Photoluminescence quenching in reverse-biased $Al_xGa_{1-x}As/GaAs$ quantum-well heterostructures due to carrier tunneling

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The photoluminescence intensity of reverse-biased $Al_xGa_{1-x}As/GaAs$ quantum wells decreases considerably with increasing electric field perpendicular to the quantum-well layers. We show that the photoluminescence intensity decrease is accompanied by an increase of the photocurrent, whose quantum efficiency is high. The phenomenon of photoluminescence quenching is therefore explained by the leakage of photogenerated carriers through the barrier layers rather than by enhanced nonradiative recombination processes within or in the vicinity of the quantum wells. We conclude that the photocurrent is determined by the tunneling of holes, and we apply a simple tunneling theory to interpret the observed data. A good agreement between theory and experiment is obtained by using the valence-band-edge discontinuity of $\Delta E_v/\Delta E_g=0.20$ at the $Al_xGa_{1-x}As/GaAs$ heterojunction.

I. INTRODUCTION

Electric-field-induced photoluminescence (PL) quenching in $Al_xGa_{1-x}As/GaAs$ quantum wells has recently attracted great attention.[1,2] A similar luminescence intensity decrease with increasing electric field has been reported for bulk material, such as $Al_xGa_{1-x}As/GaAs$ heterojunctions[3] and CdS Schottky diode structures,[4] where the PL quenching was interpreted as a consequence of field-induced sweeping of photogenerated carriers. Although the electric-field-induced luminescence quenching is qualitatively the same in quantum wells and in bulk material, several different mechanisms have been proposed to interpret the observed phenomenon in quantum well structures.

Photogenerated carriers are well confined in the quantum wells unless the electric field becomes very strong. The observed quenching of the excitonic transition has been explained by field-induced spatial separation of carriers.[5] Under flat-band condition both electron and hole wave functions are symmetric with respect to the center of the well. When an electric field is applied perpendicular to the quantum wells, the electron and hole distributions are polarized with opposite directions. Consequently, the overlap integral between electrons and holes decreases. The square of the overlap integral M_{cv}^2 has been calculated as a measure of the radiative recombination rate. However, the variation of M_{cv}^2 as a function of electric field is overly small to account for the observed results. In addition, the PL intensity quenching can be explained by this effect only if the recombination process of the biased quantum well is dominated by nonradiative processes. Otherwise, the luminescence efficiency should be insensitive to the decrease of the radiative recombination rate.

Another mechanism proposed by Mendez *et al.*[1,2] involves field-induced leakage of the wave functions into the $Al_xGa_{1-x}As$ barrier layers where a large number of nonradiative centers are assumed. However, under electric field the electron and hole distributions in the quantum wells are polarized in opposite directions. Therefore, this effect cannot enhance the nonradiative recombination in the constituent $Al_xGa_{1-x}As$ barrier layers. In addition to these two models, coupling between neighboring wells and tunneling through the barrier layers has been proposed.[6] However, at present no quantitative evaluation of the proposed models has been made.

In this paper we demonstrate that photocurrent measurements reveal important details of the electronic processes in quantum wells under electric field. In particular we show that the PL intensity quenching is closely correlated with the increase of the photocurrent which exhibits a high quantum efficiency. Therefore, we assume that the PL intensity decrease with increasing electric field is caused by the leakage of photogenerated carriers through the 20-nm $Al_xGa_{1-x}As$ barrier layers rather than by enhanced nonradiative recombination processes. We provide evidence for our assumption that the observed photocurrent is determined by the hole-tunneling process and we present a simple tunneling theory[7] to explain the experimental data. A good agreement is achieved by using a valence-band-edge discontinuity of $\Delta E_v/\Delta E_g=0.20$.

II. EXPERIMENTAL

The $Al_xGa_{1-x}As/GaAs$ quantum wells used in these experiments were prepared by molecular-beam epitaxy (MBE) on heavily Si-doped substrates kept at a temperature of 600° C. The samples are composed of three different single quantum wells with thicknesses of 5, 7.5, and 10 nm, respectively, separated by 20-nm-thick $Al_xGa_{1-x}As$ barrier layers, as shown in the inset of Fig. 1(a). The electric field was applied perpendicular to the quantum well layers through a semitransparent Schottky barrier contact formed by successively evaporating 10-nm Cr and 20-nm Au on the 200-nm-thick $Al_xGa_{1-x}As$ surface layer. The value of x in $Al_xGa_{1-x}As$ was chosen to be 0.30. The photoluminescence measurements were per-

