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Charge-transfer-state photoluminescence in asymmetric coupled quantum wells

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We have performed continuous and time-resolved photoluminescence experiments on novel double-quantum-well structures in Schottky diodes. We have directly observed the buildup of a charge-transfer (CT) state in which the electrons and holes are in separate wells because of the fact that they tunnel in opposite directions. We have studied the effect of an electric field on the CT state formation, and have observed a strong, linear Stark shift of the CT luminescence.

Tunneling in semiconductor quantum-well (QW) structures has been a subject of intense study for some time; however, only recently have time-domain techniques been applied to directly observe tunneling. Time-resolved photoluminescence (PL) has been used to determine the tunneling escape rate of electrons from a single QW through a thin barrier into a continuum,[1] and to determine the electric field dependence of this tunneling rate.[2,3] Tunneling between coupled QW's is of particular interest; time-resolved[4] and time-integrated PL (Ref. 5) and optical absorption[6] have been used to observe tunneling in double-QW structures. Time-integrated PL (Ref. 7) and PL excitation spectroscopy[8] have also been used to observe spatially indirect optical transitions in narrow-barrier structures. In this Rapid Communication we report the direct observation of the buildup of a "charge-transfer" (CT) state via electron and hole tunneling in opposite directions in $GaAs/Al_xGa_{1-x}As$ double-QW structures.

The double-QW structures used in this study are of a novel design, where the two QW's are of different widths, but the Al composition of the wider QW is adjusted so that under flat-band conditions the electron energy levels of the two wells are very close to resonance, and the hole energies are sufficiently different so that the PL energies of the two wells separated.[9,10] A 58-Å $Al_{0.15}Ga_{0.85}As$ QW (QW1) is coupled to a 26-Å GaAs QW (QW2) through an $Al_{0.45}Ga_{0.55}As$ barrier. Two samples were grown for this study: Sample *A* has a thin (43-Å) barrier so the electron states are somewhat delocalized over the two wells, and sample *B* has a thick (86-Å) barrier so the electron states are strongly localized in each QW. Twenty-five periods of the double-QW structure were grown on an n^+-type GaAs substrate and n^+-type GaAs buffer layer. A semitransparent Al Schottky contact was evaporated on the top surface so that the effect of an electric field applied along the growth direction could be studied. The calculated band diagram and electron and hole states for sample *A* are shown in Fig. 1 for both flat-band and reverse-bias conditions.

The samples were held in a cryostat at a temperature of 6 K. Photoexcitation was provided by picosecond pulses from a synchronously pumped dye (Pyridine 1) laser. We estimate that each laser pulse injected a carrier density of approximately 10^{11} cm^{-2} in each well. The resulting PL was collected by a lens and dispersed by a 0.32-m monochromator with 300 l/mm grating across the entrance slit of a Hamamatsu synchroscan streak camera operating at 100 MHz. The streaked image was integrated on a two-dimensional silicon-intensified target (SIT) detector, allowing us to obtain time-resolved PL spectra with a time resolution of 20 ps and a spectral resolution of about 3 meV. Time-integrated (CW) PL spectra were obtained using a separate system with sub-meV resolution.

CW PL spectra are shown for both samples in Fig. 2. For sample *B* the ω_1 and ω_2 transitions are about the same magnitude, but for sample *A*, the ω_1 intensity is much smaller than for ω_2, indicating stronger tunneling processes for this sample, as will be discussed below.

Time-resolved spectra for sample *B* (thick barrier) are shown in Fig. 3 for 0 and -8 V bias. The spectra show the scattered pump light (defining the time origin) and two PL lines corresponding to recombination within each of the two wells. The high-energy PL line corresponds to the QW1 transition, and the lower-energy line to QW2. The PL rise and decay times change very little with applied bias, and the Stark shift of the PL peaks is observed to be a few meV to the red as expected.[11–13] In the CW PL spectra shown in Fig. 4, however, we observe a third PL peak which shifts strongly and approximately linearly towards the red with applied bias. In the narrow barrier sample (*A*) this peak appears even more clearly in the CW spectra and presents a stronger Stark shift. The origin of this peak can be elucidated by examining the time-resolved PL spectra for the thin barrier sample; the line comes from radiative recombination between electrons in QW1 and holes in QW2, which we refer to as the charge-transfer (CT) state.

