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Effect of an electric field on the luminescence of GaAs quantum wells

E. E. Mendez, G. Bastard,* L. L. Chang, and L. Esaki
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

H. Morkoc and R. Fischer
University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

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Low-temperature photoluminescence (PL) measurements have been performed in narrow GaAs-$Ga_{1-x}Al_xAs$ quantum wells subject to an electric field perpendicular to the well plane. At low fields the PL spectra show two peaks associated, respectively, with exciton and free-electron—to—impurity recombination. With increasing field the PL intensity decreases, with the excitonic structure decreasing at a much faster rate, and becomes completely quenched at a field of a few tens of kV/cm. This is accompanied by a shift in the peak position to lower energies. The results are interpreted as caused by the field-induced separation of carriers and modification of the quantum energies. Variational calculations performed for isolated, finite quantum wells explain qualitatively the experimental observations.

I. INTRODUCTION

The main consequences of the quantum size effect in semiconductor heterostructures are the confinement of the electric carriers in a one-dimensional potential well and the formation of discrete energy states. These two results have been used extensively in the study of the properties of multiheterostructures and, in particular, in the interpretation of photoluminescence (PL) experiments. Thus, in undoped GaAs-$Ga_{1-x}Al_xAs$ quantum wells (QW), the main PL structure has been attributed to the excitonic transition between the ground quantum levels ($n=1$) of electrons and heavy holes.[1-3] Additional peaks, observed in some cases, have been interpreted as due to the recombination of $n=1$ electrons with neutral carbon acting as an acceptor.[4]

Under the influence of external parameters such as an electric field, applied perpendicularly, significant effects on the carrier confinement and energy states of the QW would be expected. The field should polarize the carrier distribution along or against its direction, depending on the sign of the carrier effective mass, and the quantum states should experience an energy shift caused by the field. These concepts, should they amount to an experimentally significant effect, could be used in a variety of optoelectronics applications, e.g., for control and modulation of both the photon energy and the intensity output of radiation-emitting devices based on quantum wells and superlattices.

In an attempt to test these basic ideas, we have done PL measurements on GaAs-$Ga_{1-x}Al_xAs$ multiheterostructures subject to an electric field perpendicular to them. The results show a strong influence of the field on both the luminescence intensity and peak energy position. The intensity decreases with increasing field, and even completely quenches for moderate fields, and its energy peak is shifted to lower energies. We believe that the main mechanisms responsible for these results are the field-induced polarization of the carriers confined in the quantum wells and the modification of their quantum energy states.

II. EXPERIMENTAL

The samples under study were prepared by growing, by molecular bean expitaxy (MBE), a sequence of layers on Si-doped GaAs (100) substrates: a 1-μm GaAs buffer layer, a 0.5-μm $Ga_{1-x}Al_xAs$ layer, a series of six identical GaAs-$Ga_{1-x}Al_xAs$ quantum wells, and finally a 0.1-μm clad layer of $Ga_{1-x}Al_xAs$. All the layers were grown at 700 °C, except the buffer layer grown at 600 °C. The width of the alloy barriers of the wells was 100 Å; that of the GaAs regions, L_z, averaged from 20 to 35 Å, depending on the sample. The $Ga_{1-x}Al_xAs$ layers were undoped, while the GaAs wells were either undoped or doped with Si to $\sim 10^{16}$ cm^{-3}. The value of x in the $Ga_{1-x}Al_xAs$ layers varied from sample to sample, ranging from 0.32 to 0.40, as determined from Raman scattering[5] and electroreflectance measurements.

The electric field was applied to the GaAs quantum wells with the use of a Schottky-barrier configuration, formed by evaporating a semitransparent Au film on the multiheterostructure. The dark-current—voltage characteristics of the diodes thus made showed rectifying behavior, with a breakdown voltage of ~ 10 V at both 300 and 6 K. The average built-in field was estimated to be $\sim 10^4$ V/cm and the space-charge width ~ 0.8 μm, so that the quantum wells were well inside the space-charge region. Luminescence was excited on the diodes, at $T = 6$ K, with the use of the 5145-, 6328-, or 6471-Å lines of an Ar^+, He-Ne, or Kr^+ laser, respectively, and was detected with a cooled photomultiplier placed at the exit slit of a $\frac{3}{4}$-m double-pass monochromator.

