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Measured transition from two-dimensional to three-dimensional electroabsorption as quantum-well barriers are lowered

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Electric-field-dependent measurements of photocurrent spectra of GaAs-$Al_xGa_{1-x}As$ multiple quantum wells are made for $0.0013 < x < 0.0250$. Strong room-temperature exciton enhancement and quantum-confined Stark effect are observed for x as low as 0.0082. Below that x value behavior is more bulklike. It is at this x value that theory predicts that the holes should become delocalized. This study indicates that a three-dimensional electron and a two-dimensional hole results in a two-dimensional exciton in this system.

Recently we reported the observation of strong room-temperature excitons in GaAs-$Al_xGa_{1-x}As$ multiple quantum wells (MQW's) for x as low as 0.02.[1] This observation has important technological ramifications for optoelectronic devices since the escape time of photoexcited carriers in MQW's with $x \leq 0.04$ is equivalent to bulk GaAs,[2] which is 2 orders of magnitude faster than MQW's with $x=0.2$.[3] This rapid escape time allows modulators using "shallow" MQW's to operate at optical intensities several times higher than "standard" MQW's.[4] In addition these shallow MQW's displayed rapid ionization of the exciton with field resulting in large electroabsorption.[1] This results in modulators with superior performance in self-electro-optic effect devices.[5]

The existence of excitonic confinement effects with such low barriers is puzzling since the electron wave functions should be delocalized when the barrier is so low. Very recently we observed Stark ladder formation (commonly observed in short-period deep superlattices[6,7]) in a $x=0.02$ MQW with 100-Å wells and 40-Å barriers.[8] This suggests that field-induced localization may be playing a role in these samples. However, no such effects have been observed in samples with wider barriers at $x=0.02$, and the exciton in these samples remains strong and unchanged at the lowest field applicable, even though the wave functions should be delocalized according to standard theory.

Here we present the completion of the empirical study started in Ref. 1 and observe the transition to bulklike behavior as the barriers are lowered in MQW. Also, we show theoretically that this transition occurs when the holes become delocalized, indicating that the combination of a three-dimensional (3D) electron and a two-dimensional (2D) hole results in a 2D exciton in this system. In Ref. 1 as the barrier was lowered to $x=0.02$ the exciton remained strong and it redshifted with field, which is commonly referred to as the quantum-confined Stark effect.[9] The bulk GaAs presented there had a weak exciton which had an apparent blueshift with field which is commonly observed and associated with field-induced broadening. Here we explore the region in between these barrier heights ($0.0250 \leq x \leq 0.0013$). The transition between 2D and 3D behavior, if one uses the metric of redshifting versus blueshifting, occurs between $x=0.0082$ and 0.0025.

The structure and molecular beam epitaxy (MBE) growth conditions of the *p-i*(MQW)-*n* samples presented here are identical to those in Ref. 1, except for the different mole fractions of the barriers. Five samples are presented here, with $x=0.0250$, 0.0160, 0.0082, 0.0025, and 0.0013. The wells and barriers are each 100 Å wide as in Ref. 1. The barriers are lowered in successive samples by reducing the temperature of the Al crucible in the MBE chamber along an extrapolated curve.

The above x values are determined by a combination of x ray and secondary ion mass spectroscopy (SIMS) measurements on the samples after growth (they differ only slightly from the design values). X-ray scattering off the $x=0.0250$ sample versus angle is shown in Fig. 1. The peak due to the substrate is marked on the figure. The peak on the left-hand side is due to pendulsulang oscillations between the $x=0.3$ *p* and *n* regions of the sample. In between these is a bump which results from scattering off the MQW region, which has a slightly larger lattice