A selection of the time-resolved PL spectra versus ap-

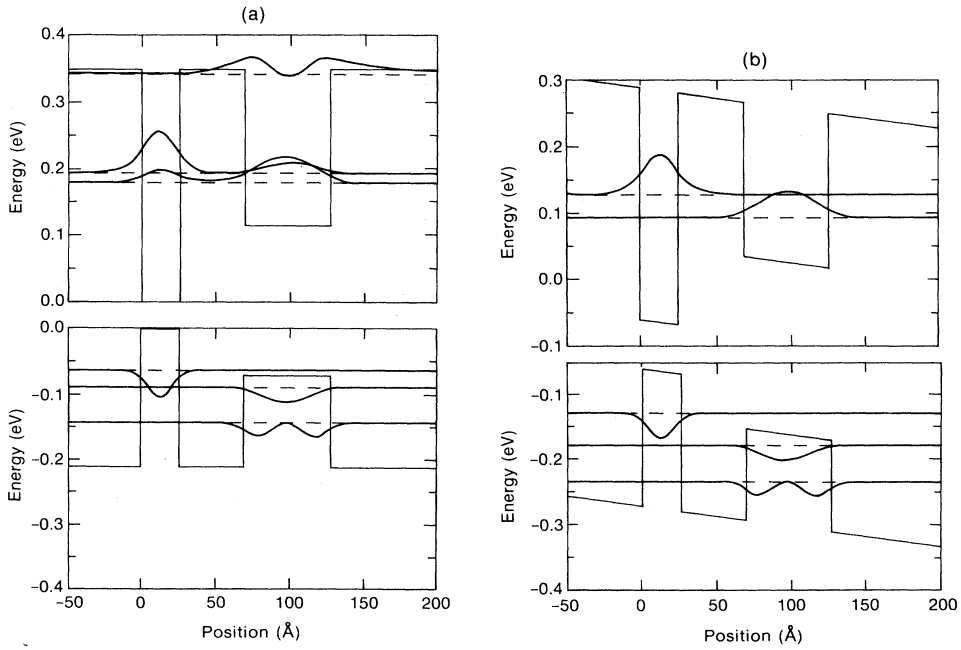

FIG. 1. Calculated band diagrams and electron and heavy-hole wave functions for sample A: (a) under flat-band conditions and (b) with an electric field of 30 kV/cm. The results for sample B are similar, but the electron states are always strongly localized in each well.

plied bias voltage is shown in Fig. 5 for sample A. At low bias one sees the scattered pump light and two PL lines corresponding to the ω_1 and $\omega_2+\omega_{CT}$ transitions. It should be noted that at resonance the PL of QW2 and the CT state have the same energy. As the electric field is increased, the ω_2 and ω_{CT} lines separate and ω_{CT} is strongly Stark shifted to lower energy. This PL has a long lifetime that, in fact, approaches or exceeds the 10-ns time interval between pump pulses and the synchroscan sweep cycle time; hence the PL signal that appears roughly constant versus time on the streaked spectrum. The Stark shift of this long-lived PL component is approximately linear with applied bias. The strong red shift and long decay time indicate that this PL is, in fact, the result of recombination

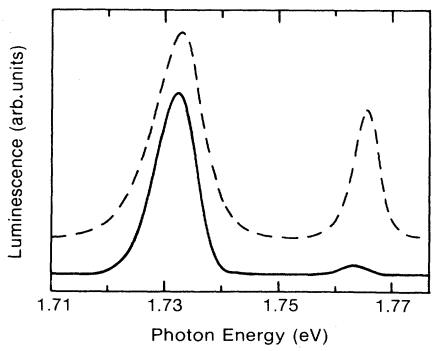

FIG. 2. Time-integrated PL spectra for sample A (solid line) and sample B (dashed line), with no applied bias voltage. The high-energy peak corresponds to the QW1 transition; the lower-energy peak to QW2.

