Tunneling-assisted photon emission from quantum wells

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Luminescence and excitation spectra were studied on molecular-beam-epitaxially grown GaAs-$Al_{0.44}Ga_{0.56}As$ quantum-well structures consisting of ten periods, each period having a 100-nm well separated from a 5-nm well by a 5-nm barrier. Luminescence below the photon energy of the heavy-hole exciton transition in the narrow well was observed and accounted for in position, shape, and strength in terms of tunneling-assisted photon emission between the narrow and wide wells.

Tunneling-assisted photon emission was proposed many years ago to explain below-band-gap emission in electroluminescent diodes[1-3] and has been invoked many times since in at least a qualitative manner. The complications of junction structure, however, have made unambiguous identification of this process difficult. Recently the process has been invoked in explaining below-band-gap photoresponse in heterojunctions.[4] By tunneling-assisted photon emission (TAPE) we mean a *single-step* process in which a photon is emitted and the electron is predominantly in a different location relative to a barrier following the emission. This is distinct from the two-step process of electron tunneling, relaxation, and then photon emission.[5] It would appear that undoped, coupled quantum wells in the GaAs-$Al_xGa_{1-x}As$ system offer a simpler and more controlled environment for the study of the TAPE process. We performed such a study and observed TAPE from quantum wells for the first time. Its observation provides a measurement of the heavy-hole binding energy in the well.

Normally in a single narrow($\sim$5-nm width) quantum well of high quality, luminescence at low temperatures occurs predominantly from the $n=1$ heavy-hole exciton.[6] If such a well were separated from bulk GaAs by a narrow barrier ($\lesssim$5-nm width), luminescence could also be expected from TAPE arising from recombination of an electron trapped in the well and a hole at the valence-band edge of the bulk GaAs. To enhance observability we used a multilayer wafer of ten periods in which the narrow GaAs well is separated by the $Al_xGa_{1-x}As$ barrier from a very wide well (henceforth called the "tub") of GaAs of $\sim$100-nm width, which approximates the bulk crystal (see Fig. 1).

The structures were grown[7] by molecular beam epitaxy on a GaAs [100]-oriented Si-doped substrate under arsenic-rich growth conditions with an As_2 beam, a substrate temperature of 650 °C, and a GaAs growth rate of 18 nm per minute. The luminescence was excited with a tunable cw dye laser beam that was at normal incidence to the plane of the layers and focused to a spot 200 μm in diameter. The signal was detected in the reflection direction 24° off normal incidence and analyzed by means of a $\frac{1}{2}$-m monochromator equipped with a cooled photomultiplier tube which provided a resolution of about 0.3 meV. The sample was mounted by one edge with rubber cement on a copper header that was part of a variable-temperature He cryostat.

Figure 1 shows the luminescence of the sample at $\sim$6 K when excited at the $n=1$ light-hole exciton well transition at 1.6367 eV. Also shown is an excitation spectrum when the luminescence detector is set at the heavy-hole exciton well transition at 1.6042 eV. The most interesting feature is the peak at $\sim$1.59 eV in luminescence. Because of its large energy above the tub luminescence (in comparison to the thermal energy) and because of its close proximity to the heavy-hole exciton well transition, it can be concluded that the transition involves an electron trapped in the lowest subband in the conduction-band well.[8]

Several considerations argue strongly against the luminescence peak at 1.59 eV being extrinsic in origin. First, a considerable amount of previous experience has

