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Voltage-controlled sharp-line electroluminescence in GaAs-AlAs double-barrier resonant-tunneling structures

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Highly resolved, acceptor-related sharp-line electroluminescence spectra in large area GaAs-AlAs doublebarrier resonant-tunneling structures are reported. Excitation of the lines is achieved by voltage-controlled tunneling in the bias range for tunneling into the lowest heavy-hole state of the quantum well of the doublebarrier structure. The sharp-line spectra are observed at low levels of current injection and are attributed to recombination of holes localized at acceptors at different monolayer planes in the 66-Å-wide quantum well, from well center to well edge, with electrons localized at potential fluctuations within the well. $[$ S0163-1829(98)50232-9]

The linewidths of photoluminescence (PL) or electroluminescence (EL) spectra in quantum wells $(QW's)$ are normally dominated by inhomogeneous broadening arising from well width or other potential fluctuations.¹ However, it has recently been shown that the contribution of discrete localized states can be resolved by the use of focused laser spots of \sim 1- μ m diameter.^{2–4} This strongly reduces the effects of spatial averaging, and permits the observation of recombination at individual quasi-zero-dimensional localized states.

We demonstrate here that selective excitation of sharpline EL can be achieved from voltage-controlled charge injection in *large area* GaAs-AlAs double-barrier resonanttunneling structures (DBRTS) embedded in $p-i-n$ junctions. Up to 30 lines are observed in a 40-meV energy range predominantly in the spectral region of electron to neutral acceptor (e- $A⁰$) transitions in the QW of the DBRTS.⁵ The sharp lines are observed at low currents in the region of resonant tunneling into the first heavy-hole $(HH1)$ confined levels of the QW. The tunneling process leads to injection of carriers into the confined states of the QW with little or no excess energy. The carriers relax from HH1 to the relatively low-lying defect levels and give rise to the EL. The spectra are observed in DBRTS grown under conditions that favor acceptor incorporation into the $QW's⁶$ and are attributed to recombination of holes localized at acceptor levels, with electrons localized in the QW's. The occurrence of sharp lines is attributed to the differing binding energies for acceptors located on the distinct monolayer planes within the $QW.⁷$

The results bear similarities to PL spectra excited with a \sim 1- μ m laser spot reported by Zrenner *et al.*³ in type-II GaAs/AlAs OW's. In that case sharp lines \sim 20–40 meV below E1-HH1 were observed, as in the present work, and were ascribed to recombination at quantum dotlike, interface fluctuations in the 30-Å QW. The energy range of the spectra was ascribed to fluctuations in well width by up to two monolayers, corresponding to a change in electron energy of 38 meV for a 30-Å QW. Furthermore, the energy spacing between the sharp lines was consistent with a lateral size for the fluctuations of \sim 1000 Å. By contrast in the present structures with a 66-Å QW, a two-monolayer change in width corresponds to only a 10-meV change in energy, making the involvement of acceptor-related localization, expected from the growth, essential to explain the energy range of the spectra.

Experiments were carried out at 2 K on two DBRTS in $p-i-n$ junctions (Table I). The samples $(A \text{ and } B)$, grown by molecular beam epitaxy (MBE) at 630 $^{\circ}$ C are two of a series studied in Ref. 6 (samples C and B of Ref. 6). The structures were grown on (100) substrates in the order p^+ , intrinsic (*i*), n^+ to facilitate doping of the upper $Al_xGa_{1-x}As$ contacts, employed to ensure efficient transmission of EL out of the structure. A by-product of this growth procedure is that Be acceptor diffusion from the lower p^+ GaAs into the QW may occur unless very wide spacer layers are employed. Sample A has the widest spacer (300 Å) in Ref. 6, while B has a 150-Å spacer. We concentrate here on sample A, since it shows the best resolved spectra. Sharp lines are also observed for sample B, but less well resolved, probably due to acceptor interaction effects.

