Ultrafast-lifetime quantum wells with sharp exciton spectra

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Sharp quantum-confined excitons in semi-insulating low-temperature-growth AlAs/GaAs quantum wells with 15 ps carrier lifetimes are demonstrated. High-quality well-barrier interfaces can be grown at low substrate temperatures and annealed up to temperatures of 700 °C, beyond which interface mixing broadens the exciton transitions. Electroabsorption from the quantum-confined Stark effect in as-grown *p-i-n* modulators approaches 10 000 cm^{-1}, which is comparable to traditional high-temperature growth quantum wells. The low-temperature growth quantum well structures eliminate the need for postgrowth processing, such as ion implantation for photorefractive quantum wells, ultrafast saturable absorption, or electro-optic sampling applications. © *1995 American Institute of Physics.*

The dual goals of achieving ultrafast lifetimes in quantum wells along with sharp quantum-confined excitons have been mutually exclusive. Ultrafast lifetimes usually require high defect densities, which broaden the excitonic spectrum. In addition, standard methods to produce semi-insulating quantum wells, such as Cr-doping[1] or proton implantation,[2] are time consuming, may require postgrowth processing, and can have ill side effects, such as Cr contamination of subsequent growths. Low-temperature growth (LTG) GaAs has attracted significant recent attention as a marketable new ultrafast subpicosecond photoconductor.[3–5] This ultrafast property would be important for photorefractive,[6] electro-optic sampling[7] and saturable absorption applications if it could be combined with sharp quantum-confined excitons in multiple quantum wells (MQWs). However, earlier investigators found that excitons in LTG AlGaAs/GaAs MQWs with excess arsenic and wide barriers were too severely broadened to be useful.[8]

In this letter, we demonstrate simultaneous ultrafast lifetimes and sharp quantum-confined excitons in AlAs/GaAs quantum wells grown at low substrate temperatures. Barriers of AlAs between the wells[9] make sharp excitons in LTG quantum wells possible. The thin barriers, and the use of As_4 fluxes instead of As_2 during molecular beam epitaxy, reduces the amount of excess arsenic deposited into the wells from the barriers.[10] During weak anneals, excess arsenic collects into precipitates in the wells, providing efficient recombination sites but producing an inhomogeneous environment for the excitons. By reducing the amount of excess arsenic, the inhomogeneous exciton broadening is reduced. Furthermore, the use of AlAs instead of AlGaAs appears to reduce the amount of interface mixing at the low growth temperature.

The samples used in our experiments were grown by molecular beam epitaxy (MBE) using As_4 on substrates at a low growth temperature, nominally of 310 °C. The structures were LTG MBE AlAs/GaAs *p-i-n* transmission modulators grown on n^+ GaAs substrates. Contact and stop-etch layers of *n*-type materials were grown on an n^+ GaAs substrate at 600 °C. This was followed by a LTG (310 °C) MQW layer consisting of a 150 period superlattice of 100 Å GaAs wells and 35 Å AlAs barriers. The LTG (310 °C) results in ≈0.2% excess arsenic in the MQW. A 2000 Å *p*-$Al_{0.3}Ga_{0.7}As$ (1×10^{18} cm^{-3}) layer followed by a 2000 Å top *p*-GaAs (1×10^{19} cm^{-3}) were grown at 450 °C on top of the LTG layers. The 450 °C growth temperature for the *p*-doped layers acts as a weak *in situ* anneal of the previously grown LTG layers and results in the formation of As precipitates in the MQW region.[11,12] The arsenic clusters deplete free carriers from the surrounding material, rendering it high resistivity.[13] Gold contacts were made to the top *p*-GaAs. The samples were epoxied to glass to remove the substrate using standard techniques.[14] Another gold contact was made to the exposed *n*-$Al_{0.5}Ga_{0.5}As$ stop-etch layer after substrate removal. Electro-optic characterization was performed with the device reverse biased using a monochromator with an incoherent tungsten lamp.

Several samples were subjected to rapid thermal anneal at several temperatures for 30 s to control the size and spacing of the As precipitates using precipitate engineering.[12] The process of annealing, which forms As clusters, converts the heavily defected LTG GaAs into a high-quality GaAs matrix. Therefore, sharper excitonic features are normally observed for higher anneal temperatures as the LTG material approaches stoichiometric GaAs. In our absorption spectra, shown in Fig. 1, we observe the opposite trend. Our sample with the *in situ* anneal of 24 min at 450 °C during the growth of the top 2000 Å *p*-GaAs epilayer gave the sharpest quantum-confined excitons without additional postgrowth anneal. The sharp excitonic features get washed out at higher postgrowth anneal temperatures, caused by progressive interface intermixing.