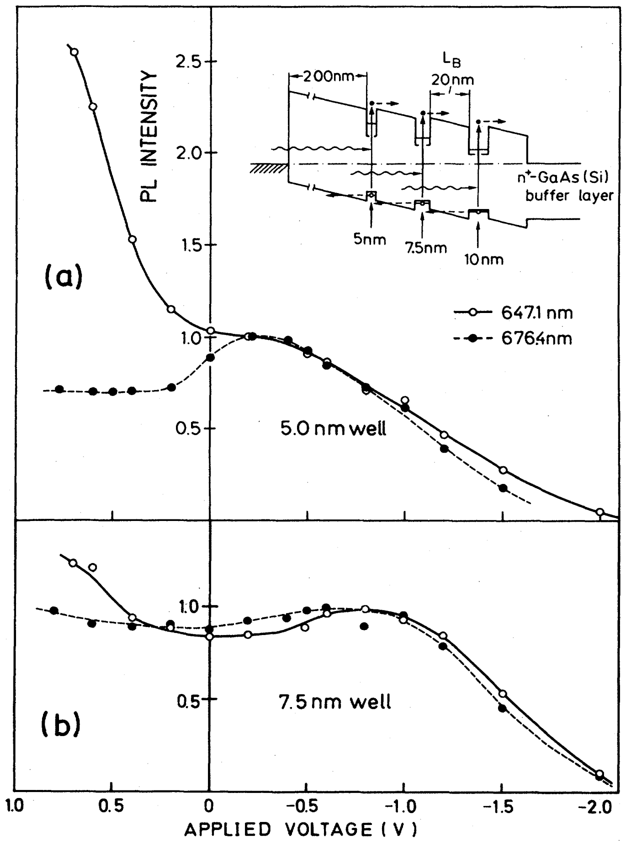

FIG. 1. Integrated photoluminescence intensity of 5-nm GaAs quantum well (a) and of 7.5-nm GaAs quantum well (b) as a function of external voltage applied perpendicular to the layers. The measurements were performed at 2 K using the 647.1- and 676.4-nm lines of a Kr^+ ion laser. The inset shows the layer sequence of the studied sample and the sweeping away and tunneling of photogenerated carriers schematically.

formed with the sample at 2 K by using the 676.4- and 647.1-nm lines from a Kr^+ ion laser for excitation. The net laser power absorbed by the sample at a spot size of 200-μm diameter was 10 mW. The resulting luminescence signal was analyzed with a 1-m Spex monochromator and detected by a GaAs cathode photomultiplier.

III. RESULTS AND DISCUSSION

The value of $x=0.30$ in the $Al_xGa_{1-x}As$ cladding and barrier layers enables us to excite either only the quantum wells by the 676.4-nm ($=1.833$ eV) line or the whole structure including the surface $Al_xGa_{1-x}As$ layer by the 647.1-nm ($=1.916$ eV) line of the Kr^+ ion laser. The observed luminescence spectrum consists of four distinct emission peaks located at energies of 1.51, 1.55, 1.59, and 1.63 eV, respectively. The low-energy line arises from the n^+-type GaAs buffer layer, and its intensity remains constant regardless of the applied voltages. This is due to the fact that no significant field which might sweep away the carriers can be established in this heavily doped (10^{18} cm^{-3}) layer. The other peaks originate from the quantum wells, and as expected from previous results, their intensities depend strongly on the applied electric field. In addition, the quantum-well luminescence shifts to lower energy by 2 meV/V for the 5-nm well and by 6 meV/V for the 10-nm well.

In Fig. 1(a) we show the observed integrated photoluminescence intensity from the 5-nm well as a function of applied voltage. The data expressed by open circles are obtained by the 647.1-nm excitation, while the dots are obtained by 676.4-nm line. Excitation by 647.1-nm line yields an open circuit voltage of 0.7 V, which is very close to the flat-band condition. The luminescence intensity decreases when increasing the bias voltage toward the reverse direction. The first steep decrease between $+0.7$ and $+0.2$ V is probably caused by sweeping away the photogenerated carriers from the $Al_xGa_{1-x}As$ surface layer. At very low electric field, both electrons and holes generated in the $Al_xGa_{1-x}As$ surface layer diffuse into the quantum wells, preferentially into the outermost 5-nm quantum well, and provide a substantial contribution to the PL intensity. Under electric field, however, holes in the $Al_xGa_{1-x}As$ surface layer are swept away toward the surface electrode and do not contribute to the luminescence of the quantum wells.

