Nonlinear spectroscopy of InGaAs/InAlAs multiple quantum well structures

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The first investigation of nonlinear absorption in InGaAs/InAlAs multiple quantum wells using picosecond and cw infrared lasers is presented. The nonlinearity is demonstrated to be due to plasma-induced excitonic bleaching. The measured saturation densities agree with those predicted by theory.

A unique feature of multiple quantum well structures (MQWS's) is that their optical and electronic properties can be tailored as compared to bulk material. This has been shown in the case of excitons in MQWS's, where the quantum confinement enhances the exciton binding energy, whereas the broadening due to scattering by thermal phonons is hardly changed.[1] This has resulted in the observation of well-resolved exciton absorption resonances at room temperature in GaAs/AlGaAs quantum wells. MQWS excitons show novel nonlinear absorption and electroabsorption effects compared to those of bulk material.[2,3] These effects have been used to demonstrate devices such as high contrast[4] and high-speed modulators,[5] optically bistable devices,[6,7] optical level shifters, linearized modulators,[6,8] and picosecond mode-locked diode lasers.[9]

Recently, well-resolved excitonic absorption lines have been observed at room temperature in infrared band-gap MQWS's such as InGaAs/InAlAs,[10,11] InGaAs/InP,[12] and GaSb/AlGaSb[13] MQWS's. This should make possible devices for use at the 1.5 μm optical fiber transparency region, similar to those already developed with the GaAs/AlGaAs MQWS. We present the first measurements of nonlinear absorption and recovery in InGaAs/InAlAs MQWS's using a ps and cw infrared tunable color-center laser (TCCL). We find low saturation intensity for the nonlinear absorption. We have also studied the saturation of a MQWS inside the extended cavity of a laser diode, using the TCCL as a probe. This first study of a saturable absorber *in situ* provides information important to the understanding of possible diode passive mode locking and demonstrates that the nonlinearity is due to plasma-induced excitonic saturation. These results also show scaling of the nonlinearities with band gap.

The experimental setup consisted of a neodymium-yttrium aluminum garnet (Nd-YAG) laser synchronously pumping a TCCL.[14] The system could either be run cw, or mode locked to produce 10-ps-long pulses at a repetition rate of 100 MHz. The tuning range of the TCCL is 1.43–1.57 μm. The samples consisted of 50 periods of $L_z = 100$ Å $In_{0.53}Ga_{0.47}As$ well, separated by $L_z = 100$ Å $In_{0.52}Al_{0.48}As$ barriers. Since the exciton Bohr diameter is about 500 Å in bulk InGaAs material, the exciton in these samples should exhibit strong quantum size effects. We have studied undoped samples as well as *p-i-n* doped samples with the MQWS in the intrinsic region.

In single beam saturation measurements the TCCL beam was focused to a 30-μm spot on the sample and germanium detectors were used to measure the incident and transmitted intensities. From this, the absorption was calculated using the below band-gap transmission to correct for the reflections at the sample faces.

The diamonds in Fig. 1 represent the absorption versus peak incident intensity for an undoped sample, measured using cw excitation at 1.52 μm wavelength. The solid curve was obtained by averaging over the Gaussian beam profile, including the effective interaction length and using the saturation intensity as an adjustable parameter.[15] Similar measurements and analyses were performed on both types of samples using cw and picosecond excitation. With cw excitation the saturation intensities obtained are $I_s = 2.5$ and 5 kW/cm^2 for undoped and *p-i-n* samples, respectively. With ps excitation the time-averaged saturation intensities are $I_s = 0.8$ and 2 kW/cm^2, respectively.

The time dependence of the induced change in absorption was measured using the pump/probe technique with ps excitation. A typical result for the undoped sample is shown in Fig. 2. A single exponential fit was used to determine the absorption recovery time, $\tau = 1.5 \pm 0.25$ and 1.3 ± 0.25 ns for the undoped and *p-i-n* doped samples, respectively.

The saturation intensity is easily understood from the model originally developed to explain nonlinear absorption in GaAs/AlGaAs MQWS's.[16] In the case of excitation long compared to the exciton ionization time, $\tau_i = 280$ fs for 100 Å InGaAs/InAlAs MQWS's,[10] an electron-hole (e-h) plasma is generated directly by nonresonant absorption, or, in the case of resonance absorption, indirectly via exciton ionization. The dominant effects on the exciton resonances owing to the occupation of states close to $k = 0$ by e-h pairs are due to the Pauli exclusion principle, i.e., phase-space filling and exchange interaction. These mechanisms prevent further formation of excitons.[16] The saturation is therefore directly governed by the carrier density N_{eh}, with the recovery time determined by the lifetime of the e-h plasma.