III. RESULTS AND DISCUSSION

Figure 1 shows the effect of the electric field on the PL of GaAs quantum wells, excited with the 6328-Å laser line (1.96 eV). The field in the well was varied by changing the terminal voltage V_{ext} of the metal with respect to the GaAs substrate. A significant photovoltage was induced by the laser excitation even at the low power densities used in the experiments. For the sample whose spectra are shown in Fig. 1, this voltage was 0.73 V for an excitation power density of 0.08 W/cm^2. The topmost spectrum corresponds to this open circuit configuration, where two well-defined structures are observed at 1.681 and 1.662 eV. The former, with a full width at half height of 6 meV, is attributed to a free exciton recombination between the electron and heavy-hole ground states of the GaAs quantum wells. The latter has a different origin, as reflected from its different behavior under the electric field, and most probably involves the recombination of ground-state electrons with carbon acting as an acceptor (e,A^0), invariably present in MBE-grown materials.

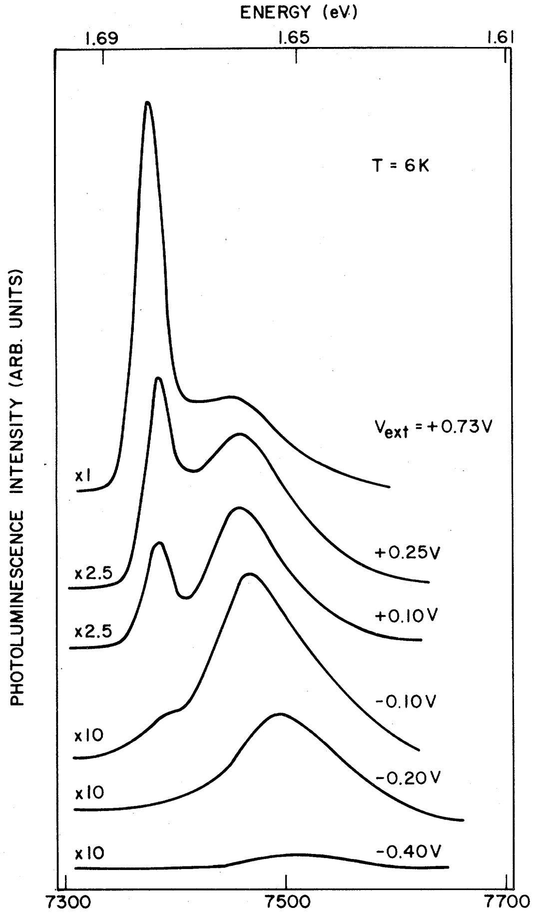

FIG. 1. Photoluminescence intensity, for various applied voltages, vs emission wavelength for GaAs quantum wells with $L_z \approx 35$ Å. The spectra were taken using 1.96-eV excitation energy and a power density of 0.08 W/cm^2.

As the terminal voltage was decreased, the intensity of both luminescence peaks decreased, but the one at higher energy did so at a much faster rate. The exciton peak was completely quenched at $V_{ext} \simeq -0.2$ V, whereas the impurity peak disappeared at $V_{ext} \simeq -0.5$ V. Analogous behavior was observed in other Schottky diodes, from different wafers, with comparable values of L_z and x. The relative quenching rate varied slightly from sample to sample. In general, for a fixed excitation photon energy, the exciton quenching rate depended on the excitation intensity. With decreasing intensity, in the range 1–0.01 W/cm^2, the quenching rate increased, and at the lowest intensity, for which the photocurrent was minimal, the exciton peak was completely quenched for $V_{ext} = 0.2$ V (for the sample shown in Fig. 1).

