under these conditions exhibit spectra which show a sharp double peak in the photoluminescence in the region of the exciton; these double peaks have also been seen by others with some samples.[5] The photoluminescence was excited with a cw-tunable dye laser at normal incidence to the plane of the layers. Luminescence emitted in the backward (reflection) direction 24° off normal incidence was detected with a photomultiplier after passing through a $\frac{1}{2}$-m monochromator. Circular polarization techniques were utilized in both the excitation and detection of the luminescence.[3] The samples were mounted in a variable-temperature cryostat and the temperatures mentioned herein were measured with a thermocouple soldered to a copper block screwed to the copper header which was in turn threaded into the tail of the cryotip. The samples were affixed to the header using a thin layer of rubber cement along one edge.

The photoluminescent spectra $I_{PL}(E)$ are functions of the excitation intensity I_p (~ 1 mW/cm^2 to ~ 10 W/cm^2) and the sample temperature T. At the lowest values of I_p one peak is usually observed and the excitation spectra show that it is due to the $n=1$ free-heavy-hole-exciton transition as in Ref. 1. As I_p is increased, a shoulder appears on the low-energy side and grows approximately as $(I_p)^{1.4}$. Finally, two distinct peaks are seen at the higher I_p's with a separation of ~ 1 meV. Data at 5 K for a MQW sample, $L=222$ Å, which exhibits two peaks, are shown in Fig. 1(a) for $I_p=0.2$ W/cm^2 at $E_p=1.537$ eV. This splitting and the position of the free-exciton peak are independent of I_p. As T is increased to about 11 K, $kT \sim 1$ meV, the free-exciton peak dominates over the lower-energy peak and its luminescence is enhanced if the sample is warmed when I_p is such that there are two peaks at 5 K. Figure 1(b) illustrates the effects on the intensity with $T=19$ K for the same sample and conditions discussed in connection with Fig. 1(a). The intensity of the lower-energy peak is estimated to vary with T as $\exp(-1.0\ \text{meV}/kT)$.

The temperature dependence mentioned above has been observed with all samples which exhibit these two sharp peaks in $I_{PL}(E)$. The change from one peak to two peaks in the luminescence with increasing I_p has been observed for samples with L in the range 81 to 327 Å and the I_p "threshold" for the

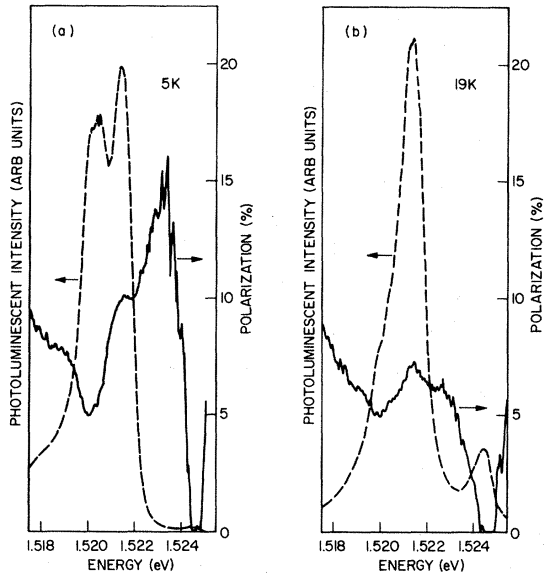

FIG. 1. Photoluminescent intensity I_{PL} and its polarization ρ vs photon energy E for a multiquantum well sample with $L=222$ Å. The sample was excited with 0.2 W/cm^2 at 1.537 eV. The peak in $I_{PL}(E)$ at 1.5214 is due to the $n=1$ free-heavy-hole-exciton transition E_{1h}, while the peak at 1.5203 seen at 5 K [(a)] is believed to be due to biexcitons. When T is increased to 19 K [(b)] the lower-energy peak disappears while that due to E_{1h} gains strength. At 5 K, ρ shows a sharp decrease as expected at the peak ascribed to biexcitons. The $n=1$ free-light-hole-exciton transition at 1.5246 eV gives rise to a negative polarization consistent with this assignment. However, the electronics for these spectra were such that negative values of ρ were recorded as $\rho=0$.