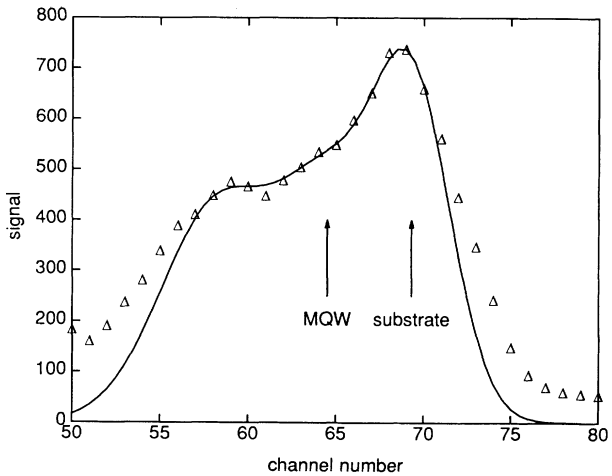

FIG. 1. X-ray scattering signal (arb. units) of the MQW sample with $x=0.0250$ Al barrier mole fraction vs channel number (angle). The solid curve is a fit based on Gaussians. The arrows mark the peak of the MQW and the GaAs substrate signal. $x=0.0250$ is obtained from the displacement of the MQW peak.

constant than GaAs. The solid curve is a fit based on 3 Gaussian peaks. The position of the MQW peak is determined from this fit and is marked. Based upon its displacement from the substrate, and assuming that the wells and barriers have equal thickness (which we checked in the SIMS measurements), and upon known dependence of lattice constant of Al fraction, we obtain $x=0.0250$.

As can be observed, at $x=0.0250$, the MQW peak is not easily distinguishable from the substrate. For lower values of x, this measurement would yield dubious results. Therefore SIMS measurements were performed on all five samples. By comparing the relative values of the Al^+ signal to that of the $x=0.0250$ sample, we obtained the x values quoted. Displayed in Fig. 2 are the SIMS values (measured during sputtering of the barrier). The precipitous drop of the Al^+ signal can be seen. The AsO^+ and GaO^+ signals were also measured as a control. The relatively unchanged values of these signals testifies for the validity of this approach. In practice, we compared the Al^+ signals normalized to the AsO^+ signals to obtain our x values.

The samples were processed into *p-i-n* diodes as in Ref. 1 and photocurrent spectra taken with a lamp-monochromator at different biases. Shown in Fig. 3 are sets of spectra taken at biases of +0.8, +0.4, 0, −0.4, −0.8, and −1.2 V applied (positive refers to forward bias) at room temperature. For all the samples, the +0.8-V spectrum had to be normalized to coincide with the other spectra well above the band gap, since a slight (about 30%) reduction in photocurrent collection efficiency occurred at this low field.

At $x=0.0250$, behavior is similar to that of the $x=0.02$ sample of Ref. 1. A strong exciton is observed that redshifts and decays due to ionization as field is applied. The pattern of Ref. 1 was that the field required to induce decay decreased as x decreased. This holds true here, with the $x=0.0160$ sample displaying more rapid decay with field. A difference is that the excitonic feature changed little from $x=0.06$ to 0.02, whereas here it has reduced

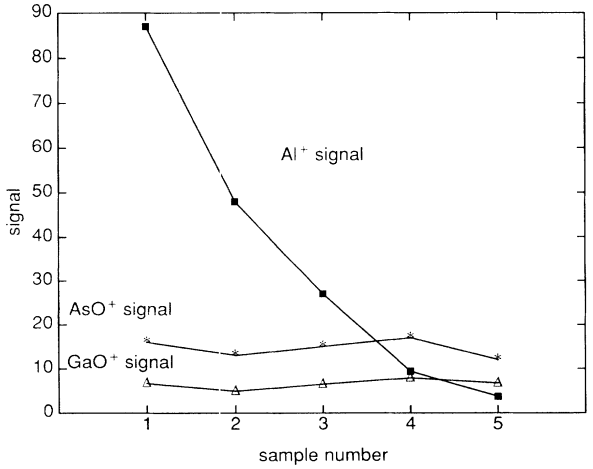

FIG. 2. Secondary ion mass spectroscopy signals for the five samples (No. 1 is the $x=0.0250$ sample). The mole fractions of samples 2–5 are obtained by comparing the Al^+ signals (normalized to the AsO^+ signal).

