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Luminescence of *n*-*i*-*p*-*i* heterostructures

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The luminescence properties of a new superlattice structure, called a *n*-*i*-*p*-*i* heterostructure, are described. The samples consist of GaAs quantum wells placed between alternately doped $Al_{1-x}Ga_xAs$ layers. The luminescence transitions between the quantized GaAs subbands are greatly influenced by the excitation-intensity-dependent electrostatic potential. The lowest subband luminescence transition agrees well with predictions for the ideal structure. In addition, distinct luminescence transitions from higher subbands are observed.

The interest in semiconductor superlattices has focused on two types of structures. One is a heterostructure, made from alternating different materials, such as a GaAs and $Al_{1-x}Ga_xAs$.[1] Rectangular potential wells are formed which confine carriers into quantized states. The potential wells for electrons and holes usually coincide spatially, so that optical excitation and recombination processes retain the excitonic character of the bulk material, but are modified by the two-dimensional confinement. The second type of superlattice is the *n*-*i*-*p*-*i* structure, formed by periodic *n*- and *p*-type doping, possibly separated by intrinsic layers, in an otherwise uniform semiconductor, resulting in a spatial modulation in the energy of the bands.[2] Optically excited electrons and holes are separated by the internal electrostatic potential so that recombination occurs by tunnelling and is typically very slow. These structures have the added characteristic that the excited carriers modify the electrostatic potential and change the effective band gap.

In this paper, we present luminescence data on a new superlattice structure that is a combination of both types.[3] A *n*-*i*-*p*-*i* structure is made by molecular-beam epitaxy (MBE), in which the doped layers are $Al_{1-x}Ga_xAs$ and the undoped layers are GaAs. The band diagram is shown in Fig. 1. The doped layers are 50 Å thick with a doping level of 3×10^{18} cm^{-3}. In addition, there are undoped $Al_{1-x}Ga_xAs$ spacer layers to keep the donors and acceptors away from the GaAs layer, a technique commonly used in heterostructure superlattices in order to suppress impurity scattering and to reduce impurity induced level broadening of the subband states in the GaAs quantum wells.[4] The *n*-*i*-*p*-*i* structure results in tilted, triangular wells, with electrons and holes spatially separated. This structure has a considerable advantage over a conventional *n*-*i*-*p*-*i* structure since the carriers are confined within the undoped region. Electrons and holes therefore occupy only weakly broadened subband states, whereas in conventional *n*-*i*-*p*-*i* structures, holes populate the acceptor impurity band. As a result, the luminescence lines more clearly reflect the occupancy of the quantized states, and, in addition, transitions from higher-energy subbands can be resolved.

Figure 2 shows luminescence spectra at different excitation intensities for one of the samples. The lowest energy luminescence band has the characteristic *n*-*i*-*p*-*i* property of an increase in energy with higher excitation intensity. The transition is between the lowest electron and heavy-hole subbands, which we denote as $(0,h0)$. As the excitation intensity varies, the linewidth increases from 15 to 40

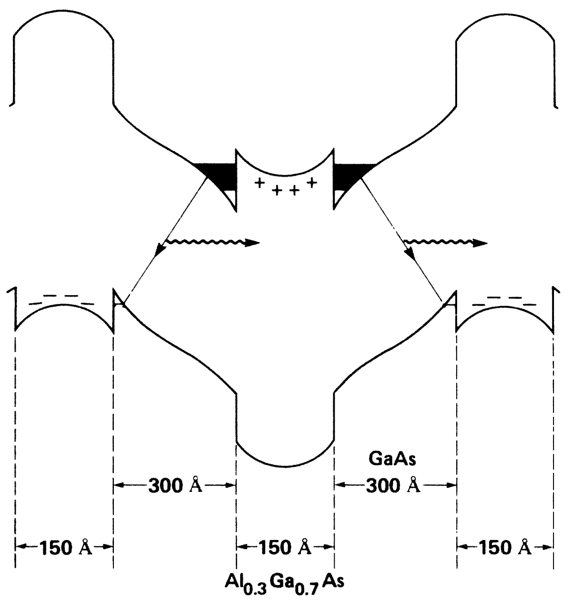

FIG. 1. Schematic band diagram of the *n*-*i*-*p*-*i* heterostructure design showing the tilted quantum well structure. Each $Ga_{1-x}Al_xAs$ layer consists of a 50-Å doped region and two 50-Å spacer layers. The expected luminescence transitions are indicated.

