Degenerate four-wave mixing in room-temperature GaAs/GaAlAs multiple quantum well structures

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We report the first observation of forward degenerate four-wave mixing (DFWM) in room-temperature GaAs/GaAlAs multiple quantum well structures near the exciton resonances. In a sample 1.26 μm thick with sixty-five 96-Å GaAs quantum wells we observe $\sim 10^{-4}$ diffraction efficiency with ~ 30 W/cm^2 average intensity from a mode-locked laser. We measure nonlinear absorption and DFWM spectra, and also a change in refractive index, per carrier pair/cm^3, of $n_{eh} \sim 2\times 10^{-19}$ cm^3 just below the heavy hole exciton peak. With 20-ns carrier lifetime this corresponds to an effective nonlinear coefficient for cw beams of $|n_2| \simeq 2\times 10^{-4}$ cm^2/W. This is appreciably larger than previous estimates and encouraging for room-temperature all-optical devices.

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Degenerate four-wave mixing (DFWM) is of current interest both for practical applications (e.g., phase conjugation[1]) and as a physical probe. As a measurement technique it can detect very small nonlinear changes in refraction, and this ability has been exploited in several semiconductor materials.[2] In this letter we report the first observations of DFWM in GaAs/GaAlAs multiple quantum well structures (MQW's) at room temperature. By combining DFWM and nonlinear absorption results we can obtain a direct measurement of the nonlinear refraction near the band gap of the MQW which we compare with previous estimates.[3] These measurements are of practical importance for possible low-power optical devices compatible with laser diodes based either on DFWM, nonlinear refraction (such as optical bistability)[4] or nonlinear absorption.

MQW structures show exceptionally strong excitonic absorption resonances at room temperature; apparently, the quantum confinement in the GaAs wells increases the exciton binding energy without increasing the phonon broadening.[3] Furthermore, this absorption is much more readily saturated than is the band edge absorption in a comparable conventional semiconductor at room temperature.[3] The room-temperature MQW excitonic saturation has been explained as due to the screening of excitons by the free carriers generated through thermal ionization of excitons.[3] DFWM has previously been seen at low temperatures in a MQW.[5] Nonlinear refraction has been implicitly observed in room-temperature MQW's in optical bistability measurements,[4] although in this case the excitonic contribution may have been totally saturated. By measuring with low intensities we avoid such saturation of the nonlinearities.

The MQW samples were grown by molecular beam epitaxy (MBE) on GaAs substrates, with the MQW layers sandwiched between GaAlAs cap layers which are transparent at the wavelengths of interest here. One sample consisted of 84 periods of 144-Å GaAs and 102-Å $Ga_{0.7}Al_{0.3}As$ (giving a total MQW thickness ~ 2.1 μm) with 0.3-μm $Ga_{0.7}Al_{0.3}As$ caps and the other had 65-periods of 96-Å GaAs and 98-Å $Ga_{0.72}Al_{0.28}As$ (giving a 1.26-μm MQW thickness) with 1.45-μm caps. The samples were cleaved to ~ 1 mm square pieces, the GaAs substrate was removed with a selective etch and the resulting epitaxial layer glued to a sapphire disc with a $\lesssim 2$ μm layer of epoxy. For the 96-Å MQW, a thin piece of glass was also glued over the sample. The sapphire and thin glue layers were used to avoid thermal effects.[3] The experimental apparatus is shown in Fig. 1. An oxazine 750 ring dye laser was synchronously pumped with a Spectra-Physics 171 krypton laser giving mode-locked pulses spaced 12 ns apart.

