Reprinted from **Optics Letters,** Vol. 8, page 477, September 1983

Nonlinear optics with a diode-laser light source

D. A. B. Miller, D. S. Chemla, and P. W. Smith

Bell Laboratories, Holmdel, New Jersey 07733

A. C. Gossard and W. Wiegmann

Bell Laboratories, Murray Hill, New Jersey 07974

Received May 18, 1983

We report results of cw degenerate four-wave mixing experiments on room-temperature GaAs–GaAlAs multiple-quantum-well material using a commercial semiconductor diode laser as the sole light source. With cw powers of ~3 mW and intensities of ~13 W/cm^2, we observe diffraction efficiencies of ~10^{-4}, corresponding to an effective nonlinear coefficient of $\chi^3 \sim 6 \times 10^{-2}$ esu.

In this Letter we report what is to our knowledge the first nonlinear-optics experiment using a semiconductor diode laser as the sole light source. We show that, by using room-temperature multiple-quantum-well (MQW) material as the nonlinear medium, degenerate four-wave mixing experiments can easily be performed with the milliwatt cw powers available from a commercial semiconductor diode laser. Because MQW material is compatible with diode lasers not only in terms of growth technology but also in terms of light wavelength and power levels, this material has great promise for future integrated-optical signal-processing elements.

Nonlinear optical effects have been observed at room temperature in MQW materials near the bandgap energy.[1-4] These observations have all used dye lasers, although cw power levels well within the range of diode lasers have been utilized for nonlinear absorption measurements.[1,2] Previous room-temperature degenerate four-wave mixing (DFWM) measurements[4] employed a mode-locked dye laser, although the average power was again low. MQW materials show exceptionally strong absorption resonances near the bandgap energy because of excitons.[2] The primary cause of the nonlinear-optical effects in room-temperature MQW material at low intensity (i.e., $\leq$1 kW/cm^2) is thought to be screening of the excitons[2] by free carriers created optically either directly by interband excitation or indirectly by thermal ionization of optically created excitons. As the excitons are screened, the absorption resonances saturate,[1,2] broaden, and shift,[4] giving rise to changes in absorption and, consequently, in refractive index.[4] Measurements of these effects led us to predict that large nonlinear effects should also be observable with low cw powers with response times of the order of tens of nanoseconds. The GaAs–GaAlAs MQW material is also ideally suited for operation with GaAs laser diodes, as its bandgap occurs at laser-diode wavelengths (e.g., ~850 nm).

To demonstrate the feasibility of nonlinear optics with diode lasers and MQW material, we set up a forward DFWM experiment with the apparatus as shown in Fig. 1. Either the dye- or the diode-laser beam could be selected by removing or replacing the kinematically mounted mirror M1. The diode laser used was a Hitachi HLP 1400 of nominal operating wavelength 851 nm. The spectrum of the diode laser at 107-mA cw driving current showed a strong mode at ~850 nm containing approximately half of the laser power, with a further group of modes at ~849 nm. The dye laser was a synchronously mode-locked oxazine 750 dye laser pumped with a krypton-ion laser. The desired laser beam was split to form a pump beam and a test beam, which were both focused on the same spot on the sample. The angle between the beams was kept shallow (~1.5°) to reduce the effects of grating diffusion, which would otherwise degrade the signal.[5] The diode-beam-focused spots were ~200 μm $1/e^2$ power diameter in the horizontal direction (i.e., in the plane of Fig. 1) and ~150 μm in the vertical direction. Detector D1 was positioned to detect the forward DFWM signal, and the output from D1 was processed as described previously[4] by a pair of lock-in amplifiers to suppress any linear scatter of transmitted pump or test beams. Because of the narrow angle between the beams, some scattered light from the transmitted pump beam could overlap the signal beam on the detector. These two beams could constructively interfere on the detector. At low signal-beam powers when the scattered pump power greatly exceeded the signal power, the apparent signal that was due to this constructive interference could greatly exceed the actual signal. To remove this systematic error, the relative phase of pump and signal beams was varied rapidly by scanning prism P1 back and forth by $\pm$20 μm at a frequency of ~4 Hz so that the interference effect would average out to zero.

The MQW sample used for this experiment consisted of 65 periods of 96-Å GaAs and 98-Å $Ga_{0.72}Al_{0.28}As$ (resulting in 1.26-μm total MQW thickness) sandwiched between 1.45-μm-thick $Ga_{0.72}Al_{0.28}As$ cap layers. This sample, which was bonded with thin epoxy layers between a sapphire disk and a thin glass disk, is the same