meV. A self-consistent calculation for the structure yields an effective band gap E_g^{eff}, which is approximately linear in the excited two-dimensional carrier concentration $n^{(2)}$,

$$E_g^{\text{eff},n}=E_g^{\text{eff},0}+\beta n^{(2)}$$

where $\beta=1.5\times10^{-13}$ cm^2 eV, and $E_g^{\text{eff},0}=1.32$ eV. The calculations assume that 60% of the band offset is in the conduction band. For an ideal *n-i-p-i* heterostructure with only the lowest subbands populated, the line shape should be approximately rectangular with a bandwidth W equal to the sum of the electron and heavy-hole (two-dimensional) Fermi energies. W is therefore proportional to $n^{(2)}$, so that the luminescence peak position is given by

$$\begin{aligned} h\nu^{\text{peak}} &= E_g^{\text{eff},n}+W/2 \\ &= E_g^{\text{eff},0}+(\text{const})W\ . \end{aligned}$$

This linear relation between linewidth and peak position is confirmed by the data of Fig. 2. The extrapolated value of $E_g^{\text{eff},0}$ is 1.34 eV which is within 20 meV of the predicted value for the structure. However, the line is not rectangular, and clearly there are additional broadening terms of unidentified origin. Furthermore, W is larger than the calculated value by about 50%. Nevertheless, the results show that dominant contribution to the line shape is the Fermi energy position. Narrower lines are observed in time-resolved luminescence spectra, which come even closer to the ideal behavior.[5]

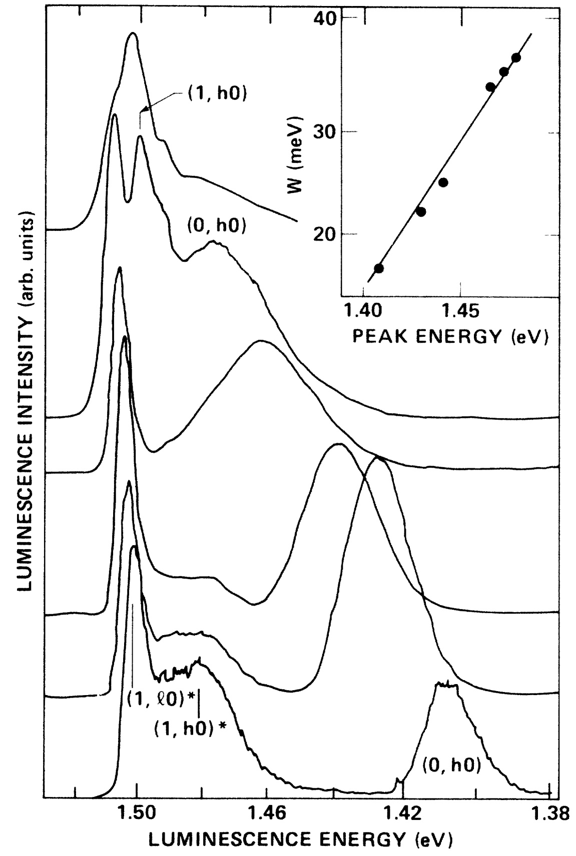

FIG. 2. Series of 1.7 K luminescence spectra taken at increasing excitation intensity ranging from approximately 5 mW/cm^2 to 30 W/cm^2 at an excitation wavelength of 1.915 eV (with the highest intensities at the top of the figure). The identification of the peaks is discussed in the text. The insert shows the relation between the linewidth and peak energy of the lowest energy transition.

One characteristic feature predicted for *n-i-p-i* luminescence is that as the recombination proceeds, the effective band gap $E_g^{\text{eff},n}$ decreases, because the electrostatic potential increases as the carriers recombine. A shift of the spectrum to low energy therefore occurs during the decay. Time-dependent energy shifts are often observed in luminescence of semiconductors, when the upper side of an emission band has a faster decay than the lower side (e.g., donor-acceptor pairs). The *n-i-p-i* case is qualitatively different since the entire peak moves in energy. Conclusive evidence for this behavior can be sought in measurements of luminescence decay, spectrally resolved from the low-energy side of peak. After the excitation is turned off, the luminescence intensity is predicted to increase as the spectrum moves down into the energy of the observations. Figure 3 shows examples of this unusual behavior, which has not been previously observed because of large inhomogeneous broadening in conventional *n-i-p-i* structure.[6] There is an increase of intensity of about a factor of 2 after the excitation pulse, and a subsequent decay time constant of about 50 μsec, characteristic of tunneling recombination. The decay is much more rapid at high energy, and corresponds to the luminescence peak moving away from the observation energy.