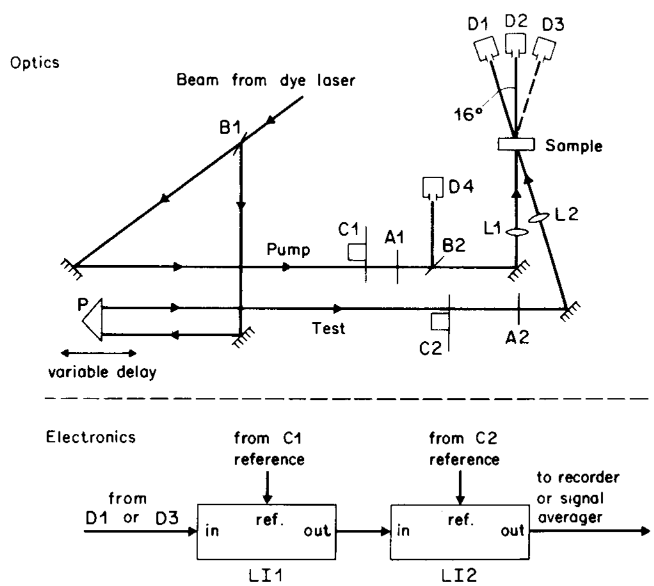

FIG. 1. Beam splitter B1 generates the test beam which goes through the optical delay line consisting of corner cube prism P on a motorized translator. The beams [attenuated by A1 (variable) and A2 (fixed) neutral density filters], are focused by lenses L1 and L2 (15- and 12-cm focal length, respectively) onto the same spot on the sample. Detectors D 1–3 are silicon photodiodes. Choppers C 1 and C 2 are set at 385 and 10 Hz, respectively. When the output of D1 or D3 is passed into the chain of two lock-in amplifiers LI1 and LI2, only those signals which depend on the product of pump and test beams give an output from LI2, thus suppressing all linear scatter. LI1 and LI2 are operated with 30 ms and 1 s time constants, respectively. Detector D 4 monitors laser power (sampled by beam splitter B2).

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The dye laser was tuned under computer control using two birefringent plates and gave pulses of second harmonic generation autocorrelation half-height width ~6 ps without a significant coherence spike. The focused spot sizes on the sample were ~45 and 30 μm $1/e^2$ intensity diameter for pump and test beams, respectively. For the $\lesssim 2$ μm active length in our samples, the slight phase mismatch in forward DFWM may be neglected. Detector D1 detects the change in transmission of the test beam due to the pump beam [i.e., (transferred) nonlinear absorption]. D2 measures transmission and D3 detects the DFWM signal.

A DFWM signal was observed near the exciton peaks on detector D3 in the vicinity of zero pump-test delay. Scanning the pump-test delay around zero gave a DFWM time delay spectrum of half-height width ~3.1 ps, which is consistent with the requirement of pump-test pulse overlap in the sample. Figure 2 shows the dependence of D3 signal on pump intensity at 1.440 eV (near the heavy hole exciton peak) for the 144-Å MQW sample. With 25-ps delay between test and pump the signal on D3 varies linearly with intensity (see Fig. 2) which we interpret as scattered nonlinear absorption. As the nonlinear absorption spectrum is experimentally identical for 0 delay and 25-ps test delay we include this scatter together with the quadratic term in the dashed fit at 0 delay; thus it confirms that the DFWM signal increases quadratically as expected up to ~0.6 mW average pump power. We interpret the roll-off at higher powers as saturation of the nonlinearity. Subsequent measurements used powers $\ll 0.6$ mW. Allowing for reflection losses we were able to attain diffraction efficiences $\sim 10^{-4}$ with ~0.5 mW average pump power incident on the 96-Å sample. Note that the low test beam powers ($\ll 0.6$ mW) and the small absolute diffraction efficiencies imply that pump depletion should be negligible in our conditions. At low pump powers the signal was approximately proportional to the test beam power.

Spectra were taken under similar conditions on detectors D1, D2, and D3 while simultaneously recording the laser power using D4. Figure 3(a) shows the linear transmission spectrum obtained from the D2 and D4 spectra. Figure 3(b) shows the signal from D1 after processing by the chain of lock-ins (with 25-ps test beam delay). Since Fig. 3(b) is the change in transmitted test beam power as a function of wavelength, it is a measure of the nonlinear absorption. Figure 3(c) shows the signal from detector D3 after the lock-ins taken near zero delay; this spectrum has the scattered nonlinear absorption signal (measured on D3 with the test delayed ~25 ps after the pump) subtracted from it. No reflection corrections have been applied in Fig. 3, and Figs. 3(b) and 3(c) are not corrected for the slight variation in laser power across the spectrum. Qualitatively similar spectra were observed with the 144-Å sample although the peaks occur at lower energies (~1.443 and ~1.450 eV) and are less clearly resolved.