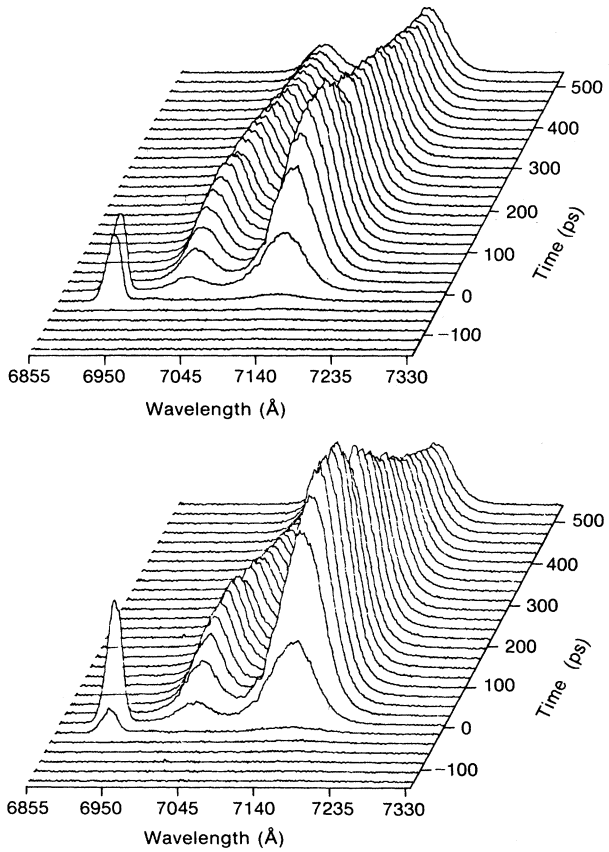

FIG. 3. Time-resolved PL spectra for sample B: (a) with no applied bias and (b) with -8 V applied bias.

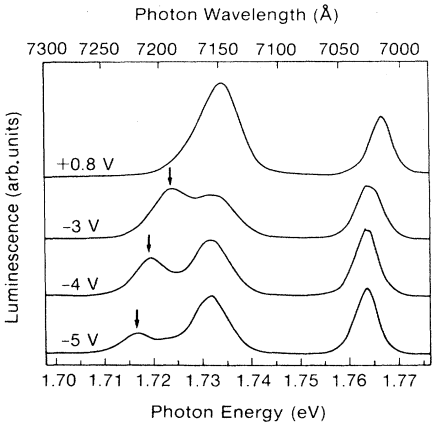

FIG. 4. Time-integrated PL spectra for sample B at various bias voltages.