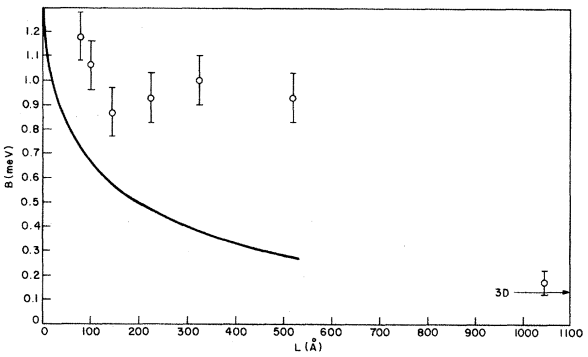

FIG. 2. Observed splitting of the double peak vs GaAs well width. The present theoretical results on the biexciton binding energy $B(L)$ are shown by the solid curve. The calculated 3D value from Ref. 8 is also indicated.

second peak tended to decrease with increasing L. Two samples with $L>327$ Å exhibited two peaks even at the lowest intensities ~ 1 mW/cm^2.

Figure 2 shows the splitting of the two peaks measured from $I_{PL}(E)$ with no line-shape analysis as a function of L. Experimental uncertainties are estimated to be ± 0.1 meV. Similar data for some samples ($L=200$ Å) grown under nonoptimum conditions, i.e., under conditions which are known to produce roughened GaAs-$Al_xGa_{1-x}As$ interfaces,[4] have been found to exhibit splittings a few tenths of a millielectron volt higher than those shown in Fig. 2.

When excitation is with circular polarized light,[2] a pronounced minimum in the polarization ρ at the lower-energy peak has been observed. A good example of this is shown in Fig. 1(a). This observation is consistent with zero polarization for the process responsible for the lower peak. We attribute the lower peak to biexcitons which, if present, would be expected to decay radiatively with zero polarization. This interpretation is also consistent with the superlinear dependence of $I_{PL}(E)$ for the split peak on I_p, not expected for excitons bound on neutral centers. Likewise it is consistent with the dependence on T which shows that the lower-energy peak involves some exciton complex rather than a completely different type of e-h recombination process. However, in some samples there was not a distinct minimum in ρ at the lower-energy peak, which we tentatively attribute to the need for a detailed line-shape analysis. Pending this detailed analysis we assign the peak separation to the biexciton binding energy $B(L)$.

We have carried out a theoretical calculation of the two-dimensional (2D) biexciton binding energy

$$B(L)=B_{2e,2h}(L)-2B_{1S}(L) \ , \tag{1}$$

where $B_{2e,2h}$ is the total binding energy of a system of two electrons and two holes, and B_{1S} is the 2D single-exciton binding energy discussed in Ref. 3. We have calculated $B_{2e,2h}(L)$ variationally using the two-parameter completely symmetric wave function $\Psi(\beta,kr)$ of Hylleraas and Ore[6]

$$\begin{aligned} &\Psi(\beta,r)=\exp[-(s_1+s_2)/2]\cosh[\beta(t_1-t_2)/2] \ , \\ &s_1=r_{1a}+r_{1b} \ , \quad s_2=r_{2a}+r_{2b} \ , \\ &t_1=r_{1a}-r_{1b} \ , \quad t_2=r_{2a}-r_{2b} \ , \end{aligned} \tag{2}$$

where 1,2 refer to electrons and a,b to holes, a symmetric kinetic energy with $m_e=m_h$, and a potential energy of the form

$$\begin{aligned} &V(r)=(e^2/\epsilon r)(1-e^{-\gamma r}) \ , \\ &V=V_{1a}+V_{1b}+V_{2a}+V_{2b}-V_{ab}-V_{12} \ . \end{aligned} \tag{3}$$

The parameter γ, a measure of L^{-1}, was determined for each L by requiring that Eq. (3) give the same B_{1S} as was obtained in Ref. 3. We define the following integrals (all functions of β) with the scaling

parameter k set to $k=1$:

$$N=\int \Psi^2 d\vec{r}\,, \quad M=\frac{1}{2}\sum_i \int (\nabla_i \Psi)^2 d\vec{r}\,,$$
$$L=\int \Psi^2 V d\vec{r}\,, \quad D=\frac{\partial L}{\partial \gamma}\,. \tag{4}$$