Another new feature of the *n-i-p-i* heterostructure luminescence is the presence of higher-energy transitions. In the low-intensity spectra of Fig. 2, there is a sharp peak near 1.50 eV and a broader line near 1.48 eV. The 1.50-eV line moves up by about 10 meV with increasing excita-

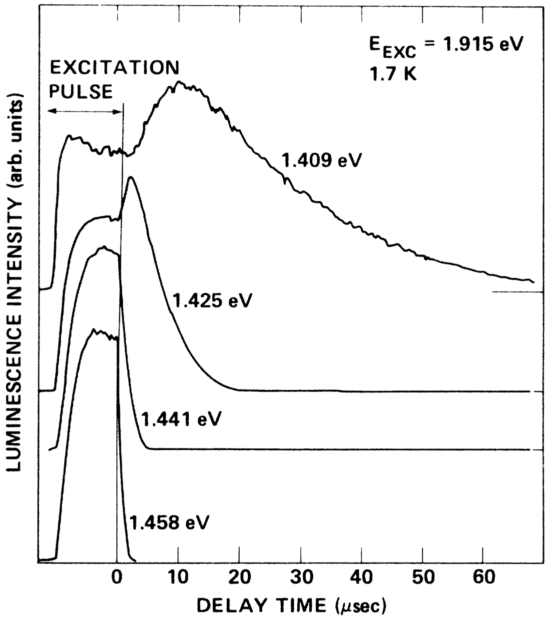

FIG. 3. Time-resolved luminescence decay measured at different emission energies. Note the increase in intensity after the end of the excitation pulse for the lowest emission energies.

tion. The intensity dependence of the other line is unclear because it is obscured by the $(0,h0)$ transition. At high excitation intensities, a further line near 1.50 eV increases rapidly above a well-defined excitation threshold, and eventually dominates the spectra. This luminescence is attributed to the equilibrium population of the first excited electron subband so that the transition is denoted $(1,h0)$. As the excitation intensity increases, the quasi-Fermi energy moves up in the band. At the same time, the internal field decreases so that the subband splitting is reduced. Our calculations predict that the electron Fermi level intersects the first excited subband at an electron density of 1.4×10^{12} cm^{-2}, and that under these conditions, the $(0,h0)$ transition is at 1.49 eV, with a bandwidth of 25 meV. These values are in good agreement with the observations.

The properties of the $(1,h0)$ transition are strongly influenced by the different electron wave function, which now extends further across the quantum well. The wave-function overlap with the heavy-hole band is therefore greater, so that the oscillator strength for the $(1,h0)$ transition is much larger than for $(0,h0)$. Consequently, $(1,h0)$ is observed as a distinct peak which eventually dominates the spectrum even though the electron population of the lower subband is much greater. The intensity dependence of the $(1,h0)$ peak energy is measured to be about half that of $(0,h0)$ as expected because of the decrease in subband splitting. Finally, the low occupancy of the upper subband accounts for the narrow luminescence line.

Although the subband splitting for holes is less than for electrons, the much larger heavy-hole density of states means that it is the electron quasi-Fermi energy that first crosses into the next subband.[7] However, at elevated temperatures, thermal excitation will first populate the hole subbands. This fortunate combination of parameters allows us to observe both types of higher-energy states. Figure 4 shows data at temperatures up to 80 K. There is a steady transfer of intensity from the $(0,h0)$ transition into a band of similar width, but higher in energy by about 30 meV. We also observe that the splitting between these pairs of lines is almost independent of excitation intensity.

The possible identifications for this thermalized luminescence line are $(0,l0)$ and $(0,h1)$, where $l0$ denotes the lowest light-hole band. Both hole levels are predicted to be at similar energies, and perhaps both contribute to the luminescence. The oscillator strength will be larger for either of these transitions than for $(0,h0)$ again because of the larger wave function overlap. As before, this is the reason that the transition is observed as a distinct peak, and also explains why the higher-energy peak completely dominates at high temperature.

It remains to identify the higher-energy transitions that are observed in Fig. 2 at low intensity and low temperature. Under these experimental conditions, equilibrium population of higher subbands will not occur. Instead, we attribute the lines to nonequilibrium population of the higher levels. After absorption of a photon, excited electrons and holes will cascade down the subband levels. Even though the scattering rate is expected to be large, the nonequilibrium states can contribute to the luminescence because of their large oscillator strength. We tentatively identify the two transitions as $(1,h0)^*$ and $(1,l0)^*$, where the asterisk denotes a nonequilibrium state. Decay measurements find that these transitions have a much faster decay than the $(0,h0)$ transition (<10 nsec), consistent with their being nonequilibrium states. The $(1,h0)^*$ transition is at a similar energy as the equilibrium $(1,h0)$ line observed at high intensity (see Fig. 2). Furthermore, the splitting between $(1,h0)^*$ and $(1,l0)^*$ is similar to that of the thermalizing pair in Fig. 4. These observations give us some confidence in the identification, although other transitions [e.g., $(0,l1)$, $(0,h2)$] could be in the same energy range.

