Photoinduced Space-Charge Buildup by Asymmetric Electron and Hole Tunneling in Coupled Quantum Wells

R. Sauer and K. Thonke
Physikalisches Institut, Universität Stuttgart, 7000 Stuttgart 80, Federal Republic of Germany

and

W. T. Tsang
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 8 March 1988)

We demonstrate, by photoluminescence measurements, the excitation-dependent opposite charging of coupled $In_{0.53}Ga_{0.47}As/InP$ single quantum wells. The formation of the dipole layer is associated with band bending lining up the electron levels in the wells. This novel effect is due to the very different tunneling rates of electrons and holes through the barrier and is observed for barrier widths $L_B = 40-100$ Å. On the basis of these findings we discuss a potential new optical bistability.

PACS numbers: 73.40.Kp, 78.55.Cr

Tunneling of electrons and holes is under intense current investigation in tailored two-dimensional semiconductor heterostructures because of academic relevancy as a basic quantum effect and wide potential applications. Coherent and incoherent, resonant, sequential, and Zener tunneling have been reported recently in double barriers, superlattices, and multiple quantum wells (QW's) of GaAs/AlGaAs, InGaAs/InP, and related compound semiconductors.[1,2] A variety of tunneling transport devices based on these effects have been suggested or demonstrated, such as high-gain photodetectors[3] and tunneling transistors.[4] Optical work on tunneling has employed photoluminescence (PL) and selective excitation to address, e.g., degeneracy splittings in resonantly coupled QW's,[5] Bloch transport of electrons and holes in superlattice minibands,[6] tunneling-assisted radiative recombination,[7] and "transferred" luminescence.[8]

This Letter reports a novel effect not dealt with in previous tunneling studies. Using optical measurements, we demonstrate photoinduced space-charge buildup on nonresonantly coupled quantum wells due to asymmetric electron and hole tunneling.[9] A set of samples is employed with two QW's per sample of $In_{0.53}Ga_{0.47}As$ lattice matched to InP, nominally 100 Å and 60 or 80 Å wide, coupled through InP barriers of various widths from 20 to 400 Å. We measure the PL intensity ratio I_1/I_2 (cf. Fig. 1) of the E_{1h} emission lines due to confined $n = 1$ electron to $n = 1$ heavy-hole recombination in the two wells as a function of the optical excitation level while keeping the sample temperature constant. This is complementary to a study on the same samples where tunneling is investigated by temperature-controlled PL excited with 514-nm laser light at a constant, low "standard" excitation power of 0.3 mW to avoid heating.[10] Here, with the samples immersed in liquid helium at 4.2 or 2 K, we find that the PL intensity ratio I_1/I_2 depends on the optical excitation level in a well defined and systematic manner. This is ascribed to space-charge buildup by asymmetric electron and hole tunneling bending the bands and shifting the energy levels of the wells so as to control the relative occupancy of the levels being spectroscopically monitored.

Let us first investigate the low-excitation case as schematically shown in Fig. 1(a). Samples with wide barriers exceeding ≃200 Å width exhibit E_{1h} emission lines of the two QW's with an intensity ratio I_1/I_2 on the order of unity. Reduction of the barrier width results, at constant excitation, in a successive intensity loss of the I_2

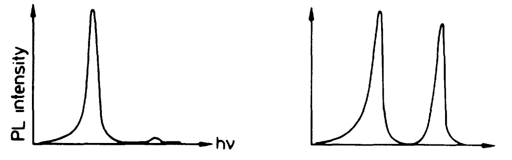

FIG. 1. Energy-band diagrams (top) and associated photoluminescence spectra (bottom) of the coupled QW1 and QW2 with $n = 1$ electron and heavy-hole states: (a) Low optical excitation with no band bending; (b) strong optical excitation with band bending lining up the electron energy levels. Electron tunneling is indicated by solid horizontal arrows, and less effective hole tunneling by dashed arrows.