Introducing the scaling parameter k, and ignoring the scaling of the volume element $d\vec{r}$ which cancels out of all results, gives

$$N\to N\,, \quad M\to k^2 M\,,$$
$$L(\gamma)\to kL(\gamma/k)\,, \quad D\to D(\gamma/k)\,. \tag{5}$$

The variational expression for the binding energy is then

$$B_{2e,2h}=[2kL(\gamma/k)-k^2 M]/N\,, \tag{6}$$

in units of the exciton Rydberg ($e^4\mu/2\epsilon^2\hbar^2$). Maximizing $B_{2e,2h}$ with respect to k leads to the relation

$$\gamma=p[L(p)-pD(p)]/M, \quad p=\gamma/k\,, \tag{7}$$

with γ^{-1} in units of the exciton Bohr radius ($\epsilon\hbar^2/e^2\mu$). It is therefore possible to calculate both $B_{2e,2h}$ and γ for any choice of β and p. Finally $B_{2e,2h}$ is maximized with respect to β by selecting p for each β so as to keep Eq. (7) constant. When this is done Eq. (6) can also be written in the form

$$B_{2e,2h}=[L(p)^2-p^2D(p)^2]/MN\,. \tag{8}$$

The limit $\beta\to 1$ in Eq. (2) describes two noninteracting excitons so we must have

$$B_{2e,2h}\underset{\beta\to 1}{\to} 2B_{1S}\,. \tag{9}$$

In this case an elementary treatment yields the simple relations that correspond to Eqs. (6) and (7)

$$B_{1S}=[2kp/(1+p/2]-k^2\,,$$
$$\gamma=p^3/[2(1+pk)^2]\,, \tag{10}$$

which provide a check on the numerical accuracy of the computations.

Any factor common to N,M,L,D cancels out of the final results; because of this and the symmetry of Ψ the eightfold integrals in Eq. (4) reduce to fivefold integrals over the volume element and limits given by

$$d\vec{r}\to r_{ab}dr_{ab}J_1 ds_1 dt_1 J_2 ds_2 dt_2\,,$$
$$J=(s^2-t^2)/[(s^2-r_{ab}^2)(r_{ab}^2-t^2)]^{1/2}\,, \tag{11}$$
$$0\le r_{ab}<\infty\,, \quad -r_{ab}<t<r_{ab}\,, \quad r_{ab}\le s<\infty\,.$$

The integrals over 1,2 were done analytically in terms of products of modified Bessel functions leaving a rather complicated single integral over r_{ab} which was done using the Gauss-Laguerre numerical quadrature algorithm.[7] The results for $B(L)$ for (100) GaAs quantum wells are shown by the solid curve in Fig. 2. There is a strong drop with increasing L at small $L<100$ Å much like that found[3] for $B_{1S}(L)$. Values used for the Rydberg (3.7 meV) and the Bohr radius (160 Å) were taken from Ref. 3.

Inasmuch as $B(L)$ is the difference of two quantities there is no guarantee that our results are a lower bound on B. However, we believe that $B_{1S}(L)$ is quite accurately known, so we suggest that $B(L)$ is probably a lower bound on the biexciton binding energy. The symmetry assumption $\sigma=m_e/m_h=1$ may seem strange until it is realized that the heavy hole for motion normal to the well has quite a small mass ($\sigma\sim 0.68$) for motion parallel to the well. Brinkman, Rice, and Bell[8] have studied the 3D biexciton using a six-parameter variational function. They find for the symmetric case ($\sigma=1$) $B=0.029$ compared with the Hylleraas-Ore value of 0.017; at $\sigma=0.68$ there is only a slight increase to $B=0.031$ (~ 0.13 meV) which suggests that our assumption $\sigma=1$ is not causing a serious error. It is reasonable to expect, on the basis of the 3D results, that the use of the Brinkamn-Rice-Bell function in our calculation would give about 70% greater binding energies. Nevertheless, our results establish that the 2D biexciton is much more strongly bound than the 3D biexciton (by a factor >10 at $L=0$), and our assignment of the ≈ 1-meV splittings to the biexciton is reasonable.

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