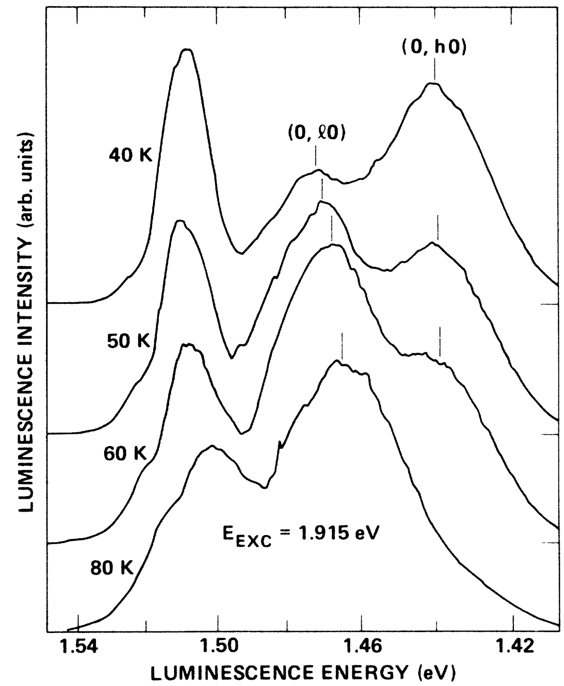

FIG. 4. Luminescence spectra at temperatures from 40 to 80 K, showing the thermalization between the $(0,h0)$ and $(0,l0)$ peaks.

The nonequilibrium transitions [principally $(1,l0)^*$] are observed at elevated temperatures, as shown in Fig. 4. Above about 100 K, luminescence intensity is transferred into this peak from the lower-energy $(0,l0)$ line, and these two form a thermalizing pair. Evidently, there is a transition from nonequilibrium to equilibrium population of this state.

Our interpretation of the excitation, time, and temperature dependence of the luminescence is supported by gradually changing results from samples with systematically changed "design parameters." These samples come from the same wafer but differ in doping densities due to an intentionally nonrotating sample holder during MBE growth. A more detailed comparison of these samples will be given elsewhere.[5]

In summary, we have demonstrated a *n-i-p-i* heterostructure that combines the quantized states of a heterojunction well, and the tunable effective band gap of a *n*-

i-*p*-*i* structure. The luminescence data approach that predicted for the ideal structure. The lowest subband transition exhibits the expected energy shift with excitation intensity, a line broadening dominated by the bandwidth of the quantized electron level, and a characteristic but very unusual luminescence decay. Luminescence transitions from higher-energy subbands are observed and given tentative identifications. Three mechanisms are observed for the occupation of the higher subbands: raising the quasi-Fermi energy into the subband, thermal equilibrium excitation of carriers, and nonequilibrium excitation.

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[1] See for example, L. Esaki, in *Proceedings of the 17th International Conference on the Physics of Semiconductors, San Francisco, 1984,* edited by J. D. Chadi and W. A. Harrison (Springer, New York, 1985), p. 473.

[2] G. H. Döhler, Phys. Status Solidi B **52**, 79 (1972); **52**, 533 (1972).

[3] G. H. Döhler and P. P. Ruden, Surf. Sci. **142**, 474 (1984).

[4] H. L. Störmer, R. Dingle, A. C. Gossard, W. Wiegmann, and R. A. Logan, in *Proceedings of the 14th International Conference on the Physics of Semiconductors, Edinburgh, 1978,* edited by B.L.H. Wilson (IOP, Bristol, 1978), p. 557.

[5] R. A. Street, G. H. Döhler, J. N. Miller, and P. P. Ruden (unpublished).

[6] W. Rehm, P. P. Ruden, G. H. Döhler, and K. Ploog, Phys. Rev. B **28**, 5937 (1983).

[7] We are neglecting heavy-light—hole mixing for the moment [see, for instance, Y. C. Chang and J. N. Schulman, Appl. Phys. Lett. **43**, 536 (1983)]. *n*-*i*-*p*-*i* heterostructures of appropriate design will represent ideal structures for the study of these phenomena because of weak excitonic effects.