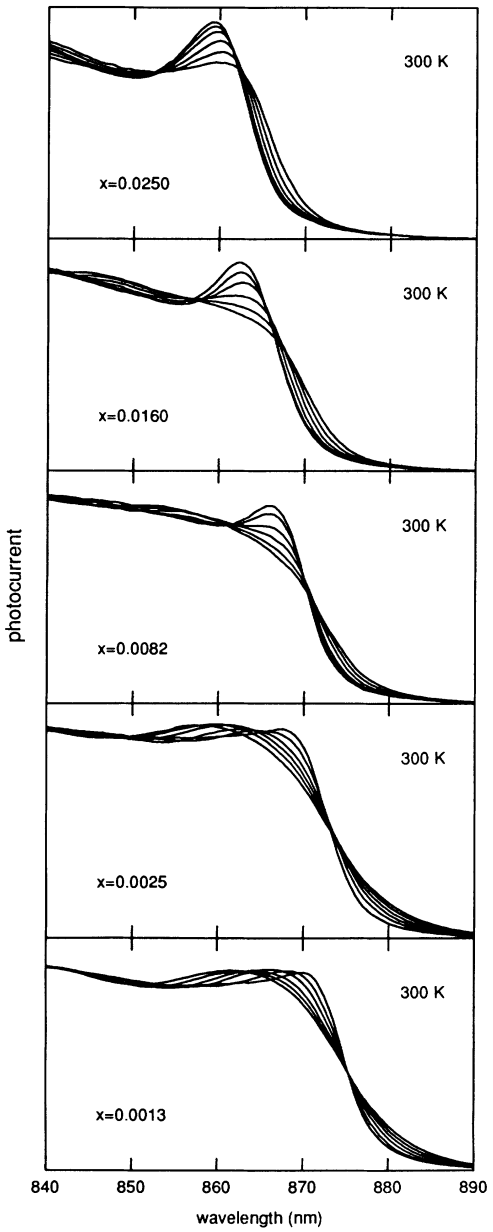

FIG. 3. Photocurrent spectra taken at +0.8, +0.4, 0, −0.4, −0.8, and −1.2 V applied bias for each sample at 300 K. For all the samples the lowest field spectrum has the sharpest edge.

prominence in the $x=0.016$ sample. The $x=0.0082$ sample shows even less prominence and much more broadening with field. By $x=0.0025$, the spectra appear very much like bulk, with the electroabsorption dominated by broadening. However, there is still a minor difference: The spectra of different biases do not all cross at the same wavelength, as is observed for bulk GaAs. For the $x=0.0013$ sample, the spectra appear identical to bulk GaAs, with all the spectra crossing at the same wavelength.

The exciton clearly redshifts with increasing field for $x=0.0160$. For the $x=0.0082$ sample the peak appears stationary with field. If one uses the definition of redshift-

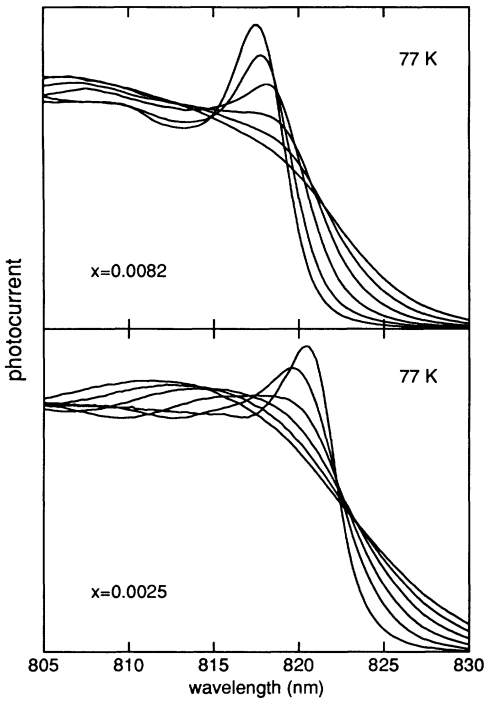

FIG. 4. Same as Fig. 3, but at 77 K. The $x=0.0082$ sample's exciton redshifts with field, whereas the $x=0.0025$ sample's exciton blueshifts, marking the delineation between two-dimensional and three-dimensional behavior.

