Mode locking of semiconductor diode lasers using saturable excitonic nonlinearities

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Multiple-quantum-well (MQW) structures of GaAs and GaAlAs have been used for passive mode locking of commercial GaAs semiconductor diode lasers. We present an extended discussion of this application of the sensitive room-temperature excitonic absorption saturation in MQW material. We review the criteria for passive mode locking and discuss two methods—carrier diffusion and proton bombardment—for reducing the aborption recovery time without destroying the excitonic nonlinearity. A simple probabilistic theory is derived for the effect of bombardment on the excitonic effects that is in order-of-magnitude agreement with experiment. We have performed experiments using MQW material to mode lock a GaAs laser. A continuous train of pulses as narrow as 1.6 psec has been obtained with a pulse-repetition rate of 2 GHz.

1. INTRODUCTION

There has been a great deal of interest recently in optical switching and signal-processing devices that can operate at picosecond speeds and at low optical energies. In order for such devices to find practical applications, however, it is necessary to have available a simple, reliable, and compact source of picosecond optical pulses. In this paper, we describe the use of GaAs/GaAlAs multiple-quantum-well (MQW) material as a saturable absorber to mode lock a commercial semiconductor diode laser. Continuous trains of pulses as short as 1.6 psec have been produced.[1] This represents about a 1-order-of-magnitude improvement over previous results and is a major step along the road to the practical application of ultrafast digital photonic signal processing.

This work represents one of the first applications of the saturable excitonic absorption in MQW material. We will discuss two different approaches that we have taken to reduce the recovery time of the saturation. This is important not only for mode locking but also for many other applications.

Passive mode locking of diode lasers has been observed by several researchers since the first reports by Morozov *et al.*[2] and Bogdankevich *et al.*[3] Ippen *et al.*[4] reported pulses as short as 5 psec for a short time before laser failure in aged laser diodes. Van der Ziel *et al.*[5] reported bursts of subpicosecond pulses from photon-bombarded lasers, and Yokoyama *et al.*[6] reported similar results from aged diodes. More recently, Harder *et al.*[7] have reported 35-psec pulses from lasers with nonuniform current injection. These experiments have demonstrated the potential of diode lasers for picosecond or subpicosecond pulsing, but, until recently, no suitable stable saturable-absorber material has been available for diode-laser mode locking.

In the past few years, much experimental and theoretical work has been done on the optical characteristics of GaAs/GaAlAs MQW material.[8] It has been found that in these structures there are prominent room-temperature exciton resonances near the band edge (Fig. 1) and that the exciton absorption saturates at optical intensities ~10× lower than those required to saturate the GaAs band-to-band transition (Fig. 2). This has prompted us to study the use of GaAs/GaAlAs MQW material as a saturable absorber for mode locking GaAs diode lasers. In Section 2 of this paper we present a review of the theories of passive mode locking of diode lasers. These theories show that one important condition is that a saturable absorber have an absorption recovery time that is short in comparison with the gain recovery time. In Section 3 we show how this can be achieved with MWQ material. Section 4 describes our diode-laser mode-locking experiments, and in Section 5 we conclude with a discussion of promising areas for future progress.

2. THEORY OF PASSIVE MODE LOCKING

Early theory of passive mode locking of lasers was concerned with pulse formation in Q-switched lasers.[9] When picosecond mode locking of dye lasers was demonstrated, much work was undertaken to understand the role of gain and absorber dynamics in the mode-locking process. In particular, Haus analyzed the case of a fast saturable absorber.[10] Later, building on the analysis of New,[11] who showed how gain saturation could provide loss for the trailing edge of a mode-locked pulse even in the case of a slowly responding absorber, Haus published a theory of passive mode locking with a slow saturable absorber.[12] In that paper, he derived the conditions that must be satisifed in order to obtain stable mode locking. Because the case of a slow saturable absorber (i.e., one whose recovery time is long compared with the mode-locked-pulse duration) is the one normally encountered with practical

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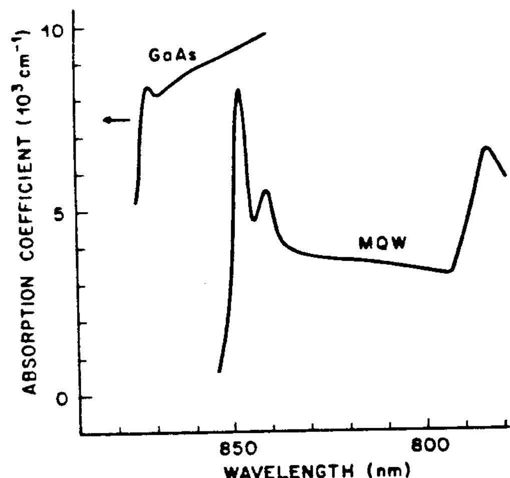

Fig. 1. Room-temperature linear-absorption spectra for bulk GaAs and GaAs/GaAlAs MQW samples near the band edge.[8]