This model can be used to compare the various results we have obtained. The saturation is determined by the carrier density N_{eh}. $N_{eh} \propto 1/\tau$, where τ is the carrier lifetime τ_r for cw excitation or the time between pulses τ_m for mode-locked excitation. In addition, in the single beam pulsed measurements the carrier density N_{eh} must also be averaged over the pulse envelope. It can be shown that, for Gaussian pulse envelope, with pulse duration much smaller than the recovery time and in the small-signal regime, the average carrier density $\langle N_{eh} \rangle$ seen by the single pulse is given by

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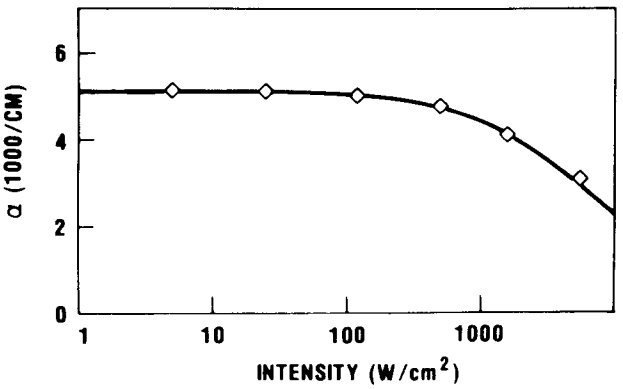

FIG. 1. Intensity dependence of the absorption at 1.52 μm wavelength of an undoped InGaAs/InAlAs MQWS consisting of 50 periods of 110 Å wells separated by 110 Å barriers. The diamonds are the experimental points measured with cw excitation. The solid curve is a fit, including correction for the Gaussian beam profile, obtained using a saturation intensity of 2.5 kW/cm^2.

$\langle N_{\rm eh} \rangle = 0.5 N_{\rm eh}$, where $N_{\rm eh}$ is the total carrier density generated by one pulse. The ratio of saturation intensities in pulsed (I_s^{ml}) and cw ($I_s^{\rm cw}$) measurements is therefore given by

$$I_s^{ml}/I_s^{\rm cw} = 0.5\tau_m/\tau_r \ . \tag{1}$$

This value is 3–4 for our samples, which explains well why the saturation intensities measured in the ps mode are about three times larger than those measured in the cw mode. For the comparison between *p-i-n* and undoped samples, some internal field sweeping of the carriers in the *p-i-n* samples can explain the small decrease in lifetime, but this is not sufficient to account for the factor of 2 in I_s, which may be related to the complex structure of the *p-i-n* samples.

In an approximation of the theory that gives results in a closed form and has compared well with the GaAs/AlGaAs experimental results,[15] I_s corresponds approximately to the intensity incident on the sample that generates one free carrier per exciton area per carrier lifetime:

$$I_s = \hbar\omega/(2A_x\tau_r\alpha L_z) \ , \tag{2}$$

where $\alpha = 10^4$ cm^{-1} is the absorption coefficient considering only the thickness of the InGaAs active layers[10] and A_x is the exciton area. For $L_z = 110$ Å Weiner *et al.*[10] obtained an exciton binding energy $E_{1S} = 5.2$ meV, giving an exciton radius of $a_x \sim 95$ Å. From Eq. (2) we obtain $I_s \sim 1.5$ kW/cm^2. Considering the uncertainties in the parameters for this material system, this result is in good agreement with our experimental value. The saturation intensities are higher than those measured in GaAs/AlGaAs MQWS's[2] of 200–500 W/cm^2. This is due to the shorter lifetime τ_r observed in the InGaAs/InAlAs MQWS as compared to the GaAs/AlGaAs MQWS, for which $\tau_r = 20$ ns, most likely because of the higher quality of the presently more developed GaAs/AlGaAs MQWS. In terms of the carrier densities (and hence energy densities) required for saturation, the InGaAs/InAlAs MQWS absorption should be easier to saturate than that of the GaAs/AlGaAs MQWS due to the smaller E_{1S} and hence larger diameter of the exciton. This comparison represents the first demonstration of the scaling behavior of room-temperature excitonic nonlinearities between different quantum well material systems and provides further support for the theory.[16]

In order to study the frequency dependence of the changes in the absorption spectrum of the MQWS, we performed a two-beam experiment in which an InGaAsP laser diode was used to saturate the absorption and the tunable color center laser was used as a probe. The diode operated at 1.5 μm, near the peak of the excitonic absorption. One facet of the laser was antireflection coated. The output from this facet was collimated and then focused to a 3 μm radius spot on the MQWS using 8 mm focal length objectives. The MQWS acts as the output mirror for the extended cavity of the laser diode. The cw output of the color center laser was introduced into the laser cavity with a beamsplitter, collinear to that of the diode, but with crossed polarization. Additional polarizers were used to isolate the beams from each other. The portion of the laser-diode beam reflected from this beamsplitter was monitored to determine the laser-diode intensity incident on the sample. The wavelength of the color center laser was then scanned to obtain a saturation spectrum.