line relative to I_1, and eventually, for $L_B = 20$ Å, the I_2 line is no longer detectable. In Fig. 2 we demonstrate directly by photoluminescence excitation (PLE) spectroscopy that the depletion of QW2 is due to electron and hole tunneling through the barriers. The sample with $L_B = 60$ Å, chosen in Fig. 2 as an example, exhibits in PL both the emission lines I_1 and I_2. The associated PLE spectrum, with luminescence-light detection at I_1, shows excited transitions E_{1l}, E_{2h}, and E_{3h} in QW1 involving $n = 1$, 2, and 3 electron and light- (l) or heavy- (h) hole subband levels. This portion of the PLE spectrum is identical to PLE spectra of correspondingly wide isolated QW's. The hatched peak gives evidence for tunneling from QW2 to QW1: Excess carriers resonantly excited in QW2 in the E_{1h} transition are transferred to QW1 through the barrier, yielding there, after relaxation to the lowest sublevels, recombination radiation in the detection channel. The intensity of the transferred peak, as a measure of the tunneling efficiency, correlates inversely with the barrier widths in different samples: The peak, absent for $L_B > 200$ Å, is weakly observed for $L_B = 100$ Å, increases for narrower barriers, and is by far the dominant PLE feature for $L_B = 20$ Å. Hence, particle transfer causes a depletion of QW2 and the related changes of the intensity ratio I_1/I_2 as stated.

Given this optical evidence for electron-hole tunneling in our samples we return to the anticipated space-charge buildup. Basically, the effect must occur since light electrons tunnel at much higher rates than heavy holes which tend to be localized in the barrier for $L_B \gtrsim 40$ Å. This has been experimentally shown by "effective mass filtering."[11,12] Very different tunneling rates are also expected from the familiar expression for the tunneling probability of a particle through a square-well potential barrier of height V_0, of the form $\exp[-(8m^* V_0/\hbar^2)^{1/2} L_B]$. Here, applicable mass values for $In_{0.53}Ga_{0.47}As$ are[13] $m_e^*/m_0 = 0.041$ and $m_{hh}^*/m_0 = 0.465$. Asymmetric tunneling of photogenerated excess carriers charges the wells with $n_1 - p_1 = p_2 - n_2$, where n and p are the steady-state electron and hole excess concentrations in the two wells. (In contrast, recombination alone would preserve charge neutrality in the wells.) At low excitation [Fig. 1(a)] the charge on the QW's is negligible, being related to the absolute excess carrier densities, although the carrier tunneling depopulates QW2 relative to QW1 and gives rise to strong I_1 and weak I_2 emission. However, at high excitation [Fig. 1(b)] the space-charge density $n_1 - p_1$ becomes significant, causing a bending of the bands which finally lines up the electron energy levels and separates the hole levels farther. In this case there is essentially no depletion of QW2 under steady-state excitation conditions if the hole tunneling rate is small compared to the recombination rate, $R_p \ll \tau^{-1}$. Consequently, comparable line intensities I_1 and I_2 are expected, as if the wells were decoupled. As a function of the excitation level, the ratio I_1/I_2 will start at a high value for low excitation levels and drop to values on the order of 1 when the pump level is sufficiently increased.

This behavior is in fact observed. Experimental data are shown in Fig. 3 for five samples with barrier widths as indicated and QW widths close to 100 Å (QW1) and 60 or 80 Å (QW2). For $L_B = 400$ Å the wells are entirely decoupled with no influence of the pump level on the relative line intensities. Smaller barrier widths cause changes in the intensity ratio which increase drastically and in succession for barriers narrower than 100 Å. In

