OPTICAL SPECTROSCOPY OF GaAs/GaAlAs QUANTUM WELLS UNDER AN EXTERNAL ELECTRIC FIELD

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We have studied by means of low temperature photoluminescence (PL) and photocurrent spectroscopy the effects of an external electric field on the excitons in GaAs quantum wells confined between GaAlAs. Increasing the field causes a Stark shift of the excitons toward lower energies with a simultaneous quenching in the PL intensity. At moderate fields, we find very good agreement (better than 0.5 meV) between the light- and heavy-hole exciton energies obtained by PL and photocurrent measurements. A significant deviation in energy of the PL relative to the photocurrent is observed at high fields, manifesting the increase in the contributions of impurity-bound excitons to the PL lineshape. A detailed PL study of the Stark shift as a function of well thickness has also been performed. The results show an increasing Stark shift with increasing well thickness, amounting to 110 meV for a 230 Å-wide well at a field of 10^5 V/cm . For very wide wells (~ 1000 Å) the behavior of bulk GaAs is recovered: the excitons become ionized before large Stark shifts can be observed. Variational calculations have been carried out and shown to account for the experimental observations of both the Stark shift and the quenching of the PL. In this light, we will discuss the mechanisms governing the optical properties of quantum wells under an external electric field.

1.-INTRODUCTION

Considerable effort is presently devoted to the study of the effects of an external electric field on the optical properties of GaAs/GaAlAs quantum wells (see, e.g., other 5 papers related to this topic in this Conference). The first studies were performed by the technique of photoluminescence (PL) spectroscopy[1] on thin wells, where the confinement effects are large. A strong quenching of the PL, together with a small Stark shift of the excitons in the wells, was observed. The fields have been applied not only perpendicularly to the layers but also in the parallel configuration.[2] In the latter case, the situation is similar to bulk GaAs, where Stark shifts are hardly observable, because of strong lifetime reduction and impact ionization of the excitons.[3] In the longitudinal configuration, the Stark shift increases with increasing well thickness, until the wells become so wide that the confinement effects are negligible and the situation of bulk GaAs is recovered. The PL quenching has been attributed to two coexisting mechanisms: spatial charge separation and field-induced tunneling of photogenerated carriers out of the well.[1,4] Time resolved PL measurements have helped to distinguish between these two mechanisms.[5,6] The decrease of the lifetime for thin wells could be explained with the Fowler-Nordheim tunneling model.[5,6] For wider wells, and in the case where non-radiative recombination processes are negligible, the lifetime increase could be satisfactorily explained by the polarization of the electron and hole wavefunctions.[6] We have found that, for wide wells, the PL quenching is consistent with calculations of the reduction of wavefunction overlap between electrons and holes, as they are separated by the field.

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The combination of PL and photocurrent spectroscopy (PCS) gives important information about the role of the impurities in the PL spectra and helps to clarify the Stokes shifts between absorption and recombination processes reported in the literature.[7-9] Our measurements show that with increasing electric field the PL shifts to lower energies with respect to the absorption, as a result of the electrons and holes being skewed toward the interfaces, where more impurities and defects are present.

2.-EXPERIMENTAL

Five heterostructures, grown by molecular beam epitaxy on (100)-oriented n^+−GaAs substrates, where investigated. Four samples, with well thickness of 130Å, 160Å, 230Å and 400Å, consisted of 5 periods of alternate GaAs and $Ga_{0.65}Al_{0.35}As$ layers, with barrier thickness of 250Å. The last sample was a 1000Å-thick single GaAs well sandwiched between $Ga_{0.65}Al_{0.35}As$ layers. A semitransparent Ni Schottky barrier was evaporated on the top of the cladding layers to apply the external electric field. Conventional PL measurements were obtained at 5K with the samples mounted on a variable temperature cryostat. The luminescence was excited either with the 5145-Å line of an Ar^+ laser or with the 6471-Å line of a Kr^+ laser. Power densities below 10mw/cm² were used to avoid excessive carrier generation. The PCS spectra were taken at 10K using the light of a tungsten lamp dispersed from a grating monochromator. The light-induced current was detected using standard lock-in techniques. The highest electric fields applied to the samples were of the order of 10^5V/cm .