The DFWM signal shows a clear peak [Fig. 3(c)] near the heavy hole exciton resonance. The nonlinear absorption spectrum is more complicated and we interpret it as follows. At the heavy hole exciton peak, the transmission of the test is increased by the presence of the pump (consistent with previous observations)[3] whereas on either side of this peak there is a clear decrease; this behavior is consistent with a broadening of the heavy hole exciton peak with increasing intensity. Preliminary attempts to fit this curve indicate also some shift

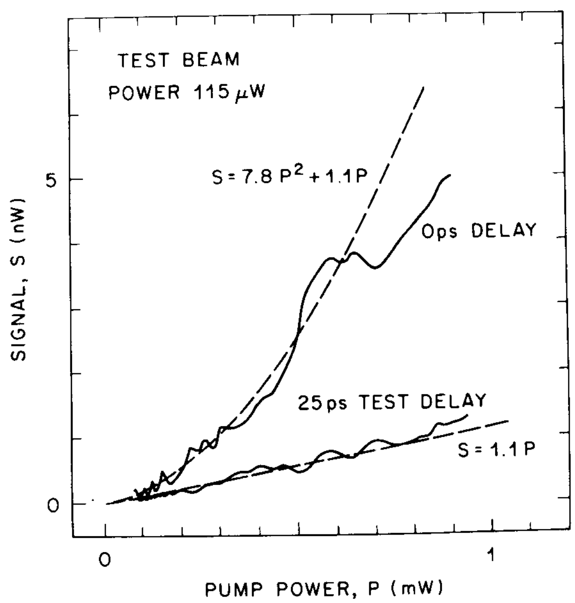

FIG. 2. Signal from $D3$ with 144-Å MQW sample showing quadratic behavior with a linear background.

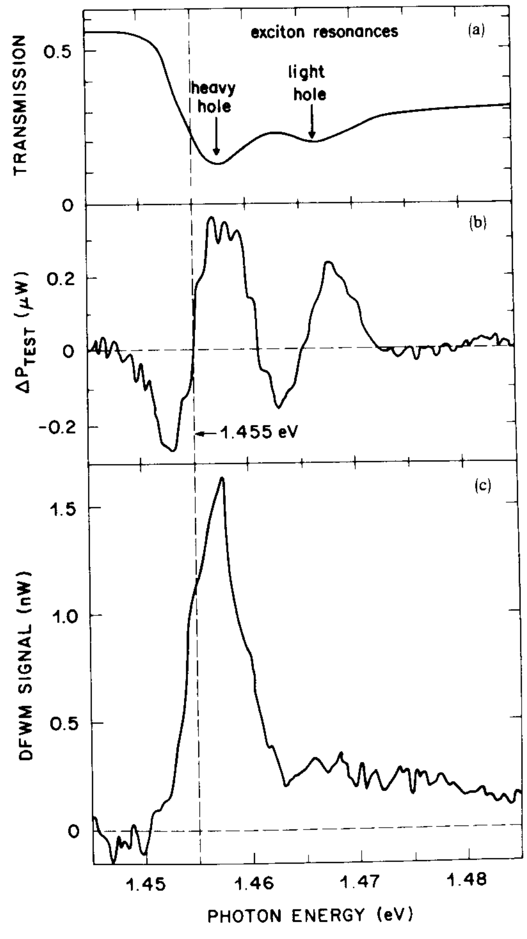

FIG. 3. Spectra for the 96-Å MQW: (a) linear absorption; (b) nonlinear absorption signal; (c) DFMW signal. For (b) and (c) the beam powers were 300 and 60 μW, respectively, for pump and test at 1.457 eV, increasing by 15% by 1.485 eV and decreasing by 15% by 1.445 eV with approximately linear variation.

of the peak to lower energy; this will be the subject of future work. However, the behavior contrasts with low-temperature results[5] which show primarily a peak shift. There is also some evidence of the same phenomena for the light hole exciton peak. The nonlinear absorption will be discussed in a subsequent paper. However, the important point at present is that where the nonlinear absorption crosses zero (e.g., at ~ 1.455 eV) the DFWM is still large. We have carefully confirmed this fact and also observe a similar phenomenon for the 144-Å sample. As the DFWM arises from either absorptive or refractive effects, the existence of DFWM in the absence of nonlinear absorption is a direct proof of the existence of nonlinear refraction. The value of the effective nonlinear coefficient can be obtained from the magnitude of the DFWM signal.

