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Excitation-intensity-dependent photoluminescence in semiconductor quantum wells due to internal electric fields

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Low-temperature photoluminescence (PL) has been studied in several III-V single-quantum-well (SQW) samples. We have observed shifts of the PL peak energy as a function of the excitation intensity which, we show, are due to the quantum confined Stark effect (QCSE) caused by the internal electric field at the surface and/or heterointerfaces in the samples. The experimentally measured PL peak shifts in $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs and $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs SQW's are compared with calculations based on the QCSE and excellent agreement is found. The magnitude of the internal field determined from the PL analysis coincides with the value for the electric field obtained from photoreflectance measurements on the same samples, at the same temperatures.

I. INTRODUCTION

The influence of static electric field on the properties of semiconductor quantum wells (QW) has received considerable attention during the past decade.[1–5] Under the influence of a perpendicular electric field, the electrons and holes become polarized in the QW, resulting in significant shifts in the energy and intensity of the optical transitions, a phenomenon that became known as the quantum confined Stark effect (QCSE).[6,7] This effect, first discussed by Miller *et al.* in 1984, forms the basis of a number of optoelectronic devices.[6]

The QCSE is most often studied by optical techniques based on absorption measurements, including electroabsorption and photocurrent spectroscopy, with the electric field typically being applied via semitransparent Schottky contacts.[8–10] It is interesting to note that while photoluminescence spectroscopy is one of the most often used experimental techniques in semiconductor physics, it has not been widely used to examine the effects of electric field on the QW's. This is in spite of the fact that the QCSE has a similar effect on luminescence transitions as it has on absorption. The question of the effect of the electric field on photoluminescence (PL) is important, however, as most PL measurements are made on samples that contain QW's near a surface or a heterointerface (i.e., the majority of QW samples) and are influenced by internal electric fields that are intrinsic to these layer structures. Since PL is often used to determine various QW parameters (such as well widths, barrier height, etc.) by comparing the PL peak energy with model calculations, it is important to understand the magnitude and consequence of the internal electric fields on the PL spectra.

In this paper we shall present calculations and experimental results that quantify the effect of the internal electric fields on the PL emission of semiconductor QW's. We shall show that the observed nonlinear intensity dependence of the PL signal can be accurately described by the QCSE resulting from the built-in electric fields. We shall also show that the magnitude of the internal electric field may be obtained from conventional PL data and this value accurately corresponds to that determined by other methods, such as photoreflectance (PR) spectroscopy.

II. EXPERIMENTAL DETAILS AND RESULTS

We have studied the excitation intensity dependence of PL of a great number of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs and $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs samples containing single and multiple quantum wells, grown by metal-organic chemical vapor deposition (MOCVD) and molecular-beam epitaxy techniques. Consistent results have been obtained for all the samples, but for the sake of simplicity we shall focus our attention in this publication on an $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs single-quantum-well (SQW) sample and an $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs sample containing two SQW's.

The $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs SQW was grown by MOCVD on an n^+-type, Si-doped, GaAs(100) substrate. The quantum well was undoped and consisted of 120-nm barriers and a single 11-nm-thick $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ ($x=0.20$) well. The $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs sample discussed in this paper included two undoped MOCVD grown SQW's, on an n^+ substrate and buffer layer. The quantum-well barriers were 50-nm-thick $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ ($x=0.54$) layers and the two wells were 4.3- and 7.6-nm-thick GaAs. PL was excited with an Ar-ion laser ($\lambda=514$ nm) and detected with a Si photodiode after having been dispersed by a 0.75-m spectrometer. The sample temperature was cooled to $T=12$ K by a closed cycle refrigerator. The excitation power was varied between 50 μW and 100 mW using calibrated neutral density filters. The excitation (laser) beam was slightly focused to a 0.5-mm diameter spot on the sample.

As the quantum wells were undoped and grown on n^+ layers, a surface electric field is generated in these samples by the ‘‘pinning’’ of the Fermi level at the surface, which results in an almost constant field in the undoped region where the QW is located. This type of structure was first used by van Hoof *et al.*[11] to generate well-controlled electric fields in GaAs, which could be accurately measured by photoreflectance spectroscopy. Our intention with the design of these specific QW structures was similar: we wished to place the quantum wells in an internal electric field that was well defined and could be measured accurately by photoreflectivity.