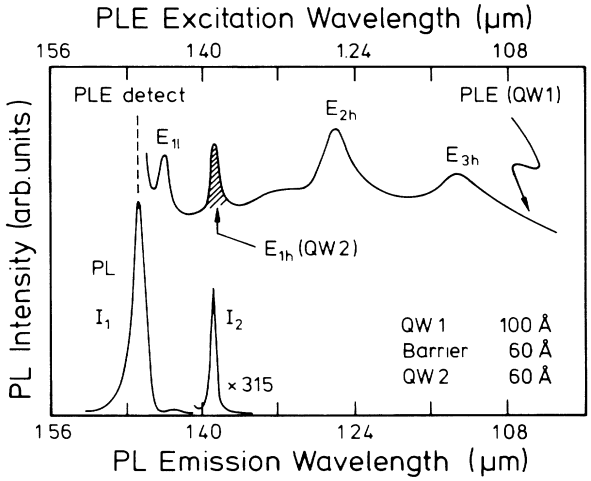

FIG. 2. Optical detection of the electron-hole tunneling. Lower curve: PL spectrum at low excitation density ($\simeq 10$ mW/cm^2). Upper curve: PLE spectrum of QW1 taken with halogen-lamp-monochromator excitation showing the peak E_{1h} (hatched) due to electrons and holes transferred from QW2. Measurement temperature is 4.2 K.

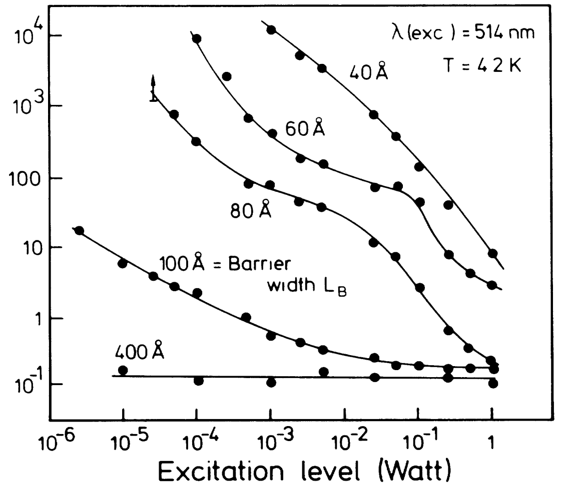

FIG. 3. Intensity ratio I_1/I_2 of the two PL lines vs excitation level for various barrier widths. For $L_B = 20$ Å the ratio is indefinitely large as the I_2 emission cannot be observed at any pump level. The curve for $L_B = 400$ Å was shifted down by a factor of 10 for clarity. Laser output of 1 W corresponds to an excitation density on the sample of $\simeq 200$ W/cm^2.

the high-excitation regime, all curves tend to a small intensity ratio as predicted by the space-charge concept. Final asymptotic values are not reached throughout with our present maximum laser output power of 1.5 W, and are in principle hard to obtain because of sample heating at even higher optical powers. The final values, though expected to be in the range of unity, are actually still subject to the small hole tunneling rates (which tend to lift up the curves), to different radiative quantum efficiences of the two wells per sample, and to different optically active well widths. For very low excitation levels, constant, asymptotic intensity ratios are not reached since the I_2 emission falls below the detection noise level, thus limiting our experimentally accessible range.

Convincing support is lent to the space-charge concept by the observation that three samples all with nominal barrier widths $L_B = 20$ Å do not show any indication of the I_2 line even at hard optical pumping: For barriers that narrow, electrons and holes tunnel efficiently[10,14] with rates R_e and R_p, respectively, which are both much larger than the competing recombination rates, $R_e \gg \tau^{-1}$ and $R_p \gg \tau^{-1}$; hence, QW2 is totally depleted of electrons and holes within their recombination lifetime so that charge neutrality in the QW's is maintained and no dipole layer can be generated. Estimates of the ratio R_e/R_p for $L_B = 20$ Å calculated with band offsets[15] $\Delta E_c{:}\Delta E_v = 0.4{:}0.6$ and the given QW widths using the earlier mentioned expression for the tunnel probability yield a value of ~600. The corresponding value for the next wider well in our series of samples, $L_B = 40$ Å, would be the square, 3.6×10^5, making plausible the rather sudden onset of the charging effect for $L_B \geq 40$ Å. We note that results equivalent to those shown in Fig. 3 were obtained with excitation by green (514 nm) or red (647 nm) laser light and for nominal sample temperatures of 4.2 or 2 K. For $T = 2$ K, the "humps" on the curves are virtually flattened out, resulting in simple decay curves. This indicates that the humps are artifacts not related to the charging effect but possibly due to unintentional impurity doping of the wells. In fact, the 60- and 80-Å barrier samples show, up to medium excitation, a weak band between the I_1 and I_2 lines which saturates with increasing excitation and shifts to higher energies consistent with donor-acceptor pair recombination.