0146-9592/83/090477-03$1.00/0

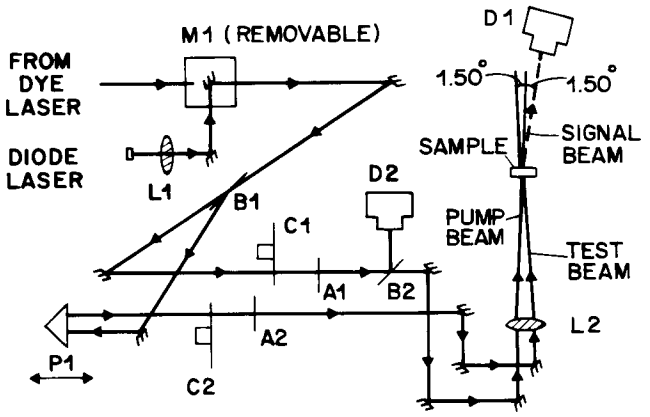

Fig. 1. Experimental apparatus. Lens L1, 8.0-mm focal-length 0.5-N.A. laser-diode lens. Mirror M1 is kinematically demountable to permit selection of dye- or diode-laser beams. The diode and dye beams are aligned along the same path. Beam splitter B1 splits off the test beam, and B2 samples the pump-beam power. Prism P1 is mounted on a computer-controlled translator to give variable delay of the test beam. A1 is a variable attenuator, and A2 is a fixed attenuator. The two beams are focused by a 50-cm lens onto the same spot on the sample. Silicon photodiode D1 detects the signal; D2 monitors the pump-beam power. The output from D1 is passed through a lock-in detector synchronized with chopper C1 at 385 Hz, and the output (with 3-msec time constant) is passed through a second lock-in detector synchronized with chopper C2 at 10 Hz; this gives the required signal while suppressing all linear scatter.

one previously used for nonlinear absorption and DFWM studies using a pulsed dye laser.[4] The heavy hole exciton feature occurs at ~851 nm in this sample. Because the diode-laser wavelengths are slightly shorter than the optimum wavelengths for DFWM in this sample at ~22°C as determined from the pulsed experiments (~851 μm), the sample was mounted on a thermoelectric cooler. The bandgap energy of GaAs shrinks by ~0.4 MeV/K (i.e., the bandgap wavelength increases by ~0.24 nm/K) around room temperature, permitting fine tuning of the optimum operating wavelength.

Figure 2 shows a typical power dependence of the forward DFWM signal with the cw laser diode. This curve was taken at 14.7°C sample temperature, which was the sample temperature for which the maximum signal was obtained, although a signal could be observed between 6 and 22°C. The curve has been fitted at low powers with a quadratic power dependence (as would be expected for this DFWM configuration) and a small linear background term. Pulsed measurements[4] have also shown a linear term, which we believe arises from a scattered nonlinear absorption signal. The curve shown in Fig. 2 was extended to relatively high power levels to show the roll-off of the signal at high powers compared to quadratic behavior. This roll-off of the DFWM signal occurs at intensities well below the measured absorption saturation intensity and may reflect the fact that, since the DFWM signal is proportional to the square of the nonlinearity, it is therefore sensitive to even small amounts of saturation of the nonlinearity. The quadratic behavior of the DWFM at low powers has, however, been checked down to <100-μW pump power. The diffraction efficiency (i.e., the forward reflectivity of the test beam) in Fig. 2 rises to $\sim 5 \times 10^{-5}$. The corrections for single surface reflections at each interface raise this to $\sim 10^{-4}$ diffraction efficiency inside the crystal. This efficiency compares well with that observed in pulsed experiments.[4]

We measured the effective carrier lifetime τ_R by using the mode-locked dye laser with the method described previously.[2] With 200-μm focused spot sizes we obtained $\tau_R \sim 31$ nsec. This is somewhat larger than the 21 nsec previously measured with smaller (~50-μm) spot sizes[2] and may indicate a small amount of diffusion of carriers out of the beam area for the smaller spot sizes. Assuming that the hole mobility is ~400 cm^2/V sec and that the electron mobility is much larger than this (as would be appropriate for pure GaAs), we calculate an ambipolar diffusion constant $D_a \sim 21$ cm^2/sec. The calculated grating diffusion time τ_D for 1.5° incident beam angle and refractive index 3.5 using this D_a is 13 nsec, implying an effective grating lifetime[5] $\tau = \tau_D\tau_R/(\tau_D + \tau_R)$ of ~9 nsec. The hole-mobility estimate may be high, however, so the only safe conclusion is that the grating lifetime is between 9 and 31 nsec.