Interpretation of the DFWM magnitude requires some care because of the existence of grating diffusion. The interference of pump and test beams for our conditions (i.e., 16° between pump and test beams) produces a carrier grating inside the material of spacing $\sim 3.0\ \mu$m, which gives rise to the DFWM signal. Using a hole mobility $\sim 400\ \text{cm}^2/\text{Vs}$[6] (appropriate for pure GaAs) we calculate[7] a grating "lifetime" ~ 120 ps so the grating disappears in the 12 ns between pulses. The actual carrier lifetime is much longer than this (we measure ~ 20 ns in these experiments for both samples using the method previously described[3]). It is precisely to avoid confusion between grating diffusion and carrier lifetime that we performed this experiment with picosecond pulses (i.e., much shorter than any relaxation times) rather than with a cw laser.

To analyze the results quantitatively at the point of zero nonlinear absorption we introduce the change in refractive index (n_{eh}) induced by one electron-hole pair per unit volume; as the grating diffuses completely between pulses but negligibly during each pulse, the DFWM signal depends on the number density of electron-hole pairs created by each pulse, $N = (\alpha/\hbar\omega)\int I(t)dt$ where I is the intensity, α the absorption coefficient, and $\hbar\omega$ the photon energy. In the small signal regime, the first order diffraction efficiency of such a grating when there is no nonlinear absorption is[8,9]

$$\rho \simeq \left(\frac{1}{2}\frac{2\pi}{\lambda} n_{eh}\, N l_a\right)^2 e^{-\alpha l},$$

where λ is the wavelength and l the thickness of the active (i.e., MQW) material; the effects of large absorption are accounted for through the effective interaction length $l_a = (1 - e^{-\alpha l})/\alpha$ and the factor $e^{-\alpha l}$. To relate this to beam powers we use the exact deconvolution technique for Gaussian beams.[10] We obtain the following results (including corrections for surface reflections): for the 144-Å MQW at 1.444 eV just below the heavy hole exciton peak where $\alpha = 7200\ \text{cm}^{-1}$, $|n_{eh}| = 2.1\times 10^{-19}\ \text{cm}^{-3}$; for the 96-Å MQW at 1.457 eV where $\alpha = 1.2\times 10^4\ \text{cm}^{-1}$, $|n_{eh}| = 2.5\times 10^{-19}\ \text{cm}^{-3}$. For steady-state nonlinear refraction with cw beams for time much longer than the carrier lifetime (~ 20 ns) these figures imply nonlinear refraction coefficients $|n_2|$ of $1.2\times 10^{-4}\ \text{cm}^2/\text{W}$ (144-Å MQW and $2.5\times 10^{-4}\ \text{cm}^2/\text{W}$) (96-Å MQW). We estimate an accuracy of a factor ~ 2 in these results. These n_2 values are considerably larger than previous estimates[3] for these materials, although in a cw DFWM configuration due account must be taken of grating diffusion.[7]

In conclusion, we have observed degenerate four-wave mixing at room temperature in multiple quantum wells near the exciton peaks. With average intensities $\sim 30\ \text{W/cm}^2$ we observe diffraction efficiencies $\sim 10^{-4}$ in samples only 1–2 μm thick. The signals arise from both nonlinear absorption and nonlinear refraction and we measure carrier density dependence of refractive index $\sim 2\times 10^{-19}$ per excited pair per cm^3. This large value considerably exceeds prevous estimates and is very encouraging for numerous applications. The fact that these large room-temperature nonlinearities are seen at low powers (< 1 mW) and at wavelengths (~ 850 nm) ideally suited to diode lasers lead us to believe that MQW structures may prove to be the key to practical optical devices for future optical communications and signal processing systems.

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