3.-RESULTS AND DISCUSSION

Photoluminescence and photocurrent spectra of the sample with 160Å-wide wells for six selected voltages (fields between ~0 V/cm and ~77 kV/cm) are shown in Fig. 1. Three different structures can be seen at the largest voltage (0.8 V) in the PL spectrum. By comparison with the PC spectrum and with envelope function calculations,[10] we identify the first two peaks at the high energy side as the 1e→1lh and 1e→1hh free excitons, respectively (e means electron, and lh and hh, light and heavy hole, respectively). The rapid decrease of the third structure with increasing temperature, as well as its saturation with increasing light power, allows us to assign it to a bound exciton (D,X).

Three main effects can be observed in the PL as the electric field is increased (i.e., the external voltage decreased): a red shift of all the structures, a decrease of the total luminescence efficiency and a merging of the free and bound excitons, so that they cannot be resolved separately. The change of the relative intensities of free and bound excitons is somehow sample dependent and shows a complicated behavior with increasing electric field.[11] The energies of the 1e→1hh

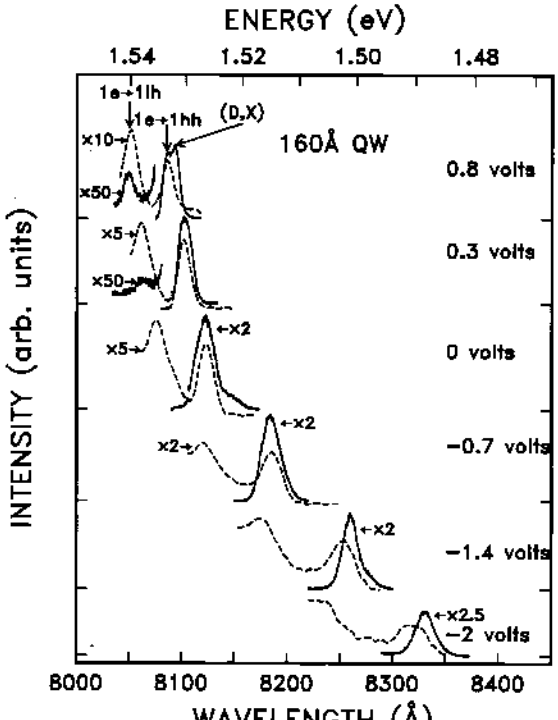

Fig. 1: Low temperature (T = 5 K) PL (solid lines) and (T = 10 K) photocurrent (dashed lines) spectra of a 160Å GaAs-$Ga_{0.65}Al_{0.35}As$ quantum well at different external voltages. Scaling factors are indicated (when not present, 1 is assumed). Total applied voltage through the sample is indicated on the right side of the spectra. The free light-hole, heavy-hole and donor-bound excitons are marked with 1e→1lh, 1e→1hh and (D,X), respectively. The spectra are displaced vertically for clarity.

excitons found in PL and PCS agree to within 0.3 meV up to fields of ~50 kV/cm. Notice also the good agreement in line-width in both kinds of measurements. An increasing Stokes shift between the PL and PCS, amounting to 2.5 meV at -2 V, is observed at higher fields. This difference can be explained as the result of enhanced bound-exciton luminescence as the carriers are skewed closer to the interfaces, where more impurities and defects exist. The increase in line-width with increasing field hinder the resolution of free and bound recombination at the highest fields. However, based on the temperature dependence of the PL at different voltages, which shows a decrease in intensity at the low energy side of the spectra as well as a pseudoshift to *higher* energies with a moderate increase of temperature (up to 20K),

