Large room-temperature optical nonlinearity in GaAs/Ga_{1-x} Al_xAs multiple quantum well structures

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We report the first measurements of optical absorption saturation in GaAs/GaAlAs multiple quantum well (MQW) structures at room temperature near the heavy hole exciton peak. Linear absorption shows distinct exciton peaks at room temperature in the MQW and we deduce this is because the confinement increases exciton binding energy without increasing LO phonon coupling. This room-temperature MQW absorption also saturates more readily than that in a comparable GaAs sample; the measured saturation intensity is 580 W/cm^2 with a recombination time of 21 ns in a MQW with 102-Å GaAs layers. From this we predict a nonlinear refraction coefficient $n_2 \sim 2 \times 10^{-5}$ cm^2/W. This large nonlinearity should permit room-temperature optical devices compatible with laser diode wavelengths, materials and power levels.

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Recently there has been considerable interest in nonlinear optical effects in semiconductors associated with absorption saturation, both for the physical insight which these give and for practical applications in low-power all-optical switching and signal processing devices.[1] We report the first measurements of optical absorption saturation in room-temperature GaAs/Ga_{1-x} Al_xAs multiple quantum well (MQW) structures,[2] showing saturation intensities as low as 580 W/cm^2 near the principal excitonic feature at the fundamental band edge. We also investigate both the reasons for the existence of the strong excitonic absorption in room-temperature MQW's and the mechanism of absorption saturation, and compare the behavior of the MQW with bulk GaAs. Previous measurements of absorption saturation and related nonlinear effects near the band edge of GaAs[1,3,4] and MQW's[5–7] have concentrated on low-temperature behavior. In GaAs the excitonic feature all but disappears at room temperature.[8] In contrast, the strong and readily saturable MQW excitonic absorption investigated here may have a major impact in optical signal processing by permitting room-temperature devices to be fabricated with materials and switching intensities compatible with laser diode sources.

The samples were grown by molecular beam epitaxy on GaAs substrates. The MQW sample consists of alternate 102-Å GaAs layers and 207-Å $Ga_{0.72}$ $Al_{0.28}$As layers of total thickness ~ 2.4 μm. The GaAlAs layers effectively confine carriers within the GaAs layers. The GaAs sample has a 3.2-μm GaAs layer between two 3-μm $Ga_{0.7}$ $Al_{0.3}$As layers. In both cases a $\sim$1-mm-diam hole has been etched away from the substrate using a selective etch, leaving the epitaxial layers exposed. For the MQW sample some etching of the epitaxial layers themselves was unavoidable so the actual thickness is less than 2.4 μm. Both samples were undoped with estimated carrier concentrations less than $\sim 10^{15}$ cm^{-3}. The laser was an oxazine 750 ring dye laser,[9] operated with propylene carbonate dye solvent,[10] pumped with a Spectra Physics 171 Krypton ion laser either cw or synchronously mode locked.

Figure 1 shows linear absorption spectra of the samples at room temperature. In GaAs, the spectrum shows a slight bump just at the band edge which is the remains of the excitonic resonance[8] whereas the MQW spectrum shows two clear resonances at $\sim$1.463 eV and 1.474 eV, well resolved from the interband absorption. These two peaks are thought to arise from the heavy hole (lower energy) and light hole (higher energy) excitons.[11,12] The carrier confinement in the MQW layers influences the nature of the exciton. In the idealized extreme of very thin layers the exciton would behave like a two-dimensional hydrogen atom,[13] giving a 2-D radius $\frac{1}{4}$ of the 3-D radius and a 2-D binding energy four times

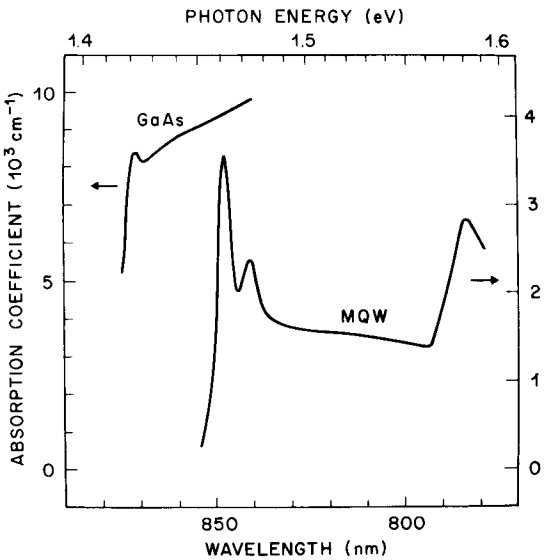

FIG. 1. Linear absorption spectra at room-temperature GaAs and MQW samples. (Note that the MQW absorption is underestimated because of thickness loss during etching.)