Figure 1 shows typical, low-temperature PL spectra measured on the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$/GaAs QW sample at several exci-

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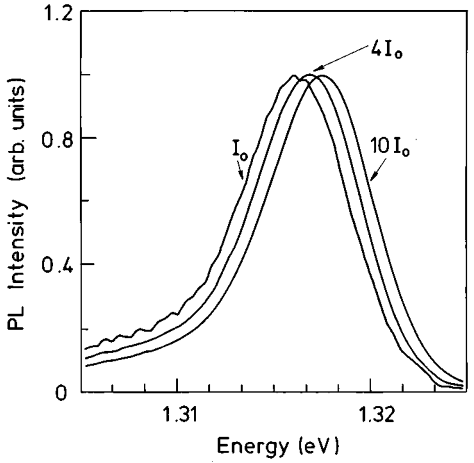

FIG. 1. Low-temperature ($T=12$ K) photoluminescence of an $In_xGa_{1-x}As/GaAs$ single-quantum-well sample at three different excitation intensities. The lowest excitation shown on this figure corresponds to $I_{exc}=100$ μW. The shift of the PL peak to higher energies with increasing excitation powers is clearly visible. The spectra have been normalized.

tation intensities and normalized so that the shift of the peak energy can be easily noticed. The lowest excitation power shown in this figure corresponds to 100 μW. The luminescence from this sample is due to the electron to heavy-hole transition and is characterized by a relatively narrow line (full width at half maximum ≈ 7 meV). What is significant in this figure is the shift of the PL peak position toward high energy (blueshift) as the excitation intensity is *increased*. We observed such a shift, at low excitation powers, for all the samples investigated, although the magnitude of the shift varied from sample to sample. In Fig. 2 we display the PL spectra for the $Al_xGa_{1-x}As/GaAs$ QW's under comparable excitation intensities, which display similar blueshifts for this type ($Al_xGa_{1-x}As/GaAs$) of layer structure. The measured energy shift was found to have a clear nonlinear excitation intensity dependence, as is shown on Fig. 3, where the PL peak position is plotted against the excitation intensity for the $In_xGa_{1-x}As/GaAs$ sample. It can be seen that at low power levels the PL peak positions vary strongly with the excitation intensity, while at higher excitation intensities the PL peak converges towards a saturation value. The intensity dependences of the PL peak positions for the $Al_xGa_{1-x}As/GaAs$ QW's are shown in Fig. 4 and are quite similar to that observed for the $In_xGa_{1-x}As/GaAs$ sample. We shall show below that this characteristic intensity dependence is the result of the screening of the internal electric field by the optically induced carriers, which alters the energy of the QW states. It is important to note that at these very low levels of excitation intensity, concern with optically induced temperature shifts[12] or band-filling effects[13] (Burstein-Moss shift) may be dismissed. The largest photoexcited carrier density in our experiments did not exceed $n=5\times10^{12}$ cm^{-2} (assuming a carrier lifetime of $t=10$ ns). Another notable point is the fact that the free and bound excitonic emission from the bulk GaAs substrate and epilayers did not show this type of nonlinear behavior. In fact, PL

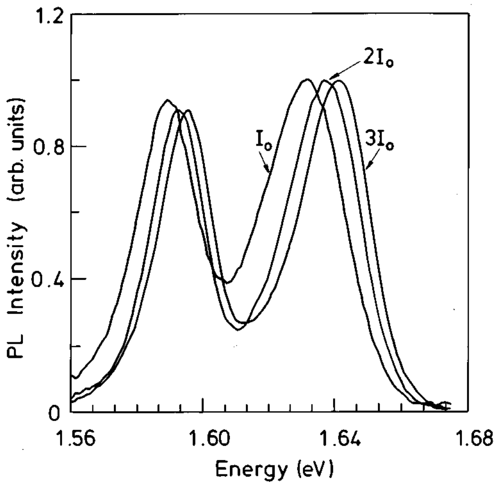

FIG. 2. Low-temperature ($T=12$ K) photoluminescence of an $Al_xGa_{1-x}As/GaAs$ sample containing two single quantum wells, measured at three different excitation intensities. The lowest excitation power corresponds to 100 μW.

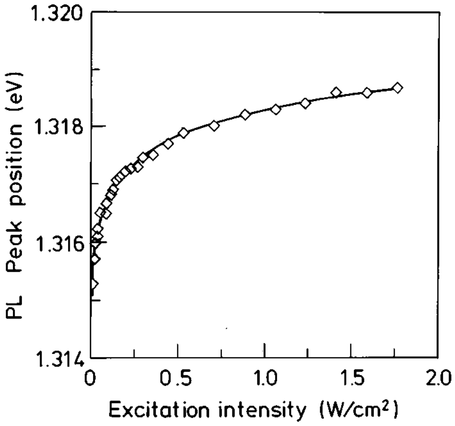

FIG. 3. Excitation power dependence of the PL peak position for the $In_xGa_{1-x}As/GaAs$ QW measured at $T=12$ K. The symbols represent the experimental points, and the full line the theoretical dependence for the given quantum well using Eq. (7).