Material	Thickness	Doping $\text{(cm}^{-3})$
GaAs	250 Å	$n = 2 \times 10^{18}$
$Al_{0,33}Ga_{0,67}As$	$1 \mu m$	$n = 1.3 \times 10^{18}$
GaAs	500 Å	$n = 2 \times 10^{18}$
GaAs	350 Å	$n = 1 \times 10^{16}$
GaAs	80 Å	Undoped
AlAs	50 Å	Undoped
GaAs	66 Å	Undoped
AlAs	50 Å	Undoped
GaAs	300 Å (A), 150 Å (B)	Undoped
GaAs	1000Å	$p = 5 \times 10^{17}$
GaAs	1000 Å	$p = 1 \times 10^{18}$
GaAs	$3 \mu m$	$p = 2 \times 10^{18}$
GaAs	Substrate	$n = 2 \times 10^{18}$

TABLE I. Layer composition of devices.

The EL was dispersed by a grating spectrometer and detected with a charge coupled device array. Compared to Ref. 6, the setup has $\sim 10^3$ times higher sensitivity, permitting observation of the new spectra that are only visible at low injection. Mesas of 200 - μ m diameters were studied with metallized finger contacts on the top surface. These contacts enhance the visibility of the sharp lines, possibly due to current injection in small regions. Nevertheless, sharp lines on a stronger monotonic background were also detected with annular contacts. A schematic band diagram is shown in Fig. $1(a)$. Under forward bias electrons and holes accumulate close to the left- and right-hand barriers, forming two-

FIG. 1. (a) Schematic band diagram of the double-barrier resonant-tunneling structures under bias in the region of the heavyhole 1 tunneling resonance. (b) Current-voltage characteristics of sample A.

FIG. 2. Sharp-line electroluminescence spectra for sample A as a function of bias from 1.596 to 1.616 eV. Excitation of sharp peaks to progressively higher energy with increasing bias is observed.

QW. *I*-*V* characteristics are shown in Fig. 1(b). Clear resonances are observed due to tunneling into the HH1, light hole 1 $(LH1)$ and E1 levels at 1.590, 1.655, and 1.705 V, respectively.⁸

FIG. 3. As in Fig. 2, but with spectra from 1.63 to 1.65 V. For biases greater than \sim 1.636 V, the E1-HH1 excitonic recombination has intensity comparable to that of the sharp-line spectra, and dominates the spectra for biases greater than \sim 1.645 V. Inset-selected sharp-line peak positions as a function of magnetic field from 0 to 14 T.

EL spectra in the bias range from 1.596 to 1.616 V in the region of HH1 are shown in Fig. 2, with spectra to higher bias close to the onset of LH1 in Fig. 3. At \sim 1.596 V sharpline EL is first observed, superimposed on the high energy tail of the GaAs band-edge EL. The lines have width ~ 0.3 meV, close to the spectrometer resolution. With increasing bias, lines to progressively higher energy are observed, the high energy cutoff of the lines occurring, e.g., at 1.594 eV for a bias of 1.616 V. The fact that the cutoff agrees within 20 meV with the bias shows that holes are injected into HH1 with very little excess energy relative to the highest energy defect line. The holes relax from HH1 into the lowest energy states available, with the energy of the highest line observed increasing with bias as the current increases and the lower energy states become filled. Recombination of the trapped holes then occurs with the low density of electrons that tunnel from the electron accumulation layer into the well.⁹ With increasing bias, the intensity of a particular line increases and then saturates, showing that a fixed number of centers contributes to each line. When significant current flow $(>5 \mu A)$ first occurs at \sim 1.645 V at the onset of LH1 tunneling (Fig. 3), the E1-HH1 excitonic peak at 1.614 eV rapidly increases in intensity and then dominates the spectra. 10 In addition, at higher currents, the sharp lines become significantly less well resolved.

The peak of the distribution of the sharp-line EL occurs \sim 28 meV below E1-HH1, close to that expected for (E1-A⁰) recombination in a $66-\text{\AA}$ QW.⁵ The binding energy of a carrier to an impurity in a QW is expected to be dependent on the impurity location in the QW, peaking at the center and decreasing in a discrete fashion for monolayer planes from the center to edge.⁷ For the present 66- \AA QW the holebinding energy (E_A) to Be acceptors at the center of the QW is expected to be 36 meV, decreasing to \sim 21 meV at the edge.¹¹ For a uniform distribution, the density of acceptor states is a maximum at an energy corresponding to the oncenter binding energy (36 meV) . Thus the e-A⁰ recombination is expected to peak 36 meV below the E1-HH1 free particle transition. After allowing for an exciton binding energy of 9 meV, 12 the e-A⁰ peak is expected 27 meV below $E1-HH1$, in good agreement with the energy difference $(28$ meV) between the peak of the sharp-line intensity distribution and $E1-HH1$ (Figs. 2 and 3). This correspondence of energy provides clear evidence for the involvement of acceptors in the sharp-line spectra. A major contribution to the energy range of the spectra then very likely arises from the variation of hole-binding energy (E_A) with position in the well. The decrease in E_A from 36 to 21 meV from the center to the edge of the QW is able to account for the occurrence of sharp-line recombination in the range \sim 1.585–1.600 eV.