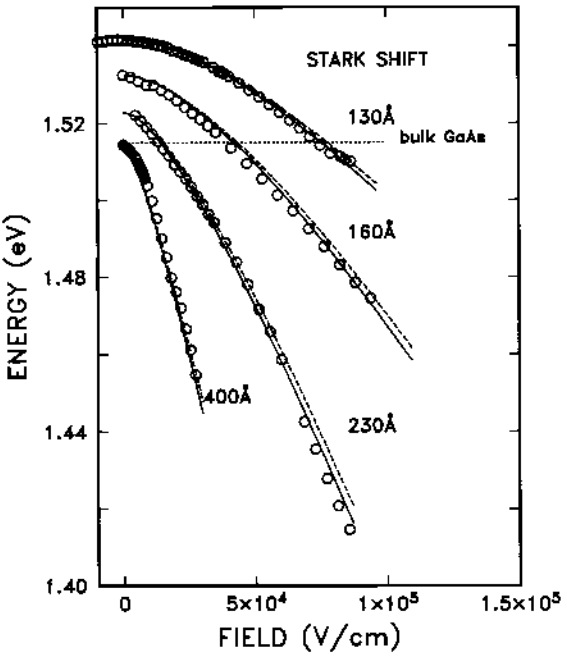

Fig. 2: Points: experimental exciton energies as a function of electric field strength for different well thickness. Solid and dashed lines: theoretical shift obtained with numerical calculations, using the matrix formalism described in the text, and variational results, respectively. The horizontal dotted line indicates the energy of the free exciton in bulk GaAs.

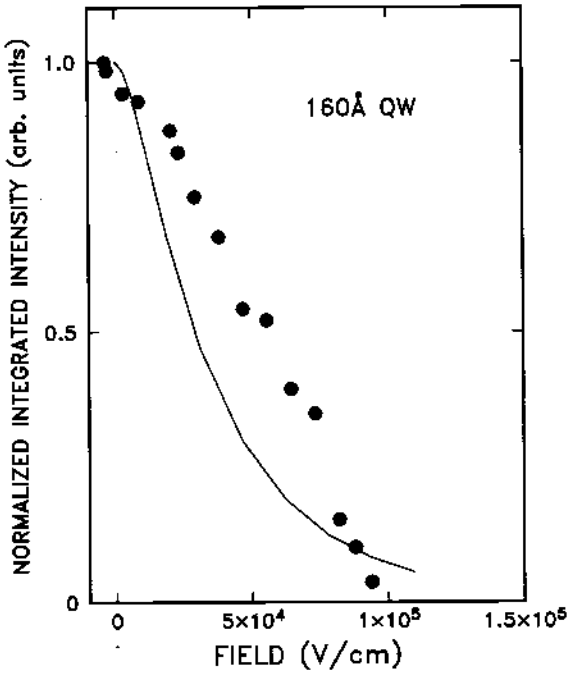

Fig. 3: Integrated photoluminescence intensity versus electric field, normalized with respect to flat band condition, for the same sample of Fig. 1. The solid line depicts the change in the overlap between electron and hole wavefunctions obtained by variational calculations.

we infer that both processes are present in the recombination mechanism. The energy difference of ~ 2.5 meV, similar to the difference between free and acceptor-bound excitons in bulk GaAs, suggests the enhancement of this channel of recombination with increasing field. The faster shift of the holes toward the interfaces[12] also sustains this hypothesis.

Figure 2 shows the Stark shifts of the dominant peak in the PL spectra for four samples versus electric field. An increasing Stark shift with increasing well thickness, for the same electric field, as expected from perturbation-theory arguments,[13] is clearly observed in this figure. This causes the energy of the excitonic PL to be less than that of the bulk-GaAs exciton (its energy is indicated by a dotted line in the figure). The results of two different calculations of the 1e→1hh energies are also depicted in Fig. 2. The dashed lines correspond to variational calculations,[12] while the solid lines represent the exact numerical solution using the matrix formalism of Smith,[14] described elsewhere.[11] The two calculations are in good agreement at low fields. In both calculations the effect of the field is more pronounced for holes than for electrons, because of the heavier effective mass and the lower barrier for the holes. The agreement between theory and experiment is satisfactory, with an uncertainty of ~ 10% from that of the experimental determination of the field strength. The large Stark shifts, with the PL below the band gap of bulk GaAs, cause the recombination to be indirect in real space, as the electrons and holes are confined in quasitriangular wells at opposite interfaces. It is also interesting to note that for wide wells (~400Å), larger than the three-dimensional exciton diameter (~300Å), the electron-hole interaction remains strong even though the Stark shift greatly exceeds the exciton binding energy.