The other apparent feature in Fig. 1 is the shift of the impurity peak to lower energies when V_{ext} is decreased. A plot of this shift versus V_{ext} is shown in Fig. 2 and reveals a linear dependence for $V_{ext} < 0.25$ V, with a slope of ~0.03 eV/V. Also represented in Fig. 2 is the position of the exciton peak as a function of V_{ext}, down to 0 V, beyond which voltage the peak is completely quenched. Qualitatively similar

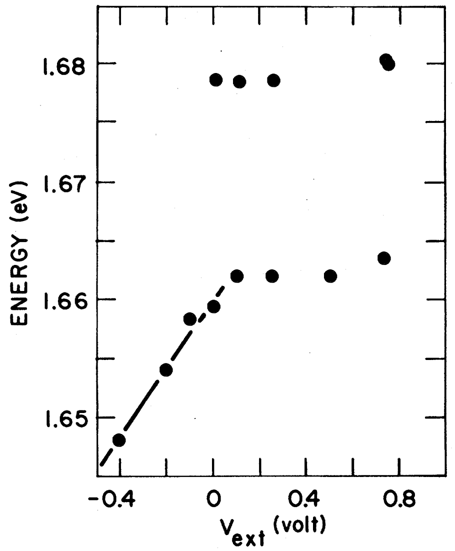

FIG. 2. Energy positions of the PL peaks for the sample of Fig. 1 as a function of the applied voltage.

shifts were observed in other samples studied, but the slope varied with the value of L_z. For example, for a diode with $L_z \approx 20$ Å the shift observed was ~ 0.005 eV/V.

The experimental observations just described were independent of the excitation energy in the range considered :1.92–2.41 eV. It should be noted that 1.92 eV is well below the bandgap of the $Ga_{1-x}Al_xAs$ layers and, in some samples, was almost resonant with the excitonic transition (only ~ 0.08 eV above it). This is most significant because it implies that the luminescence quenching has its origin in the electric field in the quantum wells.

In the past, a considerable decrease of the PL intensity in bulk CdS and GaAs under an electric field has been reported.[6,7] This has been interpreted as a consequence of the field-induced sweeping of photogenerated carriers. Under an electric field, in a Schottky-barrier configuration, the carriers excited by the incident radiation are swept away from the space-charge region and are not effective in radiative recombination. The PL intensity is thus reduced by the extent to which the space-charge width is increased by the field. This is not the case in the present study because the quantum wells, where the PL is generated, are inside the space-charge region even in the absence of any external voltage (due to the built-in field).

Luminescence quenching of excitons in ultrapure bulk GaAs (10^{13}–10^{14} carriers/cm^3), induced by an external electric field, has also been reported.[8] The suppression of exciton luminescence occurred at extremely low fields, ~ 1 V/cm, and was accompanied by an increase in the (e,A^0) recombination rate. These effects were interpreted on the basis of an exciton dissociation by impact ionization. This mechanism cannot explain our results, though, because the electric field required to quench the PL of the quantum wells is much higher, of the order of a few tens kV/cm. On the other hand, a reduction of the PL due to the (e,A^0) transition accompanied the extinction of the exciton PL in QW, opposite to the behavior observed in the ultrapure GaAs experiment.

A tempting explanation to our results would be the ionization of the exciton by the electric field. It is well known that a field of the order of $F_I = R_\infty / e a_\infty$ can ionize an exciton.[9] (R_∞ and a_∞ are, respectively, the three-dimensional effective Rydberg and Bohr radius and e is the electronic charge. For GaAs it is $F_I \simeq 5$ kV/cm.) However, this is not the case in a confined medium like a GaAs quantum well, when the electric field is perpendicular to the well and $L_z < a_\infty$.

We believe that the quenching of the excitonic luminescence is due to the spatial separation of carriers induced by the electric field. At zero field the electron and hole wave functions are symmetrical with respect to the center of the well. When a field is applied perpendicular to it, the electron and hole distributions are polarized in opposite directions and, consequently, the recombination probability is effectively decreased. The results shown in Fig. 1 are for decreasing V_{ext}, i.e., increasing total field, including both built-in and applied. Even though the actual value of the field was not known, it can be estimated, for the range of V_{ext} of this work, to be of a few tens of kV/cm. With an increase in V_{ext}, the observed luminescence intensity also increases, as expected. This is of course limited by the large associated forward current which prevented us from reaching the flat-band condition, beyond which a decrease in PL intensity is to be expected.