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greater. Our sample is in an intermediate situation (the layer thickness is $\sim\frac{1}{3}$ of the 3-D exciton diameter), and a heavy-hole exciton binding energy of ~ 9 meV, with orbit radius $a_0 \sim 60$ Å, is estimated[14] for our MQW (compared to 4.2 meV and 140 Å for GaAs). To understand why the MQW shows such clear exciton resonances at room temperature, we have measured absorption spectra in the MQW at several temperatures; we measure the half-width at half-maximum of the heavy-hole exciton peak, Γ, using the data on the low-energy side of the peak. The results show an approximately constant width at low temperatures increasing for temperatures above ~ 150 K. The low-temperature exciton linewidth (Γ_0) has been ascribed to fluctuations of layer thickness of the order of one atomic layer.[15] The usual dominant thermal broadening mechanism for exciton resonances at room temperature in 3-D semiconductors is LO phonon absorption. The degree of broadening on such a model is proportional to the number of LO phonons, so we have

$$\Gamma = \Gamma_0 + \Gamma_B / (e^{\hbar\omega_{LO}/kT} - 1). \quad (1)$$

Here $\hbar\omega_{LO}$ is the LO phonon energy and Γ_B is a measure of the phonon broadening. Using $\hbar\omega_{LO}/k = 428$ K (appropriate for GaAs), we obtain a good fit with $\Gamma_0 = 2.0$ meV and $\Gamma_B = 5.5$ meV. The broadening parameter Γ_B and the binding energy E_B of the MQW heavy-hole exciton compare very favorably with those of 3-D semiconductors with similar band gap.[16,17] The broadening parameter for the MQW is actually less than that for GaAs. The confinement therefore offers a unique method of increasing the binding energy without increasing the phonon broadening, resulting in clear room-temperature excitonic structure. The phonon contribution to Γ of $\Delta E \simeq 1.8$ meV at room temperature corresponds to a mean time τ_0 to phonon absorption of ~ 0.4 ps using the relation $\Delta E = \hbar/\tau_0$ for a Lorentzian line shape. As LO phonon absorption ionizes the exciton, this model implies that 0.4 ps is also the mean time to ionization of the exciton after its creation.

Figure 2 shows the measured intensity dependence of the cw optical absorption near the (heavy-hole) excitonic absorption peak in both the MQW and GaAs samples. The average power was reduced by a 1:100 chopper to eliminate bulk sample heating. The effective mean intensity used here is $\frac{1}{3}$ of the intensity in beam center, as suggested by numerical calculations of saturation with Gaussian beams.[18] The effects of multiple reflections are also taken into account by assuming that the cavity is on resonance. The results in Fig. 2 demonstrate directly that the MQW excitonic absorption may be saturated with milliwatt powers and that this saturation is much easier to achieve than that in the room-temperature GaAs.

TABLE I. Saturation intensities (see text definition) measured for the MQW around and above the heavy-hole exciton peak at room temperature (spot size $\sim 20\,\mu$m).

Wavelength (nm)	Energy (eV)	Linear absorption coefficient (cm^{-1})	Saturation intensity I_s (W/cm^2)
849.5	1.4595	2420	3680
847.7	1.4626	3610	580
846.8	1.4642	3380	790
831.4	1.4913	1630	$\gtrsim 8000$

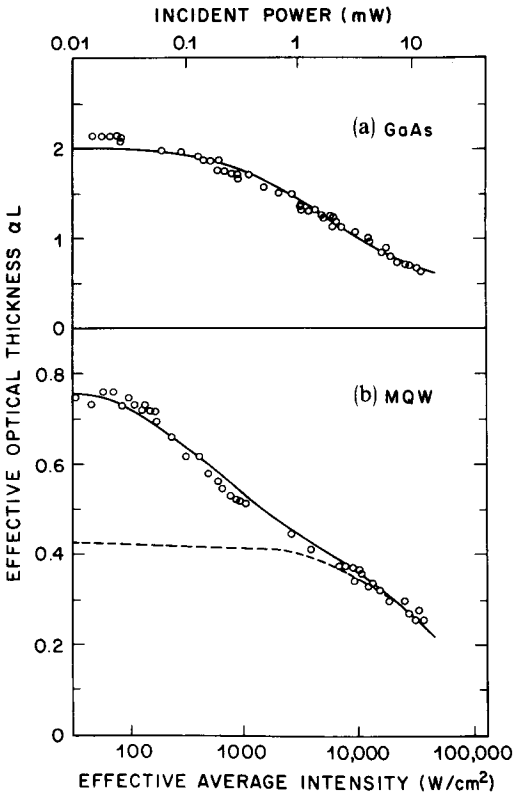

FIG. 2. Intensity dependence of optical absorption in the GaAs and MQW samples at room temperature with a ~ 5-μm-diam spot. The solid curves are semiempirical Lorentzian saturation fits to the data for comparison: (a) (GaAs) $\alpha L = 0.5 + 1.5/(1 + I/4400)$, (b) (MQW) $\alpha L = 0.35/(1 + I/580) + 0.43/(1 + I/44000)$. ($\alpha L$ is the optical thickness of the sample and I is the intensity in W/cm^2.) The broken curve in (b) is $\alpha L = 0.43/(1 + I/44000)$.