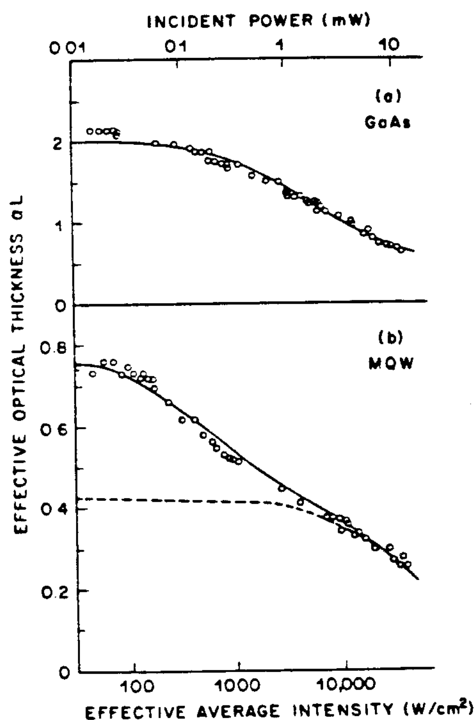

Fig. 2. Intensity dependence of optical absorption in GaAs and GaAs/GaAlAs MQW samples at room temperature measured with 5-μm spot.[6]

saturable-absorber materials,[13] we will discuss these conditions in some detail.

Figure 3 shows the time-dependent gain and absorption loss in a passively mode-locked laser system with a slow saturable absorber. Before the arrival of the pulse, the loss and gain are approaching their steady-state values, and the loss is larger than the gain. The leading edge of the pulse thus experiences loss. As the intensity of the pulse increases, however, we require *the loss to saturate faster than the gain*, so that the central part of the pulse will be amplified. When the gain becomes saturated below the unsaturable loss (mirror transmission, waveguide losses, etc.), the trailing edge of the pulse will again experience a loss. If *the recovery time of the loss is faster than that of the gain*, then the loss will remain greater than the gain everywhere except near the peak of the pulse, and spontaneous emission will not be able to build up between the pulses. Note also that the unsaturated gain is greater than the unsaturated loss so that laser oscillation can build up when the laser is turned on. On one round trip in the resonator, the combined action of the gain and loss saturation is to shorten the pulse by amplifying the center and attenuating the leading and trailing edges. In the steady state, this shortening is balanced by dispersive pulse broadening.

In order for the ideal slow saturable-absorber mode locking described in Fig. 3 to be possible, the two conditions in italics in the paragraph above must be satisfied by the saturable-absorber material. These conditions can be stated mathematically as

$$\sigma_A/A_A > \sigma_G/A_G \tag{1}$$

and

$$\tau_A < \tau_G, \tag{2}$$

where σ_A and σ_G are the effective cross sections for the absorber and the gain, respectively, A_A and A_G are the geometrical cross sections of the laser beam in the absorber and in the gain, and τ_A and τ_G are the lifetimes of the absorber and gain saturation effects. In Ref. 12, Haus shows how the pulse-width and mode-locking range depend on the gain and loss parameters. In order to have stable mode locking, it is necessary to have substantial dynamic changes in both gain and loss during the passage of a pulse. Thus it is desirable for the pulse-repetition rate to be of the order of τ_G, the gain recovery time.

The GaAs/GaAlAs MQW material can easily satisfy inequality (1), as demonstrated in Fig. 2. There is a saturation of the absorption that takes place at intensities ~10× lower than those required to saturate bulk GaAs. (The gain-saturation cross section is essentially the same as the absorption-saturation cross section). The physical reason why the MQW absorption can saturate more readily is that the saturation is due to the screening of excitons by free carriers—a mechanism qualitatively distinct from the filling of conduction and valence-band states in the GaAs laser material. The exciton screening effect is not strong in room-temperature GaAs because the exciton resonance is barely resolvable even

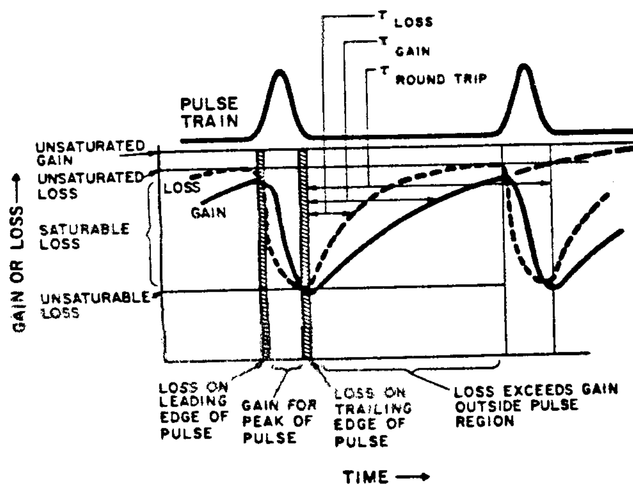

Fig. 3. Gain and loss dynamics for ideal mode locking with a slow saturable absorber.

in pure material. In laser material, excitons will be heavily screened owing to the high carrier concentrations already present. In MQW material, the exciton resonances are clearly resolved, so this sensitive saturation mechanism can be used.