The zero-field absorbance versus wavelength, shown in Fig. 1 for all samples, includes the sample without postgrowth anneal. Sharp quantum-confined exciton spectra are

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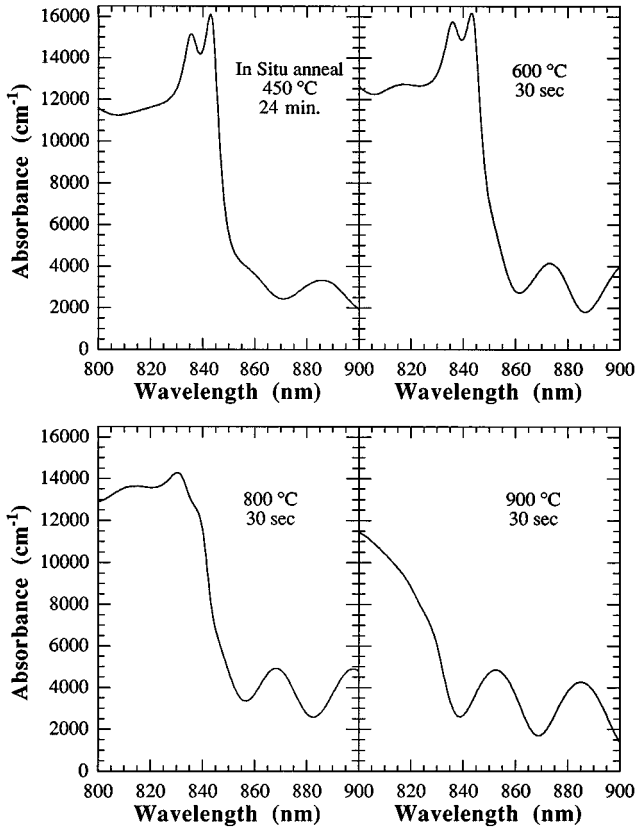

FIG. 1. Zero-field absorbance vs wavelength showing the sharpest excitonic features for the sample with *in situ* anneal of 450 °C for 24 min during growth of the top 2000 Å *p*-GaAs epilayer. All other samples experienced a postgrowth anneal in addition to the *in situ* anneal.

observed in this sample and in samples annealed at low temperature. The excitonic features are washed out for postgrowth anneal temperatures higher than 700°C, caused by interface roughening. Fabry–Perot fringes are evident in the spectra because the samples did not have an antireflection coating, but the fringes do not obscure the physical trends of our materials. For the *in situ* anneal, the full-width at half maximum of the heavy hole exciton is estimated to be 10 meV. We compared this against an identical quantum-well structure grown at standard temperature that has a full-width at half maximum of 6 meV. Therefore, in the LTG quantum wells the excitons are broadened by 4 meV.

The differential transmission versus wavelength is shown in Fig. 2 for all the samples. AC fields with a peak modulation of 10 V/μm with a dc reverse bias of 5 V/μm were applied to the sample. Sharp excitonic spectra with a differential absorption of $\Delta\alpha \approx 9700\ \mathrm{cm}^{-1}$ are observed in the sample that experienced the *in situ* 450 °C anneal. This value is comparable to high-quality MQWs grown at standard temperatures.[15] AC fields are used because LTG materials have short depletion widths. Therefore the *i*-region in these devices is not completely depleted, and high frequencies are needed to capacitively couple through the depletion layers. The differential transmission is normalized with the zero field transmission. The differential transmission below the band gap exceeds negative unity, indicating that the device is experiencing a forward bias during part of the voltage

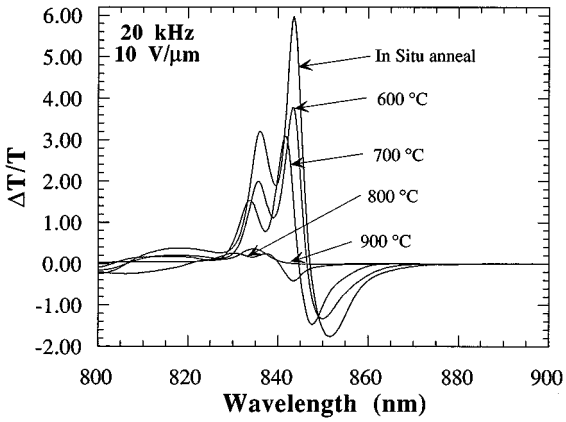

FIG. 2. Differential electroabsorption vs wavelength for all samples at 20 kHz. Note the sharp decrease in the differential absorption for the 800 and 900 °C anneals is consistent with Fig. 1.

cycle. This results in an excitonic blue shift and sharper excitons during that part of the cycle.