The observed shift to lower energies of the impurity PL peak with increasing electric field is interpreted as a direct consequence of the effect of the field on the energy position of the electron and hole quantum states.

In an attempt to interpret quantitatively our results we have done variational calculations for the ground state of a particle confined in a finite quantum well, in the presence of an electric field. The results, which will be published in detail elsewhere, can be summarized as follows. The energy shift induced by the electric field is negative, independently of the sign of the field, and for a given value the magnitude of the shift increases with the particle effective mass and the width of the QW. An application to electrons ($m^* = 0.067 m_0$; m_0 is the free electron mass) in a GaAs-$Ga_{1-x}Al_xAs$, ($x \sim 0.35$) QW leads to a shift of -0.2 meV for $L_z = 30$ Å and -1.2 meV for $L_z = 100$ Å, when the field is 50 kV/cm. The corresponding shifts for heavy holes are, respectively, -0.8 and -6.4 meV.

The calculated field-induced polarization in a GaAs QW is larger for heavy holes than for electrons, and increases with L_z. A measure of the induced spatial separation between the two kinds of carriers is the overlap integral M_{cv}, defined as

$$M_{cv} \equiv \int_{-\infty}^{\infty} dz\, \psi_e(z) \psi_{hh}(z) \quad ,$$

where ψ_e and ψ_{hh} are the wave functions describing the electron and heavy-hole distributions, respectively. Figure 3 shows M_{cv}^2 as a function of the electric field, for two different well thicknesses. The radiative recombination rate is proportional to M_{cv}^2 so that for $L_z = 30$ Å and a field of 50 kV/cm, we would expect a decrease of the luminescence intensity of $\sim 3\%$.

The calculated results, although they predict the qualitative trends correctly, give rise to too small an effect for the magnitude of fields under consideration. The calculations were done using the values of 0.4 and 0.07 eV for the depths of the electron and hole wells, respectively. These figures describe the situation for the GaAs QW studied, if we use the

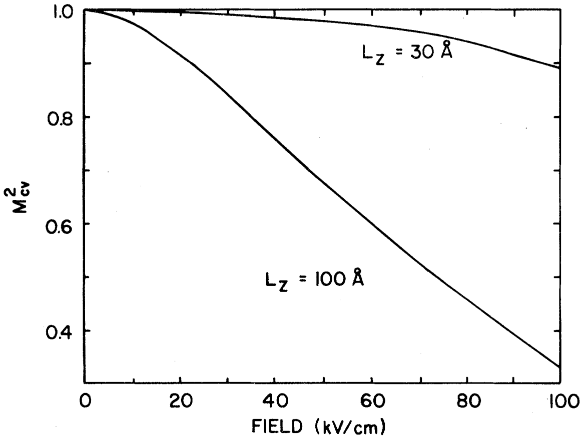

FIG. 3. Calculated values of the square of the overlap integral between electrons and heavy holes in a GaAs QW as a function of an electric field applied perpendicularly to the well.

85%-15% rule for the conduction- and valence-band discontinuities.[10] A deviation of this empirical rule that would make the hole well shallower would lead, especially for the narrow wells under consideration, to a larger polarization of the hole wave function, thus reducing further the recombination rate. An additional reduction of the PL intensity is to be expected as a result of the field-induced leakage of the wave functions into the $Ga_{1-x}Al_xAs$ barriers, which are known to have a large number of nonradiative recombination centers.

We think, however, that the major reason for the quantitative discrepancy between theory and experiment lies in the coupling of the wells, particularly in view of the narrow wells and barriers. The electric field in this case enhances both the lowering in energy of the quantum states and, more importantly, the polarization of the carrier distributions beyond what can be achieved in a simple well. Work is now in progress to incorporate these ideas in the calculations and to test them experimentally through various sample configurations.

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*Permanent address: Ecole Normale Superieure, Paris, France.

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