The proposed mechanism is confirmed by comparison with the data obtained by the 676.4-nm excitation [dots in Fig. 1(a)]. Excitation by the 676.4-nm line yields a very small open-circuit voltage, thus indicating no carrier generation in the $Al_xGa_{1-x}As$ surface layer and no significant carrier leakage from the quantum wells. When the reverse-bias voltage is increased from the flat-band condition at 0.7 V, the luminescence intensity exhibits no appreciable decrease. [Actually, the data of Fig. 1(a) indicate a slight increase between the voltages $+0.2$ and -0.2 V. However, this characteristic variation depends upon the specific sample. Most of the investigated samples do not exhibit a significant PL intensity variation between $+0.7$ and -0.2 V when excited by the 676.4-nm line.] Between 0 and -0.4 V, there is a plateau in the luminescence intensity versus voltage characteristics for both the 647.1- and the 676.4-nm excitation. The intensity decreases again with further increase of the reverse-bias voltage. We assume that this decrease is due to the escape of photogenerated carriers from the quantum well.

The photoluminescence intensities from the wider quantum wells of the sample show a similar bias-voltage dependence, except that the voltage at which the intensities begin to decrease from the plateau changes slightly with different well widths. In Fig. 1(b) we display the integrated luminescence intensity of the 7.5-nm well as a function of the applied voltage. The forward-bias characteristics do not exhibit a significant difference between the 647.1- and the 676.4-nm excitation. This observation indicates that the influence of carriers photogenerated in the surface $Al_xGa_{1-x}As$ layer on the luminescence intensity is relatively small in the more distant wells. The voltage at which the PL intensity begins to decrease is about -1 V, which is slightly higher than that observed for the 5-nm wide quantum well. This result provides first evidence that the observed luminescence decrease with increasing electric field is not due to the polarization effect,[5] but due to carrier escape from the quantum wells. The ef-