We have also studied the MQW sample with a larger (20–50-μm diameter) spot, so that filamentary heating time constants will be relatively longer, and looked at the onset of saturation using the chopper at a focus to give a rise time ~ 2 μs on a pulse $\sim 300\,\mu$s long. A thermal absorption change of time constant 120 μs, and corresponding to ~ 1.6 K/mW heating, was resolved; the detector output was electronically gated within the first 5 μs in subsequent measurements to eliminate these thermal effects. We deduced effective saturation intensities from small increases ($\lesssim 10\%$) in transmission with increasing incident power at several wavelengths; these are listed in Table I. The saturation intensity I_s is defined and calculated as follows. If the local absorption coefficient $\alpha \simeq \alpha_0 - \alpha_2 I$, where α_0 is the linear absorption and α_2 represents the first order nonlinear absorption, I_s may be defined $I_s \equiv \alpha_0/\alpha_2$. [This definition is consistent with the saturation intensity for a Lorentzian absorber $\alpha = \alpha_0/(1 + I/I_s)$]. We correct for single surface reflections and linear absorption in calculating I from the incident power; Fabry–Perot effects are neglected because of the relatively large linear absorption at low intensities. Gaussian beam effects are deconvoluted exactly for these small-signal measurements using an analytic technique.[19]

For purposes of comparison we have fitted the results in Fig. 2 with Lorentzian saturation curves [i.e. $\alpha = \alpha_0 / (1 + I/I_s)$]. The GaAs results are a best fit; the MQW results are fitted at low intensity with a Lorentzian saturation model using the measured value from Table I. This fit confirms good agreement between small and large spot results. The simple Lorentzian model is inadequate for the MQW at high intensities but the fit can be improved by adding a further Lorentzian absorption saturation component with much higher saturation intensity. The detailed nature of excitonic absorption saturation in both GaAs and MQW structures remains to be resolved. However, a Lorentzian model also enables us to make an estimate of the nonlinear refraction coefficient n_2 (defined through $n = n_L + n_2 I$, where n_L is the linear refractive index) which we deduce to be 2×10^{-5} cm^2/W, corresponding to an effective degenerate four-wave $\chi^{(3)}$ of $\sim5\times10^{-3}$ esu. This value is $\sim10^9$ larger than CS_2 (Ref. 20) and $\sim10^5$ larger than room-temperature silicon.[21]

The recovery time of the absorption change has been measured using the laser synchronously mode locked where it produces a train of pulses ~8 ps long and 12 ns apart. The recovery was too long to be measured accurately by the normal pump/probe delay line technique, but could be deduced from the difference in induced transmission for the two cases of (a) probe arriving just after pump and (b) probe arriving just before pump, i.e. 12 ns after the previous pump pulse. Assuming the absorption varies linearly with excitation and an exponential decay we obtain a lifetime $\tau\simeq21$ ns. This time did not change measurably for intensities in the range 20–250 W/cm^2.

With regard to the actual saturation mechanisms we expect that because of the high temperature ($kT\sim3E_B$) a substantial fraction of the excitons ionized by LO phonon absorption remains ionized in the steady state; assuming Boltzmann statistics and using a simple two-dimensional density of states, we have modified the Saha equation[22] of plasma physics to estimate this fraction. Using the measured $\tau = 21$ ns as the recombination time we estimate a created exciton density of $\sim3.5\times10^{11}$ cm^{-2} ($\equiv3.3\times10^{17}$ cm^{-3} effective bulk density in the GaAs layers) at 500 W/cm^2. Even at this relatively high intensity ($\sim I_s$) we estimate that the total free carrier density (i.e. $2\times$the density of ionized excitons) of $\sim4.8\times10^{11}$ cm^{-2} is much greater than the density of nonionized excitons ($\sim1.1\times10^{11}$ cm^{-2}).[23]

These densities correspond to ~0.12 nonionized excitons and 0.54 free carriers per excitonic area (πa_0^2). The measured saturation intensity in low-temperature GaAs[4] at the excitionic resonance corresponds to a created exciton density ~1.6 per excitonic volume. As with the low-temperature GaAs saturation it is not possible to distinguish conclusively between the various possible mechanism for "saturation" of (1) exciton-exciton collisions, (2) exhaustion of potential exciton-forming states in the conduction and valence bands (which occurs for approximately one exciton per excitonic volume or area), and (3) screening of the electron-hole interation by created free carriers. However, this relatively low calculated nonionized exciton density at the saturation intensity argues against mechanisms (1) and (2) while the significant calculated free-carrier concentration argues for a screening mechanism. The calculated 2-D screening length parameter[24] for 4.8×10^{11} cm^{-2} free-carrier density is ~78 Å.

In conclusion, we have demonstrated that clear exciton resonances which can be saturated with milliwatt optical power can be observed in MQW structures at room temperature. We show that one reason for the existence of such clear resonances is that LO phonon broadening is apparently not enhanced by the MQW confinement while exciton binding energy is. We argue that a likely mechanism for the saturation is screening of the electron-hole interaction by created free electrons and holes.

The discovery of this large room-temperature nonlinearity with materials, wavelengths, and powers compatible with laser diodes is an important step towards the realization of practical all-optical switching and signal processing devices.[25]

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