To compare the present cw measurements with previous pulsed measurements,[4] we can compare diffraction efficiencies. The diffraction efficiency ρ can be expressed for both cw and pulsed conditions as the square of the product of a cross section times an effective number of carriers[6]; $\rho \propto (N\sigma)$.[2] The cross section σ contains the change of absorption coefficient and the change of refractive index per carrier.[7] It can be shown for an experiment with cw beams that $N = \alpha\tau I_p/\hbar\omega$ is the number of carriers accumulated during one grating lifetime. In an experiment with pulsed beams, N =

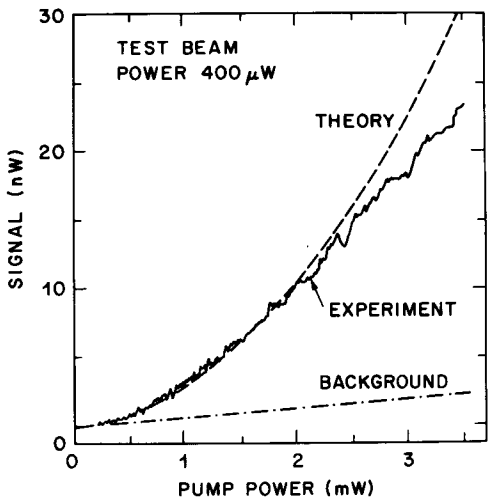

Fig. 2. DFWM signal and diode-laser light source. The sample was held at 14.7°C, and the diode current was 107 mA. The dashed line is a theoretical fit showing quadratic behavior up to ~2 mW with a small linear background (shown separately as the dotted–dashed line). Saturation of the nonlinearity at higher pump powers shows up as a deviation from the theoretical curve.

$(\alpha/\hbar\omega) \int I_p(t)dt$ is the number of carriers generated during one pulse, provided that the pulse duration is much smaller than the grating lifetime, which is itself much smaller than the pulse separation. In the present experiment we find that 1.3×10^{-14} cm$^2 \leq \sigma \leq 4.3 \times 10^{-14}$ cm^2, the two limits corresponding, respectively, to $\tau = 31$ nsec and $\tau = 9$ nsec. Our previous measurement with picosecond lasers gave[8] $\sigma \simeq 2.8 \times 10^{-14}$ cm^2. The two results are in remarkable agreement, especially if one considers that in the present experiment, because the output of the diode is not single frequency, the measured diffraction efficiency corresponds to an average value over the frequency distribution. These cross sections correspond to extremely large effective optical nonlinearities $\chi^{(3)} \sim 6 \times 10^{-2}$ esu. This value is about 10^6 times the $\chi^{(3)}$ for room-temperature silicon.[5]

In this Letter we have reported the first known use of room-temperature multiple-quantum-well material with a diode-laser source to perform a nonlinear-optics experiment. The extremely large optical nonlinearities associated with room-temperature excitons in this material should permit the design of integrated-optical signal-processing elements operating at submilliwatt power levels.

We acknowledge the invaluable assistance of D. J. Eilenberger in performing these experiments and of G. D. Aumiller in sample preparation.

References

1. D. A. B. Miller, D. S. Chemla, P. W. Smith, A. C. Gossard, and W.T. Tsang, Appl. Phys. B **28,** 96 (1982).
2. D. A. B. Miller, D. S. Chemla, D. J. Eilenberger, P. W. Smith, A. C. Gossard, and W. T. Tsang, Appl. Phys. Lett. **41,** 679 (1982). For other room-temperature linear absorption measurements see also, for example, S. W. Kirchoefer, N. Holonyak, Jr., K. Hess, D. A. Gulino, H. G. Drickamer, J. J. Coleman, and P. D. Dapkus, Appl. Phys. Lett. **40,** 821 (1982).
3. H. M. Gibbs, S. S. Tarng, J. L. Jewell, D. A. Weinberger, K. Tai, A. C. Gossard, S. L. McCall, A. Passner, and W. Wiegmann, Appl. Phys. Lett. **41,** 221 (1982).
4. D. A. B. Miller, D. S. Chemla, D. J. Eilenberger, P. W. Smith, A. C. Gossard, and W. Wiegmann, Appl. Phys. Lett. **42,** 925 (1983).
5. R. K. Jain and M. B. Klein, Appl. Phys. Lett. **35,** 454 (1979).
6. It can be shown that
$$\rho = \left(\left|\frac{2\pi}{\lambda} n_{eh} + \frac{i}{2}\sigma_{eh}\right| \frac{1}{2} N l_a \frac{w_s}{w_l}\right)^2 e^{-\alpha l},$$
where $l_a = (1 - e^{-\alpha l})/\alpha$, w_s, and w_l are the spot radii for the signal and test beams, and n_{eh} and σ_{eh} are defined as in Ref. 7. The cross section introduced in the text is
$$\sigma = \frac{1}{2}\left|\frac{2\pi}{\lambda} n_{eh} + \frac{i}{2}\sigma_{eh}\right|.$$
The derivation and discussion of these expressions as well as the rigorous definition of N will be dealt with in a forthcoming paper.
7. H. J. Eichler and F. Massman, J. Appl. Phys. **53,** 3237 (1982).
8. Since the publication of our paper,[4] we have improved the calibration of our energy measurements; our previous measurements were found to be high by a factor of 1.7. This has no bearing on the measurement of the diffraction efficiency but implies that the average pump intensity was not 30 W/cm^2 as quoted in Ref. 4 but 18 W/cm^2.