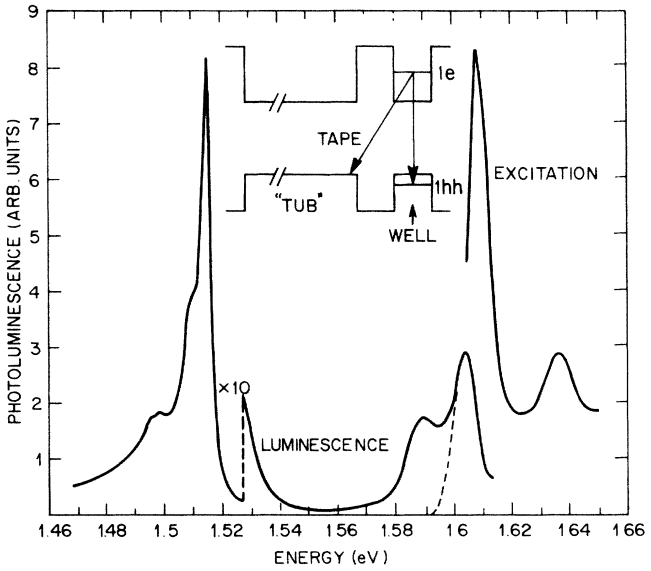

FIG. 1. Luminescence from the "tub" (near 1.515 eV) and well when excited at 1.6367 eV, the light-hole exciton well transition, and an excitation spectrum taken with detection luminescence at 1.6042 eV, the heavy-hole exciton well transition (see the text concerning the dashed line). Inset: schematic band-edge diagram with relevant well states and transitions shown.

shown that unintentional extrinsic luminescence in quantum wells depends sublinearly on excitation intensity within the accessible range. However, the 1.59-eV peak retained its strength relative to the heavy-hole exciton transition to the highest excitation intensity available (55 W/cm^2). Second, the strength of luminescence from heavy-hole excitons bound to neutral acceptors has been found[9] to decrease markedly as the sample temperature is raised from 5 to 19 K. When the temperature of the present sample was raised to 29 K, the luminescence peak at 1.59 eV increased its strength relative to the heavy-hole exciton transition by a small amount. Third, in the study of over 100 quantum-well wafers the only unintentional extrinsic luminescence observed[10] is that believed to be due to carbon which enters during the AlGaAs deposition, floats on the growth surface, and then is deposited in a thin layer of GaAs when the Al deposition is terminated. This luminescence has been found[10] to be avoidable by growing only thin AlGaAs layers. This was done in the present sample.

The 1.59-eV luminescence is thus a candidate for interpretation as a group of TAPE transitions involving an electron in the lowest well state and holes in the lowest few states of the tub. Transitions of this sort should have photon energies beginning at about 1.59 eV and extending to higher energies in a closely spaced series. The effects of line broadening and Stokes shift would appear to allow those transitions to account for the emission band centered at 1.59 eV. To make this identification more precise it is necessary (a) to determine the energy levels of the tub-well system, (b) to separate the 1.59-eV transition from the heavy-hole exciton well transition, and (c) to determine the carrier temperature. We will describe these procedures in order.

From the excitation spectrum of Fig. 1 the heavy- and light-hole exciton transitions in the well are seen to be 1.6087 and 1.6367 eV, respectively. Subtracting the low-temperature band-gap energy of GaAs, 1.5192 eV, from these energies yields the sum of the electron and hole energy levels in the well minus the exciton binding energy, which must be determined self-consistently for the tub-well system parameters deduced.

The exciton binding energies for the tub-well system were computed variationally using the energy expression

$$E(\beta)=\frac{\hbar^2}{2\mu\beta^2}-\frac{e^2}{\epsilon_0}\int_{-\infty}^{\infty}dz\,D(z)\left[\frac{2}{\beta}\right]^2 \times\int_0^{\infty}dr\frac{re^{-2r\beta}}{(r^2+z^2)^{1/2}}\ , \tag{1}$$

where β is a parameter to be determined by minimizing the energy, μ is the reduced mass for planar motion, r and z are the electron-hole relative coordinates in the plane and normal to the plane, respectively, and $D(z)$ is a normalized density distribution in z which depends on the quantum-well wave functions.

A simplified model was used for representing $D(z)$, which facilitates the evaluation of Eq. (1), based on the superposition of two Gaussians, one centered on the well and one on the tub, in which each Gaussian is multiplied by an occupancy factor depending on P_e (or P_h) which gives the probability of finding the electron (or hole) in the well. This leads to

$$D(z)=\frac{P_eP_he^{-z^2/2c^2}}{c(2\pi)^{1/2}}+\frac{(1-P_e)(1-P_h)e^{-z^2/2b^2}}{b(2\pi)^{1/2}} +\frac{P_e(1-P_h)}{[\pi(b^2+c^2)]^{1/2}}e^{-(s+z)^2/(b^2+c^2)} +\frac{P_h(1-P_e)}{[\pi(b^2+c^2)]^{1/2}}e^{-(s-z)^2/(b^2+c^2)}\ , \tag{2}$$

where s is the separation of the tub and well centers, and b (c) is the width of the well (tub) Gaussian. The widths are determined by the well and tub widths so as to reproduce the correct exciton binding energy[11] of a single well.