It is likely that a large number of defect states contributes to each line for these large area samples, in contrast with the PL of Ref. 3 where $1-\mu m$ areas were investigated. The number of states contributing to each line can be estimated from the increment in current (ΔI) leading to excitation of one line. For example, the line at 1.59 eV is excited to its full intensity for a ΔI of 1.2 μ A, as the bias is increased from 1.608 to 1.616 V. Assuming all the current is radiative, the number of carriers *p* injected into the well for the change of current ΔI can be obtained from $p = \delta I \tau_R / e$, where τ_R (~1) ns) is the radiative recombination time. For $\Delta I = 1.2$ μ A, a

FIG. 4. Electroluminescence spectra for sample B at biases of 1.59, 1.60, and 1.61 V. The structure in the range from 1.55 to 1.56 eV, attributed to acceptor pairs, is stronger and less well resolved relative to the single acceptor 1.575 to 1.585 eV structure than in sample A, consistent with the higher expected acceptor density.

value for *p* of \sim 7.5 \times 10³ is obtained. Although this estimate is rough, and may be an overestimate since recombination also occurs in the GaAs contacts, it shows that any particular line almost certainly arises from the excitation of many centers.

In spite of the large number of centers contributing to each line, we propose that sharp lines are nevertheless observed since the recombination energies from the many Be acceptors on a given (100) plane are the same as a result of the well-defined acceptor energies for the lattice sites from well center to well edge.⁷ This argument assumes that the acceptors are isolated; acceptor interaction effects, which would perturb the energies, are highly unlikely given the acceptor density⁶ (N_A) in the well of $\sim 10^9$ cm⁻², and consequent average separation >1000 Å.¹³ For a 66-Å well there are 23 distinct (100) Ga planes (the degeneracy of the left and right halves of the QW will be lifted by the electric field), reasonably consistent with the number (~ 15) of the most prominent sharp lines. Furthermore, the number of centers contributing to each line $(p=7.5\times10^3)$ is close to the number of acceptors (m_A) per plane in the QW. For a 200- μ m device, a QW of 23 planes, and $N_A = 10^9$ cm⁻², $m_A = 1.3 \times 10^4$, close to the value of *p*, supporting strongly the model for the origin of the individual lines as arising from large numbers of acceptor impurities on particular monolayer planes.

To investigate further the origin of the lines, magneticfield (parallel to the current) studies were performed. Results for some of the prominent lines are shown in the inset to Fig. 3. All lines exhibit similar behavior, with a diamagnetic shift of \sim 7 meV from 0 to 14 T. This demonstrates that both the

hole *and* the electron in the recombination are localized, since if free electrons were involved a shift to higher energy of $1/2\hbar\omega_c = 11.6$ meV at 14 T would be expected. The electron localization most likely occurs at interface potential fluctuations.³ A two-monolayer fluctuation in width corresponds to \sim 10-meV change in energy, and provides the additional localization energy to account for the sharp lines in the range \sim 1.575–1.585 eV. At the relatively low levels of electron injection ($\sim 10^7$ cm⁻² or less at the HH1/LH1 *hole* resonances), and at the low experimental temperatures, electrons are only likely to populate the lowest energy states available and thus do not contribute significant extra broadening to the spectra. As the current is increased the spectra do broaden as mentioned above $(Fig. 3)$; this may arise from population of a wider range of electron states or from carriercarrier interaction effects, as discussed in Ref. 14.