The resulting spectra are shown in Fig. 3(a). The solid and broken curves represent the absorption without and

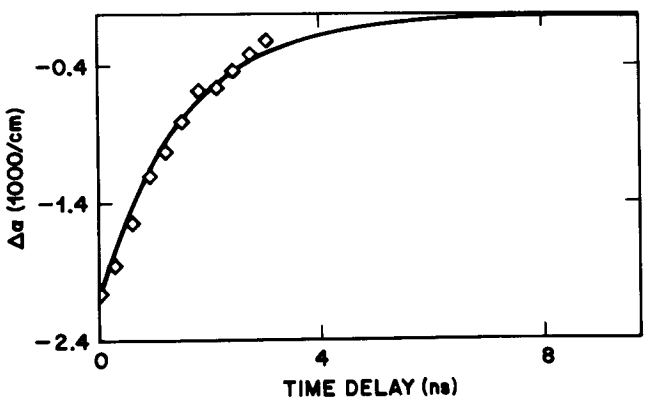

FIG. 2. Time dependence of the absorption induced in an undoped MQWS containing 110 Å wells. The laser wavelength was 1.52 μm. The diamonds are the experimental points. The solid curve is a single exponential fit using a decay time of 1.5 ns.

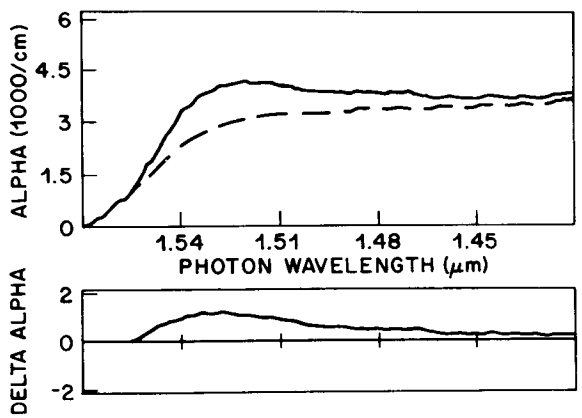

FIG. 3. (a) Absorption spectra of a 110 Å InGaAs MQWS. The solid and broken curves were obtained with diode laser pump intensity of 0 and 19 kW/cm^2, respectively. (b) Differential absorption spectrum obtained by taking the difference between the two spectra in (a).

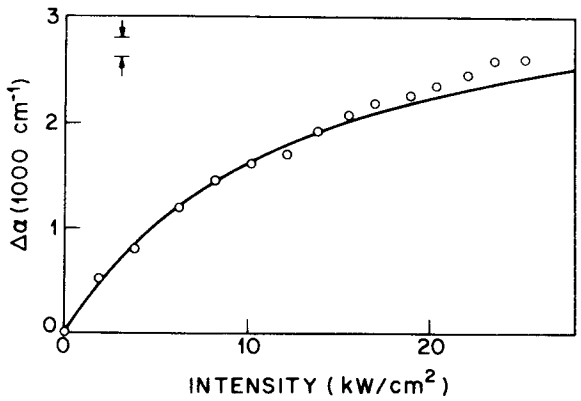

FIG. 4. Induced change in the MQWS absorption as a function of incident intensity, measured at the exciton peak wavelength of 1.524 μm. The circles are the experimental points. The solid curve is a fit obtained assuming only saturation of the exciton absorption with a saturation intensity of 10 kW/cm^2.

with diode laser pump, respectively. The difference between the two spectra is shown in Fig. 3(b). The induced absorption bleaching is as large as 1500 cm^{-1} at the exciton peak, decreasing at higher energy. Clearly, at the high intensity shown the excitonic feature has completely disappeared, leaving only the steplike band to band absorption in good agreement with the current interpretation of exciton saturation in MQWS's.[2]

In order to determine the saturation intensity in this geometry the color center laser was set to the exciton peak wavelength of 1.524 μm and the change in its absorption was observed as a function of the laser diode power (Fig. 4). The maximum induced change in absorption is as large as 2600 cm^{-1}. The curve is linear in intensity at low power, saturating at higher power. The theoretical curve was obtained in the same manner as that of Fig. 1, with a constant term included in the absorption to represent the band to band absorption, which is much harder to saturate than the excitonic absorption.[1,2] A best fit was obtained assuming that the exciton contribution to the absorption at this wavelength is 3500 cm^{-1}; the saturation intensity was determined to be 10 kW/cm^2. This is much larger than the 2.5 kW/cm^2 saturation intensity measured at resonance using the color center laser alone. This may well be due to the difficulty of obtaining complete overlap between the very small diode laser and color center laser focal spots. A portion of this difference can also be explained by a rapid diffusion of carriers out of the much smaller diode laser focal spot used here, reducing the carrier density.

In conclusion, we have presented the first detailed study of nonlinear absorption in InGaAs/InAlAs MQWS's. The results are satisfactorily interpreted in terms of the theories developed for GaAs MQWS's using scaling of the material parameters. These results are important to the development of the mode-locked semiconductor lasers and optoelectronic devices for the 1.5 μm wavelength transparent region of optical fibers.

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