The sum of the electron and heavy or light hole energies in the well is used to determine the well and tub widths self-consistently. The calculation of the energy levels of the tub-well system uses continuity of the wave function ψ and $m^{-1}\partial\psi/\partial z$ at the interfaces, where m is the [100] effective mass of the carrier. The dependence of the effective masses and the bandgap on Al concentration in the alloy was taken from Ref. 12. The fractional splitting of the band-gap discontinuity at the interfaces of differing composition was taken as 0.60 in the conduction band and 0.40 in the valence band.[13] The nonparabolicities of the conduction band were calculated from the five-band model of Lawaetz as 4.9×10^{-15} cm^2 for GaAs (Ref. 14) and 2.0×10^{-15} cm^2 for AlAs. The nonparabolicities of the heavy-hole valence band were taken as negligible in the two materials and those of the light-hole valence band as equal to the conduction-band values. Values for the alloys were interpolated linearly. The fractional Al concentration was set at 0.44, a value between 0.42 determined from barrier luminescence and 0.45 determined from the growth. The barrier was set at 5.25 nm determined from transmission electron microscopy (TEM). The fitting then led to a well width of 5.04 nm and a tub width of 90.3 nm compared to 4.7 and 91 nm observed in TEM. The exciton energies, significantly affected by the tub-well interaction, were found self-consistently with these values to be 7.6 and 8.1 meV for the heavy and light holes, respectively. Besides the energy levels of the tub-well system the wave functions and occupancy probabilities in the tub and well and surrounding barriers were calculated.

The separation of the 1.59-eV luminescence and the heavy-hole exciton luminescence was determined by a comparison of luminescence and excitation spectra shown in Figs. 1 and 2. The excitation spectra of Fig. 2 were taken for four wavelengths of luminescence detection in the 1.59-eV luminescence band. First, it should be noted that the heavy-hole exciton peak shifts as the luminescence detection wavelength is changed. This indicates that these excitation curves cannot be interpreted directly as absorption curves. Rather, they are affected by the thermalization routes (hence populations) contributing to the luminescence. The curves, however, can be used in the following way: The strength of the heavy-hole exciton in excitation is taken in ratio to the strength in the lumines-

cence spectrum at the wavelength used for excitation spectrum determination. For luminescence photon energies of 1.561, 1.585, and 1.590 eV this ratio is 1.13, while at 1.604 eV (Fig. 1) it is 2.85. These two ratios indicate two different thermalization routes and can be used to apportion the strength of the two processes at the intermediate luminescence energy of 1.596 eV where the ratio is 1.36. This apportionment indicates that only 13% of the luminescence at 1.596 eV arises from the heavy-hole exciton. This allows drawing the low-energy side of the heavy-hole exciton peak as the dashed line in Fig. 1.

Next the carrier temperature at the time of recombination is determined by accounting for the Stokes shift of the heavy-hole exciton in Fig. 1. A Gaussian $\exp[-(E-E_H)^2/a_H^2]$ was first fit to the absorption spectrum (see Fig. 3), which determined $E_H=1.6087$ eV and $a_H=0.0057$ eV. The carrier temperature was then found from the Boltzmann factor $\exp[-(E-E_H)/kT]$ that must relate emission and absorption.[15] The fit to the luminescence spectrum shown in Fig. 3 was obtained for $kT=0.0037$ eV corresponding to 43 K. This is considerably above the lattice temperature of ~ 6 K as determined by a thermocouple. The latter is believed to be a correct lattice temperature since no line broadening was observed in this sample as the excitation intensity was increased in the range of intensities used for the measurements in Figs. 1 and 2. Thus, the hot carrier temperature at the time of luminescence results from the excitation photon energy, not the excitation beam energy.[16]

The above fit of the heavy-hole exciton luminescence gives a scaling constant representing the strength of this exciton on the scale plotted. That constant was found to be $g_H=15.3$. A relation has been found[11] between the integrated strength of this exciton transition and the amplitude A_H of the continuum absorption resulting from this state. If the integrated strength of the exciton transition is in turn related to the amplitude of a Gaussian line shape, that relation[11] is given by $A_H=\pi^{1/2}\beta^2 a_H g_H/4R$, where R is the Rydberg (3.7 meV) and β^{-2} is a strength constant[11] calculated to be 0.88. This gives $A_H=0.776 g_H=11.9$ on the scale of g_H.