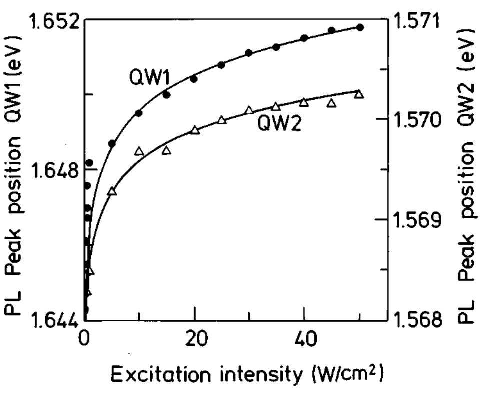

FIG. 4. Excitation power dependence of the PL peak position for the $Al_xGa_{1-x}As/GaAs$ QW's measured at $T=12$ K. The symbols represent the experimental points, and the full lines the theoretical dependence for the given quantum wells.

was not observed from the bulk GaAs until the excitation levels reached approximately 2 mW, after which the PL peak positions were independent of the excitation intensity.

III. DISCUSSION

The effect of an *external* electric field on the PL spectrum was studied by several research groups.[1,14–17] It was found that the PL intensity is quenched by the external electric field, in good agreement with theoretical predictions. The reduction in the PL intensity is expected as the electric-field-induced tilting of the QW energy bands results in the reduction in the overlap of the electron and hole wave functions. Fafard, Fortin, and Merz[17] have shown, for example, that the external field-induced quenching and peak shift may be altered by varying the excitation intensity. They have attributed these effects to a local flattening of the bands in the well region induced by the screening of the carriers trapped in the well.

It is well known that semiconductor heterostructures have *built-in* electric fields caused by the ‘‘pinning’’ of the Fermi levels at the surface and at heterostructure interfaces.[18] The magnitude and penetration of the electric field is determined by the total band bending, the carrier density, and the dielectric constant. In a PL experiment, free carriers are generated close to the surface by the exciting light (typically a laser), and are separated by the built-in surface field, which results in a reduction of the initial electric field. By varying the excitation intensity, the electric field ‘‘felt’’ by the QW’s is modified and consequently, the PL peak position is shifted as described by the QCSE. When the excitation intensity is low, the electric field in the sample is high and, therefore, the energy of the PL peak is significantly shifted from its theoretical (zero field) value. As the excitation intensity is increased, the internal field is reduced, thus the PL peak moves towards the hypothetical ‘‘no field’’ position. As the internal field tends towards the flat-band condition with increasing light intensity, the increase in the PL peak energy is reduced. For most samples containing QW’s, therefore, we expect to see an intensity-dependent PL spectrum, characterized by an increase in the energy of the PL peak with excitation intensity, and a saturation of the PL peak position at high intensities as the surface field tends towards the flat-band condition. In the following, we shall derive an expression for the internal field-induced PL peak shift in terms of the built-in electric field, the excitation intensity, and the temperature.

The magnitude of the surface electric field in a semiconductor can be calculated using the model developed by Kanata *et al.*:[19]

$$F_s=(-2V_s\rho/\epsilon\epsilon_0)^{1/2}, \tag{1}$$

where F_s is the surface electric field, V_s is the equilibrium surface voltage, ρ is the net charge density, ϵ is the low-frequency dielectric constant, and ϵ_0 is the permittivity of free space. Under steady-state conditions, an expression for the surface voltage V_s in the presence of photoinduced current is given by[20]

$$V_s=V_{so}\pm kT/e\ \ln(bN+1), \tag{2}$$

where V_{so} is the surface potential (in the dark), k is the Boltzmann constant, T is the absolute temperature, e is the electronic charge, N is the excitation rate of free carriers per unit area, and b is an intensity-independent constant given by

$$b=\exp(eV_{so}/kT)(e/AT^2), \tag{3}$$

where A is the modified Richardson constant. The surface potential, V_{so}, has been measured by a number of research groups and its value as a function of temperature may be obtained from Ref. 21.

The parameter N is determined by the excitation intensity P:[19]

$$N\approx Pg(1-R)/h\nu, \tag{4}$$

where g is the quantum efficiency (which is of the order of unity), R is the reflectivity coefficient of the material which, for GaAs, we took as $R=0.3$, and $h\nu$ is the photon energy of the exciting light ($h\nu=2$ eV).