The above model accounts semiquantitatively for the dominant features in the spectra. However, there are sharp lines from 1.56 to 1.57 eV that cannot be explained by this model. Their most likely origin is the recombination of holes localized at acceptor pairs. The average separation between acceptors is large $(>1000 \text{ Å})$, and so for a random distribution the formation of a significant density of interacting pairs is highly unlikely. However, preferential pairing of acceptors during MBE growth of GaAs thick layers has been reported previously in sharp-line PL spectroscopy.15,16 The hole binding energy to these centers was \sim 45 meV [compared to 27 meV (Ref. 17) for isolated acceptors]. This increase in E_A between isolated and paired acceptors in GaAs is qualitatively consistent with the increase in E_A deduced from the energies of the 1.585 eV (38 meV) and 1.565 eV (58 meV) peaks in intensity, supporting the pair attribution. The involvement of acceptor complexes in the lower energy spectra is supported by the results in Fig. 4 for sample B that has 150-Å spacer as opposed to 300 Å in sample A. Qualitatively similar spectra to Fig. 2 are found, but with the lower energy lines at 1.55–1.56 eV of greater intensity than the group peaking at \sim 1.58 eV. This is consistent with the attribution to pairs of the 1.55–1.56 eV features, for this sample of smaller spacer thickness and hence higher expected acceptor concentration.

In conclusion sharp-line spectra have been reported in GaAs-AlAs DBRTS, grown under conditions that favor acceptor incorporation in the QW active regions. The spectra have been attributed to the recombination of holes bound to acceptors located at positions varying from well center to well edge, with electrons also localized within the well. Each sharp line has been shown to arise from a large number of acceptors on a particular plane. Lower energy spectra have been ascribed to acceptor pairs.

- ¹C. Weisbuch, R. Dingle, A. C. Gossard, and W. Wiegmann, Solid State Commun. 38, 709 (1981).
- 2 K. Brunner, U. Bockelmann, G. Abstreiter, M. Walther, G. Böhm, G. Tränkle, and G. Weimann, Phys. Rev. Lett. 69, 3216 $(1992).$
- $3A$. Zrenner, L. V. Butov, M. Hagn, G. Abstreiter, G. Böhm, and G. Weimann, Phys. Rev. Lett. **72**, 3382 (1994).
- 4D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Phys. Rev. Lett. **76**, 3005 (1996).
- ⁵R. C. Miller, A. C. Gossard, W. T. Tsang, and O. Munteanu, Phys. Rev. B 25, 3871 (1982).
- 6H. B. Evans, L. Eaves, and M. Henini, Semicond. Sci. Technol. **9**, 555 (1994).
- ⁷G. Bastard, Phys. Rev. B **24**, 4714 (1981) .
- ⁸P. M. Martin *et al.*, Semicond. Sci. Technol. 7, B456 (1992).
- ⁹Electron tunneling below the E1 resonance may occur into disorder-induced tail states in the QW, or as a result of excitonic interactions between emitter electrons and holes in the well. See H. Buhmann et al., Solid-State Electron. 37, 973 (1994); A. Nogaret *et al.*, in *Proceedings of the 23rd International Conference on the Physics of Semiconductors, Berlin, 1996,* edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore, 1996), p. 2059.
- 10 The identification of E1-HH1 has been established from photoluminescence excitation studies on these samples, see H. B. Evans, L. Eaves, C. R. H. White, M. Henini, P. D. Buckle, T. A. Fisher, D. J. Mowbray, M. S. Skolnick, and G. Hill, Surf. Sci. **305**, 387 (1994).
- 11A. Pasquarello, L. C. Andreani, and R. Buczko, Phys. Rev. B **40**, 5602 (1989).
- 12P. Dawson, K. J. Moore, G. Duggan, H. I. Ralph, and C. T. B. Foxon, Phys. Rev. B 34, 6007 (1986).
- ¹³The change in on-center acceptor energy for a one-monolayer change in well width is only ~ 0.4 meV (see Ref. 11), and thus only contributes an energy shift of the order of the observed linewidths, as opposed to >1 meV change in energy between acceptors on successive monolayer planes.
- 14R. Steffen, A. Forchel, T. L. Reinecke, T. Koch, M. Albrecht, J. Oshinowo, and F. Faller, Phys. Rev. B 54, 1510 (1996).
- 15M. S. Skolnick, D. P. Halliday, and C. W. Tu, Phys. Rev. B **38**, 4165 (1988).
- 16M. S. Skolnick, C. W. Tu, and T. D. Harris, Phys. Rev. B **33**, 8468 (1986).
- ¹⁷ D. J. Ashen, P. J. Dean, D. T. J. Hurle, J. B. Mullin, and A. M. White, J. Phys. Chem. Solids 36, 1041 (1975).