A fit to the 1.59-eV luminescence peak can now be attempted in terms of TAPE transitions. Figure 4 shows the 1.59-eV luminescence after separation from the heavy-hole exciton luminescence by the procedure described above. Also shown are discrete lines representing positions of the TAPE transitions calculated from the quantum-mechanical tub-well system model as lowered slightly by exciton binding (see below). They represent transitions of the electron in the lowest well state to holes in the lowest few states of the tub. They are shown schematically weighted by a Boltzmann factor for 43 K and with light-hole transitions reduced by a factor of $\frac{1}{3}$.

In fitting to the observed luminescence spectrum the absorption spectrum of each TAPE transition is modeled by a function

$$re^{-(E-E_i+E_X)^2/a_P^2}+\begin{cases} e^{-(E-E_i-w)^2/a_P^2} & \text{if } E-E_i-w\le 0 \\ 1 & \text{if } E-E_i-w>0 \end{cases}, \tag{3}$$

where r represents the strength of the exciton relative to the continuum, E_i is the transition energy from the quantum mechanical model, E_X is the exciton binding energy, and w is equal to the broadening constant a_P multiplied by $(\ln 2)^{1/2}$. On the basis of Ref. 11 $r=4R/\pi^{1/2}\beta^2 a_P$ and β^{-2} is calculated to be 0.134 for heavy holes and 0.110 for light holes. On the same basis E_X is calculated to be 0.0021 eV for heavy holes and 0.0022 eV for light holes. The functions (3) are summed over all the transitions (with the functions for light hole transitions multiplied by $\frac{1}{3}$ times the ratio 0.05/0.04 of reduced light- to heavy-hole exciton masses for motion in the plane of the well), multiplied by the same Boltzmann factor as used for the

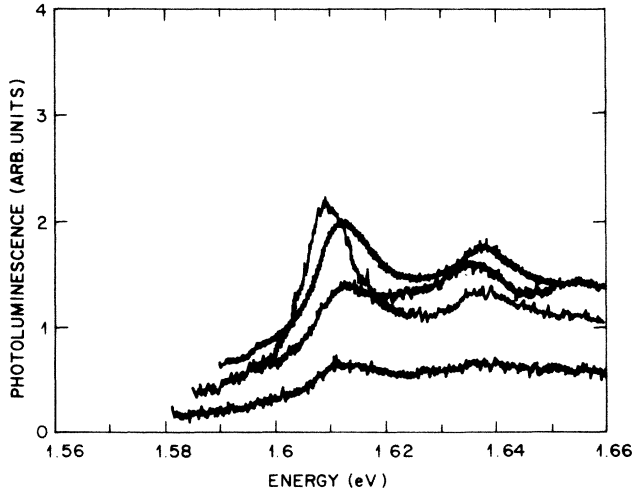

FIG. 2. Excitation spectra taken with four detection luminescence photon energies of 1.581, 1.585, 1.590, and 1.596 eV, each beginning slightly above its detection photon energy.

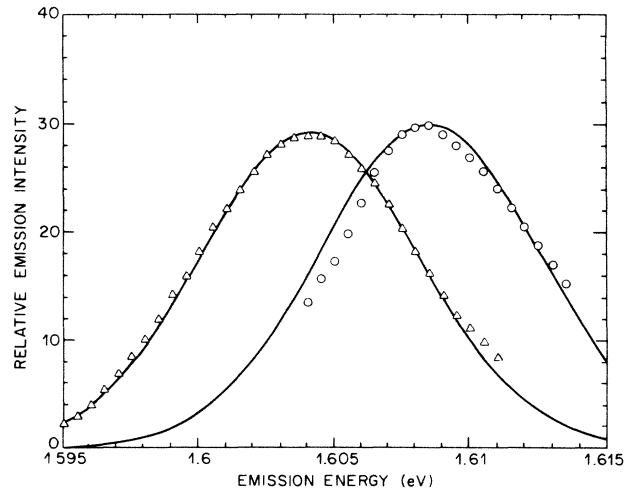

FIG. 3. Gaussian curve fit to the excitation (absorption) curve of the heavy-hole exciton well transition (circles) and a Stokes-shifted Gaussian curve fit to the corresponding luminescence curve (triangles).