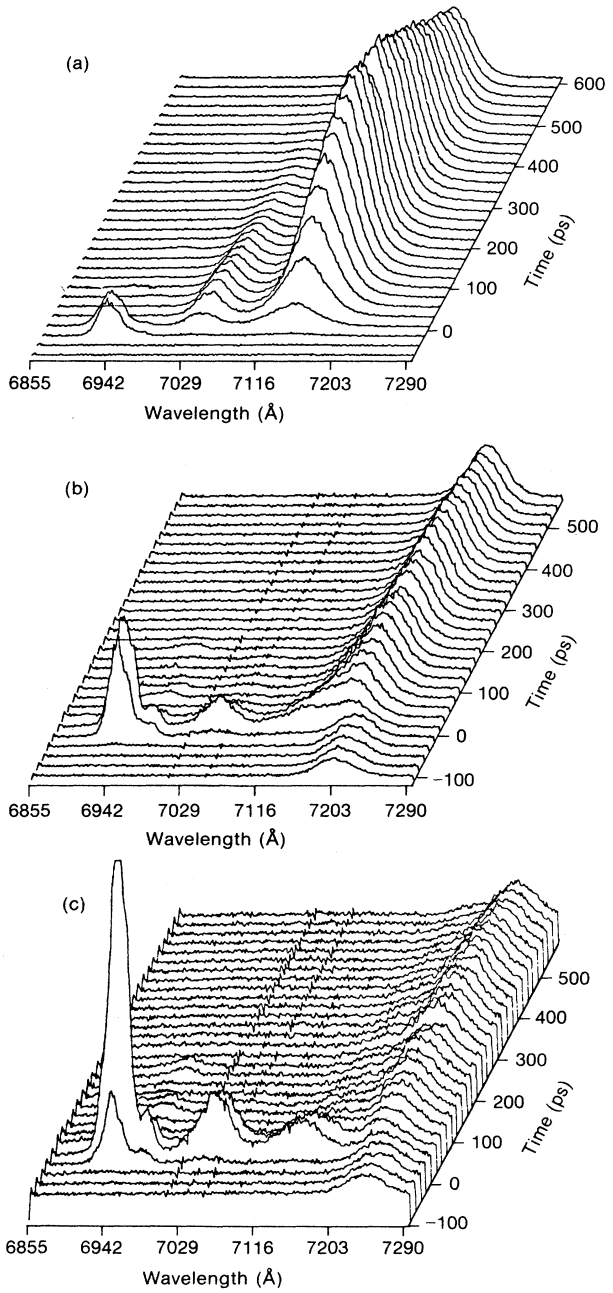

FIG. 5. Time-resolved PL spectra for sample A: (a) with 0 V, (b) -3 V, and (c) -4.75 V applied bias.

of electrons and holes in the CT state. The CT state is built up by electrons tunneling from QW2 to QW1 and holes tunneling from QW1 to QW2. The long lifetime is the result of the small overlap of the electron and hole wave functions, but the CT luminescence is nevertheless observable due to the nearly complete charge separation that takes place. This explains why the CT luminescence is observable in the CW PL spectra but not in the time-resolved spectra for sample B. In this case the charge separation is not so strong due to the much slower electron and hole tunneling rates between the wells, so although the long-lived radiative recombination is observed as a line in the CW Pl spectrum, the instantaneous intensity is low and therefore extremely weak on the time-resolved spectrum.

One further observation confirms that the origin of the strongly red-shifted PL line is from the CT state. The Stark shift of this line is dynamic, since as the carriers recombine, the screening of the applied electric field by the CT state is reduced and the red shift increases. This is most easily observed by comparing the PL wavelength just after the pump pulse with the CT PL wavelength at a very long delay time (which in practice is done by observing the PL at "$t < 0$" on the synchroscan streak image).

We mentioned previously that the CT state has a lifetime that can exceed the pump pulse period. As a consequence a dc charge can build up in the wells that screen the applied field. The separated charge density can be estimated by comparing the observed CT PL Stark shift (which gives the average electric field in the double-QW region) to the calculated electric field in the Schottky diode without the presence of charge carriers. We can then estimate an effective lifetime τ for the separated carriers from $n = g\tau$ where g is the generation rate. We find values for τ which increase with applied field, which is expected because the electron-hole separation increases with the field. We also find, however, that τ depends strongly on the pump intensity. For example, for sample B at -3 V bias and 9 W/cm^2 pump power, the lifetime τ is 2 μs. For sample A at -3 V bias and 3 W/cm^2 pump power, τ is 1.2 μs, but at 300 W/cm^2 τ is 20 ns. A more refined analysis treating the charged QW's in a self-consistent manner will be required to elucidate the processes that determine the lifetime, which is not necessarily the radiative lifetime for the CT transition.

We finally note that the ω_1 transition strength in both the time-resolved and cw experiments for sample A is surprisingly small. Two possible explanations exist: (i) The ω_1 transition is in $Al_{0.15}Ga_{0.85}As$ and, thus, the nonradiative transition rate is so fast that the electrons and holes recombine faster than the transfer time of electrons

from QW2, or (ii) the hole transfer rate from QW1 to QW2 is faster than the streak camera resolution. The latter explanation would require a hole tunneling rate orders of magnitude greater than one would expect from the simple theory of tunneling from a QW into a continuum. On the other hand, the first explanation is not quite satisfactory either, since the ω_1 transition in sample *B* grown under the same conditions is strong.

In summary, we have performed time-resolved and CW PL spectroscopy on a novel double-QW structure, and have directly observed luminescence from a charge-transfer state, where the charge separation occurs by electron and hole tunneling in opposite directions induced by an electric field.

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