ing versus blueshifting to describe 2D versus 3D behavior, then for room temperature the crossover occurs at $x=0.0082$. However, the broadness of the peaks at room temperature complicates this observation. Therefore we reduced the temperature to 77 K and repeated the measurement. This is shown in Fig. 4 for the $x=0.0082$ and 0.0025 samples (same biases as before). The peak can clearly be seen to redshift in the $x=0.0082$ sample's spectra, whereas it blueshifts in the $x=0.0025$ sample's spectra. Therefore the crossover from 2D to 3D behavior occurs at about one-half of a percent of Al mole fraction in the barrier. Other differences exist between these two samples. In the lowest-field spectrum for the $x=0.0082$ sample, a dip exists at an energy just higher than the exciton's. For the $x=0.0025$ sample, this dip does not exist. This could be due to a higher exciton binding energy in the $x=0.0082$ sample, which displaces it from the continuum states.

The blueshifting nature of bulklike samples does not seem to have been discussed much in past literature. Indeed, theoretically a small redshift should occur.[10] The authors have considered the blueshift observed at room temperature to be an illusion caused by the broadening of the exciton. However, for the $x=0.0025$ sample in Fig. 4 it does not seem that the movement of the peak can be attributed simply to broadening. Indeed, a second peak can be seen in the +0.4-V spectrum near 815 nm. The two peaks have equal height in the 0-V spectrum, giving it a "flat-top" appearance. The behavior in bulk samples may require further study.

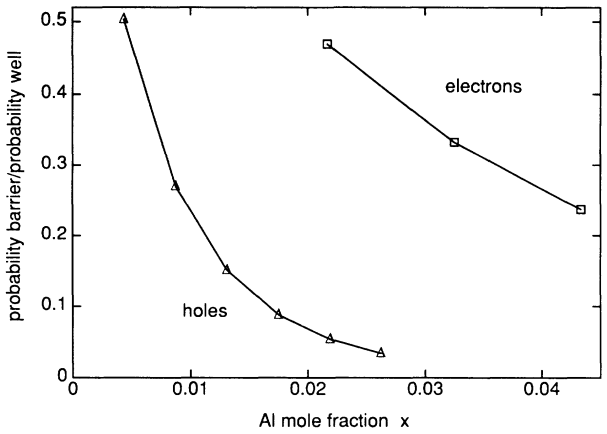

FIG. 5. Calculated ratio of the probabilities of finding a carrier in the center of the barrier/well of a GaAs/$Al_xGa_{1-x}As$ MQW for the lowest eigenstate as a function of x at flatband. The observed transition to three-dimensional behavior occurs when the holes delocalize even though the electrons are already delocalized at higher x.

As mentioned, the electron wave functions are delocalized at x values much higher than studied here according to standard models. In Fig. 5 we plot the theoretical ratio of the probabilities of finding a carrier in the center of a barrier over finding it in the center of a well versus x, for a 100/100-Å GaAs-$Al_xGa_{1-x}As$ MQW at zero field (we assume a 60:40 split for the conduction-band:valence-band offset). This ratio displays the degree to which the wave functions are delocalized. For electrons, this ratio is already high (compared to unity) at $x=0.05$, indicating large delocalization. For holes, the ratio remains small until x falls below $x=0.01$. This roughly correlates to the x value at which we observe the transition from 2D to 3D behavior. We conclude then that the combination of a 2D hole and a 3D electron results in a 2D exciton in this system, presumably due to a modification of the potential experienced by the electron because of the presence of the hole. This argument, if one exchanges electrons and holes, has been used to explain excitons observed in II-VI MQW's with low valence-band offsets.[11] However, the situation here is much more exacerbated since the 3D carrier in II-VI's are (heavier) holes. Also, in GaAs/$Al_{0.3}Ga_{0.7}As$ 30/30-Å short-period superlattices, when the electron becomes delocalized (as the electric field is lowered), the exciton is observed to become bulklike,[6,7] even though the hole is still localized. Therefore, in that system the combination of a 3D electron and 2D hole results in a 3D exciton, in contrast to what we observe in the shallow system. Clearly, new theoretical studies are necessary to resolve this issue.

In conclusion, we have observed the transition from 2D to 3D electroabsorption behavior in GaAs-$Al_xGa_{1-x}As$ samples as x is lowered below 2%. The transition occurs near $x=\frac{1}{2}\%$, if one uses the definition of redshifting versus blueshifting behavior with field.

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