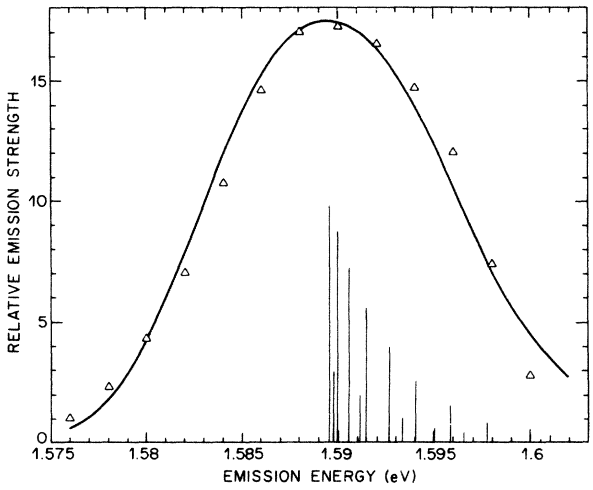

FIG. 4. Fit of the tunneling-assisted photon emission (TAPE) transitions to the luminescence curve (triangles). The vertical lines indicate the predicted TAPE transitions, without broadening, but with relative weighting expected at 43 K and with a factor of $\frac{1}{3}$ multiplying light-hole transitions.

heavy-hole well exciton with the temperature taken as 43 K as found above, and then fitted to the observed spectrum of Fig. 4 by varying only the broadening constant a_P and an overall scaling A_P (which is on the same scale as A_H found above). As can be seen in Fig. 4 a satisfactory fitting can be obtained for $a_P=a_H=0.0057$ eV and $A_P=0.0445$.

The scaling constants A_H and A_P found above can be used to find the relative strengths of a TAPE transition to the heavy-hole well transition provided the relative populations of holes in the tub and well can be determined. One way is to calculate the absorption at the exciting energy 1.6367 eV used for producing the luminescence spectrum and apportioning the relative excitation in the tub and well on the basis of the integrated occupancy probabilities of the excited states in the two spatial regions. This is done with the use of the quantum mechanical model of the tub-well system described above. This gives the relative hole populations at absorption as 0.927 in the tub and 0.073 in the well. The combination of low lattice temperature and the lack of intentional doping prevented any significant equilibrium hole concentration contributions from acceptors in the tub or well to these numbers. A significant hole concentration from unintentional acceptor doping in the barriers can also be ruled out because the holes would collect in the tub only and not in the well. That would make the TAPE transition dependence on excitation intensity weaker than that of the heavy-hole exciton well transition, a situation which is different from the closely linear dependence observed for each. If we assume that on average carriers created by absorption in the tub (well) thermally relax in the tub (well), then the difference of the chemical potentials relevant to the TAPE and heavy-hole exciton well transitions can be calculated and used in interpreting the ratio of A_P/A_H. This procedure leads to the conclusion that a TAPE transition is about six times stronger than the heavy-hole exciton well transition, a clearly unrealistic result.

This reasoning leads us to consider that the holes (but not the electrons) in the tub and well are in thermal equilibrium, a situation which would reduce the number of holes available for heavy-hole well transitions relative to TAPE transitions by 10^3. This can happen only if a near-coincidence of energies of heavy-hole tub and well states occurs causing resonant tunneling of holes between the well and tub. The quantum-mechanical model, in fact, shows that there are three heavy-hole tub states within the transition energy linewidth which are available for resonant tunneling. This model also explains why TAPE transitions involving a well hole and a tub electron are too weak to observe near 1.54 eV.

We thus conclude that the strength of a TAPE transition is given simply by $A_P/A_H=3.7\times10^{-3}$. If the TAPE transition strengths as calculated by the quantum mechanical tub-well model are weighted by the fractional tub-hole populations and taken in ratio to the heavy-hole—electron well transition, a result of $A_P/A_H=1.3\times10^{-3}$ is found. We believe that the effects of subband mixing[17] can account for the difference of the observed and calculated strength of the TAPE transition.

Based on accounting for the position, shape, and strength, we believe we have observed tunneling-assisted photon emission from quantum wells. It is worth noting that the fit to position and shape gives additional support to the 60-40% split between conduction- and valence-band offsets at interfaces of GaAs and $Al_xGa_{1-x}As$.

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states" or "tub states." To find the photon emission probability, the overlap integral of the entire envelope wave functions of initial (conduction miniband) and final (valence miniband) states is calculated. If, as just defined, the initial state is a well state and the final state is a "tub" state (or vice versa), the transition is a TAPE transition.

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