The carrier lifetimes in the LTG quantum wells were measured using a pump-probe technique with a 100 fs Ti:sapphire laser at a repetition rate of 1.8 kHz, tuned at 825 nm, with a photon energy slightly above the heavy-hole exciton energy with a transition at 840 nm. The normalized saturated absorption as a function of probe delay is shown in Fig. 3 for a sample annealed after growth at 600 °C for 30 s. The recombination lifetime for electrons in this sample is 15 ps. The response of a traditional high-temperature growth $Al_{0.1}Ga_{0.9}As/GaAs$ quantum well structure is shown in the figure for comparison. The high-temperature growth sample was proton implanted with a dose of $1\times10^{12}\ \mathrm{cm}^{-1}$ and energy of 160 keV. This dose is near the limit for implants before significant broadening of the excitonic transition occurs. This high-temperature growth sample has an electron lifetime of 160 ps, consistent with previous measurements for this implant dose.[16,17] The LTG quantum wells have much faster lifetimes while retaining sharp excitonic transitions. In the future, lifetimes in LTG quantum wells may be adjusted to have even faster lifetimes approaching 1 ps using appropriately engineered structures.

The availability of semi-insulating multiple quantum wells that have fast carrier lifetimes and sharp quantum-confined excitons will be especially helpful for photorefractive and saturable absorption applications. Transverse-field photorefractive quantum wells[18] rely explicitly on semi-insulating quantum wells with sharp excitons to ensure large electroabsorption and large photorefractive sensitivity. Quantum well saturable absorbers rely on ultrafast carrier lifetimes and also on sharp excitons. Sharp excitons are required because peak electroabsorption and saturated absorption depend inversely on the square of the linewidth for weak perturbations. Therefore, relatively small broadening of the exciton transition can lead to large degradation of the electro-optic and saturable absorption response. In addition, the ultrafast lifetimes in the LTG materials are ideally suited in photorefractive applications for high spatial resolution during two-wave[19] and four-wave mixing.

We speculate that the annealed LTG quantum wells are

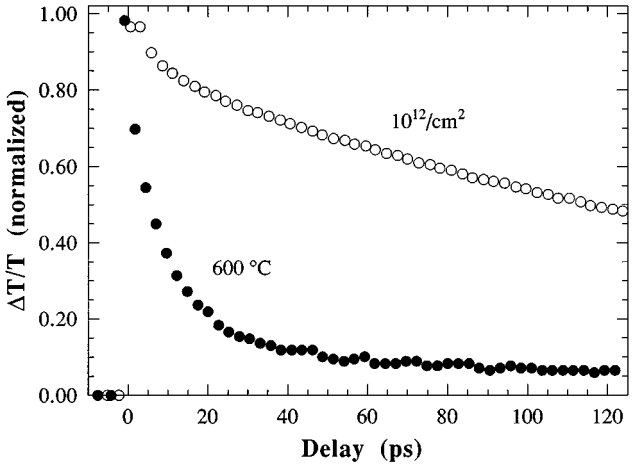

FIG. 3. The normalized saturated absorption as a function of probe delay for the 600 °C annealed sample. Normal high-temperature growth MQW results are also shown for a proton implant dose of $1\times10^{12}/\text{cm}^2$. A mode locked Ti:sapphire laser with a pulsewidth of 100 fs at a repetition rate of 1.8 kHz was used.

able to retain sharp excitonic features while achieving fast recombination times because of the different recombination and compensation properties of arsenic precipitates compared with point defects. Point defects induced by radiation damage are typically self-compensated[20] by native defect pairs of deep donors and acceptors, which ensures that the material is semi-insulating. But this compensation mechanism also ensures that the point defects, which are the recombination sites, are predominantly ionized and therefore produce significant Stark broadening of the excitonic transition.[21,22] In annealed LTG materials, which are compensated by arsenic precipitates, the point defect concentrations are low and the precipitates are predominantly neutral, without long-range Coulomb potentials to perturb the excitonic transitions. However, neutral precipitates are still efficient recombination centers, providing ultrafast carrier recombination.[5] Therefore, annealed LTG materials rely on a different compensation mechanism than radiation damage, which may represent a new technique for achieving ultrafast devices without the adverse effects of long-range Coulomb perturbations on optical transitions.

In conclusion we have demonstrated sharp quantum-confined excitons in ultrafast low-temperature growth AlAs/GaAs quantum wells with linewidths comparable to high-quality multiple quantum wells grown at standard temperatures. These new materials eliminate the need for artificial methods to produce semi-insulating quantum wells, such as Cr-doping or proton implantation. The ability to grow high-quality quantum wells with sharp excitonic spectra, that are semi-insulating and have ultrafast carrier lifetimes, should represent an advance in many applications that use semi-insulating semiconductor nanostructures.

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