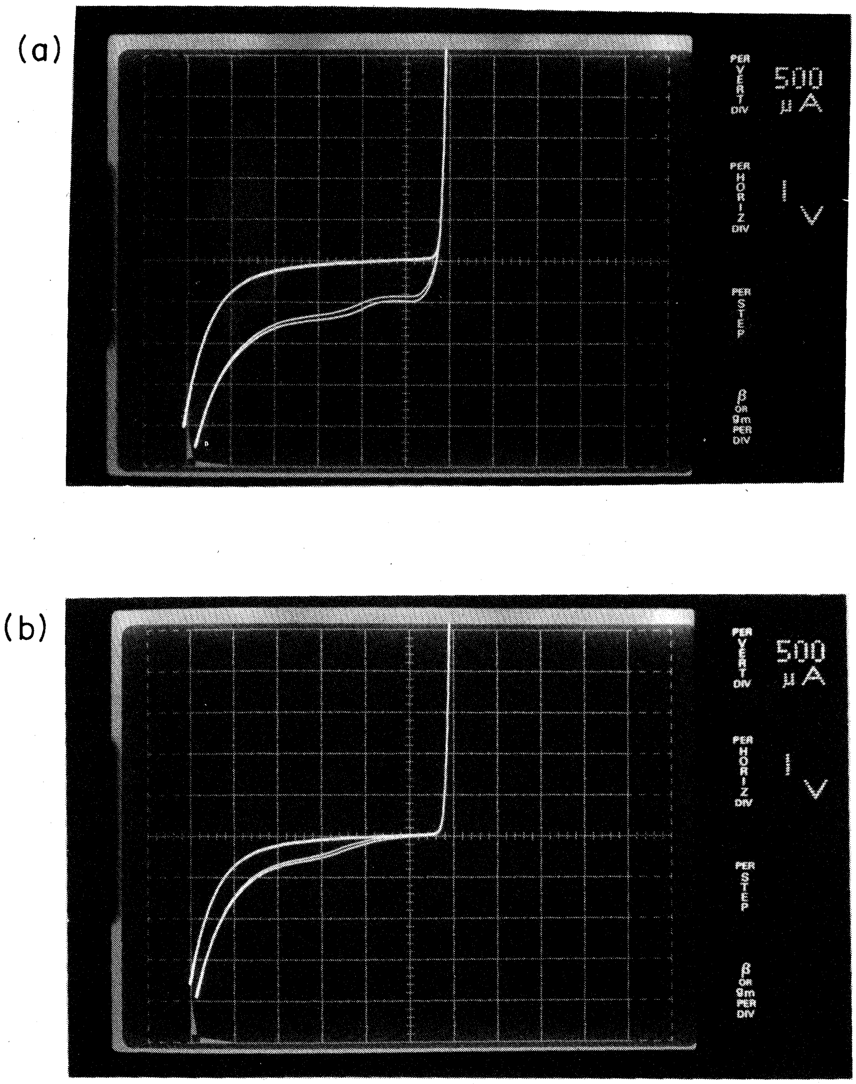

FIG. 2. Current-voltage characteristics of $Al_xGa_{1-x}As/GaAs$ quantum-well heterostructure. The upper curve represents the dark current, while the lower trace shows the photoresponse; (a) under 647.1-nm excitation, (b) under 676.4-nm excitation.

fective barrier height for carriers populating the quantized levels is higher for the wider quantum wells, and thus the voltage needed to sweep these carriers out of the well is expected to be higher.

We now discuss the observed current-voltage characteristics of the quantum-well sample in the dark and under illumination, which are depicted in Fig. 2. When the sample is excited by the 647.1-nm line, the photocurrent increases steeply as the bias voltage increases toward the reverse direction as shown in Fig. 2(a). The photocurrent in the $+0.7-+0.2$-V range is caused by the field-induced sweeping away of photogenerated carriers from the surface $Al_xGa_{1-x}As$ layer, because no photocurrent response is observed in this bias range under 676.4-nm excitation, as shown in Fig. 2(b). While holes easily reach the metallic surface contact, electrons swept away from the surface $Al_xGa_{1-x}As$ layer must pass through the quantum wells before reaching the n^+-type GaAs buffer layer. It is important to note that obviously the quantum wells can be easily overcome by these electrons since the photocurrent increase in the $+0.7-+0.2$-V range is very steep. We interpret this phenomenon as follows. The quantum wells have a small trapping efficiency for the photogenerated electrons swept away from the surface $Al_xGa_{1-x}As$ layer at a certain electric field, because these electrons have energies higher than the barrier height and they are accelerated by the external field. In addition, the energy relaxation of hot electrons in quantum wells is apparently very slow.[7] Therefore, these photogenerated electrons have large probabilities to overcome the barrier layers. This process is enhanced by the absence of charge neutrality in the quantum wells. At about $+0.2$ V, the photocurrent saturates and the current-voltage characteristic exhibits a plateau

[Fig. 2(a)]. Using a bias of about −0.5 V, the photocurrent increases again stepwise and then saturates for reverse bias voltages larger than −1.5 V. The latter step of the photocurrent increase is due to the escape of photogenerated carriers from the quantum wells, because the luminescence intensity of the quantum well decreases considerably when the photocurrent increases, as shown in Fig. 1. This interpretation of the experimental results is confirmed by the data of Fig. 2(b). When only the quantum wells are excited by the 676.4-nm line, we observe only the second photocurrent step.

The detected second step may include the photocurrent arising from the excitation of the n^+-type GaAs buffer layer. The photocurrent of Fig. 2(b) saturates at about −2.0 V which is slightly higher than the voltage for the second-step photocurrent saturation of Fig. 2(a). This difference is probably due to the contribution from n^+-type GaAs buffer layer, because the 676.4-nm line excites the buffer layer more strongly than the 647.1-nm line. However, the resulting photocurrent of Fig. 2(b) shows a saturation value very similar to that observed in the second-step photocurrent of Fig. 2(a). This result implies that the effect of carriers generated in the n^+-type GaAs buffer layer on the total photocurrent is very small even under strong excitation. This minor contribution to the photocurrent is consistent with the fact that no discernible bias dependence is observed in the buffer layer luminescence intensity. In the following quantitative evaluation we therefore neglect the photocurrent due to the buffer layer, especially when considering the 647.1-nm excitation.