Using Eqs. (1)–(4), we can calculate the internal electric field F_s as a function of the incident illumination, P:

$$F_s=\left(F_{so}^2-\frac{F_{so}^2kT\ \ln\{[bPg(1-R)/h\nu]+1\}}{eV_{so}}\right)^{1/2}. \tag{5}$$

Assuming that the electric field is constant over the undoped QW region, it is this electric field which, by way of the QCSE, modifies the PL emission originating from QW’s. As we see from Eq. (5) the internal field is a nonlinear function of the excitation intensity.

The internal field-induced PL peak shift may be calculated using the model developed by Bastard *et al.*,[22] and may be summarized as follows:

$$E_{n/h}=k_{n/h}F^2, \tag{6}$$

where E_n (E_h) is the shift of the electron (hole) energy level due to an electric field F, k_n (k_h) are constants which contain the QW parameters. Using Eqs. (2)–(5), the PL peak energy at a temperature T, in the presence of an internal electric field F_{so} and an illumination intensity P, may be written as

$$E_{\rm PL}=E_0-k_{n/h}\left(F_{so}^2-\frac{F_{so}^2kT\ \ln\{[bPg(1-R)/h\nu]+1\}}{eV_{so}}\right), \tag{7}$$

where $E_{\rm PL}$ is the PL emission energy from the QW under illumination, E_0 is the energy of the PL peak assuming zero internal electric field, and F_{so} is the built-in field under zero illumination intensity.

In Figs. 3 and 4 the PL peak energy, $E_{\rm PL}$ is plotted (unbroken line) as a function of the excitation intensity for the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ and the $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ SQW’s, respectively, and are compared with the experimental results for these samples (symbols). It is important to note that all but one of the parameters required for this calculation are known or may be obtained from the literature. The only fitting parameter used in the calculation was F_{so}, the built-in electric field for the given samples. This parameter, however, may be measured independently, on the same samples, at the same temperature using PR spectroscopy. The values obtained for the F_{so} from the PL fitting of Eq. (7) were 44 kV/cm for the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ sample and 157 kV/cm for the $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ sample at the given temperature (T

$=12$ K). We should remember that the parameter F_{so} represents the *maximum* value of the internal field, that is, the field in the "dark."

In order to judge the accuracy of the electric field values obtained from the PL measurements, we have measured the photoreflectance spectra of the same samples, at the same temperatures and determined the internal field from the observed Franz-Keldysh oscillations[23] (FKO). This method relies on the known dependence of the FKO maxima/minima (E_n) on the internal electric field. For each sample, we plotted $4/3\pi(E_n-E_g)^{3/2}$ against the index n of the oscillation, and determined the built-in field from the slope of the plot, using

$$n\pi=\frac{2}{3}\left(\frac{E_n-E_g}{h\Omega}\right)^{3/2}$$

relationship, where the Ω parameter depends on the internal field. A typical plot for the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ sample is shown in Fig. 5. For the sample shown in the figure, the value of F_{so} as determined from the plot is 40 kV/cm is in excellent agreement with the value determined from PL. The agreement between the values for the internal field obtained from PR and PL techniques for all the samples studied was good.

In conclusion, we have measured the PL spectra of several III-V QW's as a function of the excitation intensity and have compared the experimental data with calculations based on the QCSE due to the internal electric field. The only fitting parameter in the calculation was the value of the internal field, which was compared with field values obtained from the photoreflectance spectroscopy. The two types of measurements gave consistent results.

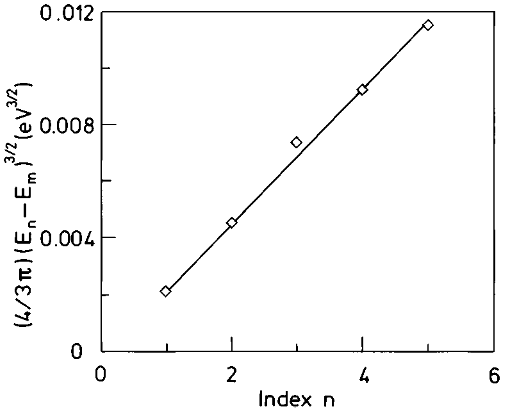

FIG. 5. Plot used to determine internal field from photoreflectance data. Symbols represent maxima and minima of the Franz-Keldysh oscillations and the full line is the best fit, the slope of which is proportional to the internal electric field.

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