In the following we discuss three effects that might arise in conjunction with space charge and high optical powers:

(1) *Saturation.*—To make sure that the behavior of the QW luminescence in Fig. 3 does not originate in any kind of PL saturation phenomena we have measured the excitation dependence of the I_1 line for all samples employed. The dependence is nearly perfectly linear in all cases over 6 orders of magnitude corresponding to the range in Fig. 3.

(2) *Stark shift.*—The electric field $\mathcal{E}$ related to the dipole layer of a QW structure should cause a Stark shift of the PL lines. Experimentally, a shift is not observed and, hence, is below the detection limit of $\simeq 2$ meV given by the optical linewidths. By application of Poisson's equation the relative shift of the electron energy levels is related to the maximum field as $\delta(\Delta E_e) = \frac{1}{2}(L_1 + 2L_B + L_2)\mathcal{E}$, where $\mathcal{E} = -(n_1 - p_1)q/\epsilon\epsilon_0$ and the L's are the widths of the QW1, the barrier, and QW2, respectively. The shift necessary to line up the electron energy levels is $\simeq 40$ meV for our QW structures, so that $\mathcal{E}_{\max} = 3 \times 10^4$ V/cm. Stark shifts of InGaAs/InP QW's have been studied in externally applied electric fields,[16] and for this $\mathcal{E}_{\max}$ the associated shift of the E_{1h} line has been found to be $\simeq 3$ meV. Realistically, the field is $\mathcal{E}_{\max}$ only at the interfaces between the barrier and the wells, and an average field extending over the region of maximum probability density of the particles in the wells would cause undetectably small shifts.

(3) *Band filling.*—As a result of the constant limited density of states per energy interval in the quasi-two-dimensional wells, $D = m^* A/\pi\hbar^2$, band filling with associated PL line shifts and broadenings could happen. We have estimated the shift for the highest powers experimentally available, taking 1 ns as a representative recombination lifetime and an excitation area $A = 0.25$ mm^2 to calculate approximate steady-state excess carrier densities. For these data, the band filling amounts to no more than $\simeq 10^{-2}$ meV.

The present discussion relies on qualitative arguments to show that all experimental findings agree with the space-charge concept. In addition we note briefly without presenting details that the observed characteristic trends can be modeled by rate equations. These involve the kinetic parameters of carrier generation and recombination in the QW's, and tunneling between the wells either directly (QW2 → QW1) or activated via the thresholds ΔE_e or ΔE_h [cf. Fig. 1(a)] for the reverse tunneling direction. The effective thresholds, by virtue of Poisson's equation, are subject to the space charge $n_1 - p_1$, representing a feedback which causes the changes of I_1/I_2 as a function of excitation. Details of this analysis will be published elsewhere.