The important result of our measurements is that the photoluminescence intensity decrease is closely correlated to the increase of photocurrent perpendicular to the quantum wells. The first steep increase of photocurrent depicted in Fig. 2(a) is caused by sweeping away photogenerated carriers from the $Al_xGa_{1-x}As$ layers, especially from the surface $Al_xGa_{1-x}As$ layer. At about −2 V, the second-step photocurrent saturates and the observed photoluminescence vanishes almost completely. This means that at the electric field corresponding to this voltage almost all the photogenerated carriers in the quantum wells escape through the barrier layers. When the sample is excited by the 647.1-nm line, the photocurrent due to the quantum wells at this bias can be calculated as follows:

$$I=qI_0e^{-\alpha_b L_s}[(1-e^{-\alpha_w L_{z_1}}) + e^{-(\alpha_b L_b+\alpha_w L_{z_1})}(1-e^{-\alpha_w L_{z_2}}) + e^{-(2\alpha_b L_b+\alpha_w L_{z_1}+\alpha_w L_{z_2})}(1-e^{-\alpha_w L_{z_3}})], \tag{1}$$

where I_0 represents the number of incident photons per second and q the electronic charge. L_s, L_b, and L_{z_i} denote the thicknesses of the surface $Al_xGa_{1-x}As$ layer, barrier layer, and quantum wells, respectively. α_b and α_w correspond to the absorption coefficients of $Al_{0.3}Ga_{0.7}As$ and of GaAs for 647.1-nm light, respectively. Using the net incident power of 10 mW, $\alpha_w=3.7\times10^4$ cm^{-1},[8] and $\alpha_b=1\times10^4$ cm^{-1},[9] the expected photocurrent due to the quantum wells is estimated from Eq. (1) to be 330 μA. From the saturation value of the second-step photocurrent in Fig. 2(a) we obtain the measured photocurrent arising from the quantum wells amounting to 190 μA, which corresponds to a high internal quantum efficiency of 57%. This result manifests that the quenching of the photoluminescence intensity at reverse bias is not due to increased non-radiative recombination processes, but due to the escape of photogenerated carriers from the quantum wells.

We next discuss the mechanism of escape of photogenerated carriers by tunneling through the barrier layers. Since the photogenerated carriers are effectively confined in the quantum wells and the measurements are performed at 2 K, the tunneling process through the barrier layers is the most plausible mechanism for explanation of the observed photocurrent. Using the 0-K approximation, the current through the barrier can be expressed by the equations derived by Simmons,[10]

$$J=\frac{q}{2\pi h L_b^2}\left\{\left[\phi-\frac{qV}{2}\right]\exp\left[-\frac{4\pi L_b}{h}(2m^*)^{1/2}\left[\phi-\frac{qV}{2}\right]^{1/2}\right] - \left[\phi+\frac{qV}{2}\right]\exp\left[-\frac{4\pi L_b}{h}(2m^*)^{1/2}\left[\phi+\frac{qV}{2}\right]^{1/2}\right]\right\}, \tag{2}$$

for weak field ($0\leq V\leq\phi/q$), and

$$J=\frac{2.2q^3F^2}{8\pi h\phi}\left\{\exp\left[-\frac{8\pi}{2.96hqF}(2m^*)^{1/2}\phi^{3/2}\right]-\left[1+\frac{2qV}{\phi}\right]\exp\left[-\frac{8\pi}{2.96hqF}(2m^*)^{1/2}\phi^{3/2}\left[1+\frac{2qV}{\phi}\right]^{1/2}\right]\right\}, \tag{3}$$

for high field ($V>\phi/q$).

In these equations, h and m^* represent Planck's constant and the effective mass of the tunneling particle, respectively. The effective potential ϕ/q represents the barrier height measured from the Fermi level. V and F are the voltage across the barrier layer and the electric field in the barrier layer, respectively. Equations (2) and (3) were developed for tunneling currents between three-dimensional materials separated by insulating material. We assume that these equations are also valid for the present case. Within the 0-K approximation the quasi-Fermi levels for electrons and holes in the well are as-