The decrease of the integrated PL intensity, for the same sample of Fig. 1, is shown in Fig. 3 versus electric field. The change of the overlap integral between electron and hole wavefunctions, obtained from the variational calculations, is also displayed as a solid line. Two different mechanisms, a decrease of the overlap integral and an increase of the nonradiative processes, have been proposed to explain the quenching of the PL. The field-induced polarization of the electron and hole wavefunctions toward opposite directions in the wells results in a decrease of the

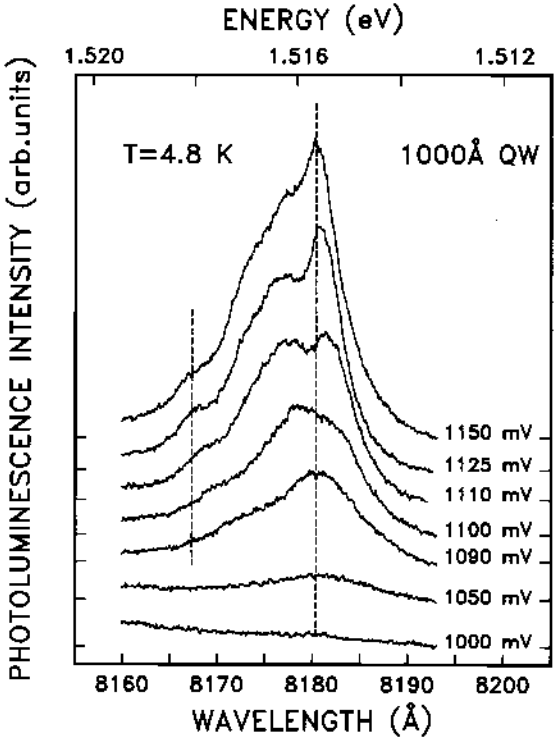

Fig. 4: Low temperature (T = 4.8 K) PL spectra of a 1000Å-wide well for different forward bias voltages. The PL rides on a background from the n^+—GaAs substrate not shown in the figure. The vertical lines are shown as an aid to observe the small shift of the different excitons.

overlap integral, and therefore a decrease in the oscillator strength and the PL intensity. The observed quenching of the PL indicates that nonradiative channels are present in our samples. Our measurements do not allow us to discern between either a decrease of the nonradiative recombination time or an increase in the radiative recombination time with increasing field as the cause of the decrease in the PL efficiency. However, the good agreement between our experimental data and the calculations strongly indicates that the decrease in oscillator strength is the main mechanism for the PL quenching in thick quantum wells ($\geq$ 100Å), in agreement with recent time resolved measurements.[6]

With increasing well width, the PL spectra evolve to that of bulk GaAs, where generally bound excitons dominate over free excitons in recombination. For very wide wells it is also expected that the situation of bulk GaAs is recovered, where impact ionization of the excitons takes place before any Stark shift is detected.[3] This situation is clearly seen in Fig. 4, where PL spectra from a 1000Å-wide well are shown for different external voltages. At a voltage of 1V, corresponding to $\sim 5 \times 10^3$ V/cm, the PL is completely quenched. A small shift of the order of 1 meV is still observable. The quenching of the bound excitons is faster than the quenching of the free excitons, similar to the case of bulk GaAs.[3] However, large fields, compared to the 10 V/cm reported in Ref. 3, are still necessary in this case to completely suppress the PL.

In summary, we have shown the strong dependence of Stark shifts on well thickness. A satisfactory agreement between the observed quenching of the PL and the decrease in the overlap integral between electrons and holes has been obtained. It has been found that confinement effects are still strong in the electron-hole interaction for well widths larger than the three-dimensional exciton size. For very wide wells the situation of bulk GaAs, where Stark shifts cannot be observed because of impact ionization of the excitons, is recovered. Finally, the combination of PL and PCS techniques allowed us to confirm the role that the impurities play in the Stokes shift between absorption and PL measurements.

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