Finally, we speculate on a new optical bistability based on the present data. As the two QW's per sample in our study have different widths and quantum energies of the confined particles we are concerned with nonresonant tunneling. This is basically a much less efficient process than resonant tunneling since in the two-dimensional energy dispersion along the layers phonon participation is needed in the tunneling process to ensure conservation of the parallel momentum $k_\parallel$. When intense optical pumping causes the electron states to become energetically degenerate within their level widths, a switch-over to resonant tunneling will occur. The resonantly enhanced tunneling rates reinforce the lining up of the electron levels and tend to maintain resonance for lower pump levels than were initially needed at the onset of the resonance. The process represents a hysteresis cycle in Fig. 3, following a particular excitation curve, changing over to a curve with fictitiously much narrower barrier, and finally jumping down to the original curve. The bistability should be effective even at high temperatures if the well widths are sufficiently different to yield large energy separations ΔE_e and ΔE_h.

In conclusion, we have given evidence from photoluminescence measurements for asymmetric electron and hole tunneling between coupled single QW's. The asymmetry causes the formation of a dipole layer which, at appropriate barrier widths of 40–100 Å, lines up the electron energy levels and controls the relative radiative-recombination intensities. This effect could also lead to a nonresonant-resonant tunneling switch-over.

The authors thank H. L. Stormer for a helpful discussion of the QW charging effect. The financial support of the Sonderforschungsbereich 329 of the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

---

[1] See the reviews by F. Capasso, K. M. Mohammed, and A. Y. Cho, IEEE J. Quantum Electron. **22**, 1853 (1986); E. Esaki, IEEE J. Quantum Electron. **22**, 1611 (1986).

[2] J. Allams, F. Beltram, F. Capasso, and A. Y. Cho, Appl. Phys. Lett. **51**, 575 (1987).

[3] F. Capasso, K. Mohammed, A. Y. Cho, R. Hull, and A. L. Hutchinson, Phys. Rev. Lett. **55**, 1152 (1985).

[4] F. Capasso and R. A. Kiehl, J. Appl. Phys. **58**, 1366 (1985).

[5] H. Kawai, J. Kaneko, and N. Watanbe, J. Appl. Phys. **58**, 1263 (1985).

[6] B. Deveaud, J. Shah, T. C. Damen, B. Lambert, and A. Regreny, Phys. Rev. Lett. **58**, 2582 (1987).

[7] D. F. Nelson, R. C. Miller, D. A. Kleinman, and A. C. Gossard, Phys. Rev. B **34**, 8671 (1986).

[8] B. A. Wilson, R. C. Miller, S. K. Sputz, T. D. Harris, R. Sauer, M. G. Lamont, C. W. Tu, and R. F. Kopf, in *Gallium Arsenide and Related Compounds—1986,* edited by W. T. Lindley, IOP Conference Proceedings No. 83 (Institute of Physics, Bristol, United Kingdom, 1987), p. 215.

[9] Space-charge buildup in a different context, double-barrier resonant tunneling, has recently been shown to be important for an explanation of the experimental current-voltage curves: V. J. Goldman, D. C. Tsui, and J. E. Cunningham, Phys. Rev. B **35**, 9387 (1987).

[10] R. Sauer, T. D. Harris, and W. T. Tsang, to be published.

[11] F. Capasso, K. Mohammed, A. Y. Cho, R. Hull, and A. L. Hutchinson, Appl. Phys. Lett. **47**, 420 (1985).

[12] D. V. Lang, A. M. Sergent, M. B. Panish, and H. Temkin, Appl. Phys. Lett. **49**, 812 (1986).

[13] K. Alavi, R. L. Aggarwal, and S. H. Groves, Phys. Rev. B **21**, 1311 (1980).

[14] R. Sauer, T. D. Harris, and W. T. Tsang, Surf. Sci. **196**, 388 (1988).

[15] D. V. Lang, M. B. Panish, F. Capasso, J. Allam, R. A. Hamm, A. M. Sergent, and W. T. Tsang, Appl. Phys. Lett. **50**, 736 (1987).

[16] I. Bar-Joseph, C. Klingshirn, D. A. B. Miller, D. S. Chemla, U. Koren, and B. I. Miller, Appl. Phys. Lett. **50**, 1010 (1987).