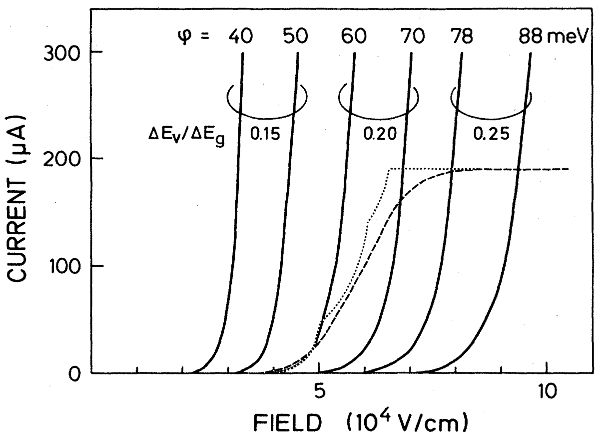

FIG. 3. Calculated hole-tunneling current (solid lines) and observed photocurrent (dashed curve) in $Al_xGa_{1-x}As/GaAs$ quantum-well heterostructures. The values of $\phi=40$, 60, and 78 meV correspond to the barrier heights of the 5-nm well with $\Delta E_v/\Delta E_g=0.15$, 0.20, and 0.25, respectively, while $\phi=50$, 70, and 88 meV correspond to those of the 10-nm well. The dotted curve represents the calculated photocurrent with $\Delta E_v/\Delta E_g=0.20$ and using the measured internal quantum efficiency of 57%.

sumed to be fixed at the lowest quantized electron and hole levels, respectively, under excitation. We have calculated the tunneling current within the illuminated spot-size area with a 200-μm diameter for both electrons and holes using Eqs. (2) and (3), and the results for holes are depicted in Fig. 3.

At a reverse-bias voltage of -1.5 V, the second photocurrent step is saturated almost completely as shown in Fig. 2(a). The resulting electric field at this voltage is estimated to be as high as 8×10^4 V cm^{-1}. At this electric field, the electron tunneling current through the 20-nm thick barrier is too small to explain the observed photocurrent. The electron-leakage current from the 5-nm well is only 0.1 μA when $\Delta E_c/\Delta E_c=0.85$ is assumed ($E_c=0.318$ eV, $\phi=0.227$ eV), and still as low as 3.5 μA even when $\Delta E_c/\Delta E_g=0.70$ ($E_c=0.262$ eV, $\phi=0.176$ eV) is assumed. Although the estimated hole tunneling current would be sufficiently large to account for the experimental data, the measured photocurrent is determined by the slower process because of charge neutrality, i.e., in our case by the electrons. The proposed tunneling mechanism seems therefore not to be able to explain the observed photocurrent. Photogenerated electrons in the quantum wells, however, have very high initial energies. Electrons excited by the 647.1-nm line have energies larger than the barrier height, and those excited by the 676.4-nm line approximately equal the barrier energy, even if the largest well depth reported ($\Delta E_c/\Delta E_g=0.85$) (Ref. 11) is assumed, because of the higher effective mass in the valence band. Two processes compete, namely thermalization (relaxation) of the electrons in the quantum wells and acceleration by the external field. Ryan *et al.*[7] reported an extremely slow energy relaxation of photogenerated carriers in quantum wells. The electron-phonon scattering lifetime was estimated to be about 7 ps. When we take into account these effects, we can expect that the electron tunneling current is enhanced as compared to the situation where all electrons are located at the bottom of the band and may be even higher than the hole tunneling current. In this case the slower hole tunneling process determines the overall photocurrent.

In Fig. 3 we show the hole tunneling current as a function of the electric field calculated for different ϕ values. The values of $\phi=40$, 60, and 78 meV correspond to the effective barrier heights of the 5-nm well assuming different values for the band-edge discontinuity ($\Delta E_v/\Delta E_g=0.15$, 0.20, and 0.25, respectively), while $\phi=50$, 70, and 88 meV correspond to the barrier height of the 10-nm well. The dashed curve represents the measured photocurrent which shows saturation at about 190 μA due to the excitation rate. The dotted curve represents the calculated photocurrent assuming a band-edge discontinuity of $\Delta E_v/\Delta E_g=0.20$ and the observed 57% internal quantum efficiency. In our calculation, the photocurrent from each well is assumed to saturate at the electric field where the calculated tunneling current reaches the value corresponding to the excitation rate. Therefore, the structures of the calculated dotted curve arise from the saturation of the photocurrent from each constituent well of our sample. Although this particular structure is not observed in the experimental curve, the agreement between theory and experiment is good. This result implies that the valence-band discontinuity, $\Delta E_v/\Delta E_g$, for $Al_xGa_{1-x}As/GaAs$ heterojunctions is 0.20. This value coincides with Dingle's rule and is in agreement with a recent reevaluation of the conduction-band-edge discontinuity $\Delta E_c/\Delta E_g$.[11,12] The present estimation of $\Delta E_v/\Delta E_g$ is, however, tentative because the exact electric field in our device structure is not known. In addition, effects arising from "hot" holes and defect centers near the heterojunction have been neglected in this study.

IV. CONCLUSION

The application of an external electric field perpendicular to the layers quenches the photoluminescence of $Al_xGa_{1-x}As/GaAs$ quantum wells. The decrease of luminescence intensity with increasing reverse bias voltage is directly correlated with an increase of the photocurrent which reaches an internal quantum efficiency as high as 57%. Our photocurrent measurements provide direct experimental evidence that the phenomenon of electric-field-induced luminescence quenching in quantum wells is caused by the leakage of photogenerated carriers through the barrier layers rather than by enhanced nonradiative recombination processes. The photogenerated electrons have energies higher than the barrier height, and they are therefore easily accelerated by the external field and not thermalized in the quantum wells. The overall photocurrent process is thus determined by the hole-tunneling behavior. A simple tunneling theory is applied to interpret the observed photocurrent, and a good agreement between theory and experiment is obtained, if the valence-band-edge discontinuity at the $Al_xGa_{1-x}As/GaAs$ heterojunction is assumed to be $\Delta E_v/\Delta E_g=0.20$.

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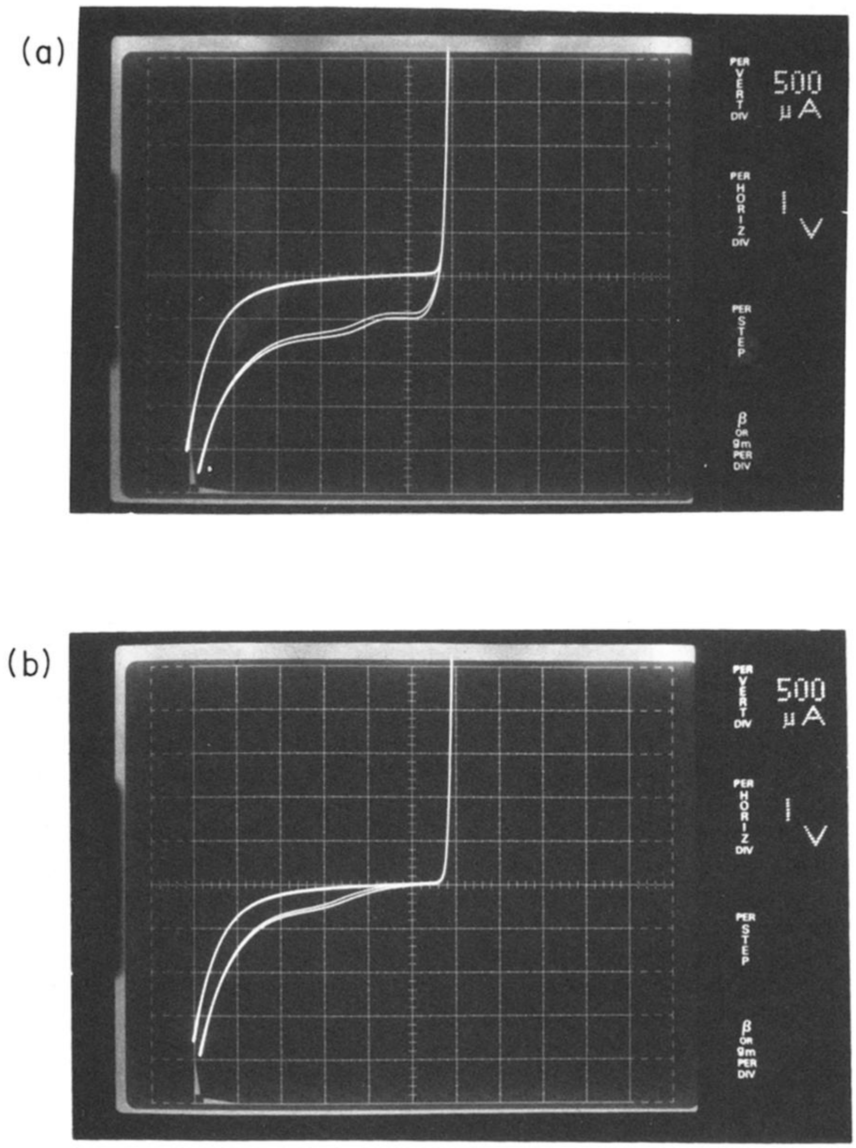

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