Because MQW absorption saturation is due to screening of optically excited carriers, the absorption recovery time is governed by the carrier recombination time, which is 30 nsec for our unbombarded samples.[8] In order to satisfy inequality (2), we must find a way to reduce the absorption recovery time below the value of ~400 psec, which is the value of τ_G appropriate for GaAs lasers.[14] In Section 3 we discuss how this can be done.

3. MATERIAL STUDIES

In this section we discuss two techniques that we have explored for shortening the absorption recovery time in MQW structures. One involves focusing the light beam into the MQW structure; the other uses a controlled damaging of the structure by proton bombardment. Note that reducing the absorption recovery time increases the saturation intensity of the absorber but does not necessarily affect the absorption cross section, which is the important parameter for mode locking.

A. Tight Focusing

When carriers are generated by a tightly focused light beam in a small region of the MQW layers, they may diffuse laterally out of the interaction region in times comparable with or shorter than the recombination time. The relaxation time of the nonlinear absorption may then be described approximately as a combination of the two times,

$$\frac{1}{\tau_A} = \frac{1}{\tau_r} + \frac{1}{\tau_d}, \tag{3}$$

where τ_r is the recombination time and τ_d is the diffusion decay time. Using the two-dimensional diffusion equation, as appropriate for carriers diffusing in the plane of the MQW layers, and assuming a Gaussian-beam excitation, the diffusing decay time can be related to the diffusion coefficient D and to the Gaussian beam waist w_0 by the equation[8]

$$\tau_d = w_0^2/8D. \tag{4}$$

Experiments yield a value of $D = 13\ \mathrm{cm^2/sec}$ for ambipolar diffusion in the plane of the MQW layers for GaAs/GaAlAs MQW structures[6]; this is in good agreement with the known carrier mobilities. The parameter τ_d is, strictly speaking, a characteristic time, not an exponential lifetime. However, it may be concluded that for excitation beams with waist $w_0 \leq 17\ \mu\mathrm{m}$, the decay time is dominated by diffusion rather than by recombination. If the beam is focused to a waist of $<2\ \mu\mathrm{m}$, the absorption recovery time should be <400 psec. This should be possible to achieve optically, and so the lifetime criterion in inequality (2) can be satisfied. However, such a small spot size causes absorption saturation at low power levels.

B. Proton Bombardment

It would be useful to be able to reduce the lifetime without restricting the optical power at which absorption saturation occurs. It is well established that bombardment of semiconductors with high-energy ions increases the carrier recombination rate by generating damage centers, which serve as recombination sites. This has been verified with a variety of different ions and semiconductor materials, mainly in studies related to photoconductive devices.[15] Although this is a direct method for increasing the recombination rate, it is not clear that it can be used in MQW structures without severely affecting the excitonic absorption feature, thus reducing or eliminating the nonlinearity associated with it.

The effects of disturbing the crystalline structure of a semiconductor can generally be expected to perturb the states near the center of the Brillioun zone, as these have the longest wavelengths (smallest wave vectors) and hence are most sensitive to small densities of imperfections. However, these are the very states from which the exciton wave function is formed. Simple Fourier analysis shows that wavelengths up to of the order of the exciton Bohr diameter and/or thickness are required to make up the exciton wave function. If the density of damage centers is such that coherent wave functions of these wavelengths cannot exist (e.g., an average of one damage center per wavelength), then the exciton will be strongly perturbed. The exciton is strictly an extended state of the crystal and cannot be viewed as existing at only one point in the crystal, but an effectively equivalent statement of the same phenomenon is that if there is a crystalline defect or damage center within an excitonic volume, the exciton state may cease to exist. Thus we should expect that if there is a high probability of finding a damage center within any given exitonic volume, the excitonic absorption resonance may be destroyed. It is not clear what the precise nature of damage centers is, but if they are charged another mechanism can come into play that gives a similar prediction; when the density of charged centers is of the order of 1/(excitonic volume) the field from the charged centers is of the same order as the field that binds the exciton, and hence the exciton may be field ionized or otherwise strongly perturbed. Recent direct measurements with uniform fields[16] confirm that fields of the order of the classical ionization field do significantly perturb the exciton resonance when the fields are in the plane of the MQW layers. Although neither of these arguments is rigorous, they do suggest destruction of the exciton resonance when there is of the order of one damage center per exciton volume. Therefore we can construct simple probabilistic arguments to predict the bombardment densities at which the exciton resonance will disappear. The simplest such argument is to presume that the excitonic absorption and/or the nonlinearity associated with it is proportional to the probability of finding no damage center with a given excitonic volume. We derive the expression for this probability in Appendix A and hence obtain for the excitonic absorption strength

$$\alpha = \alpha_0 \exp\{-FA_x[1 - \exp(-L_x/L_d)]\}, \tag{5}$$

where F is the bombarding ion flux, A_x is the exciton area, L_d is the mean distance between damage centers on a damage center track, L_x is the exciton thickness, and α_0 is the initial absorption value. From Eq. (5) it can be estimated that heavy ions, which generate a nearly continuous track of damage centers, will strongly affect the absorption at a dose of $\sim 10^{12}$ ions/cm^2, which corresponds to about one damage track per exciton area.

In our experiments we used protons as the bombarding ions in order to ensure penetration through the entire thickness

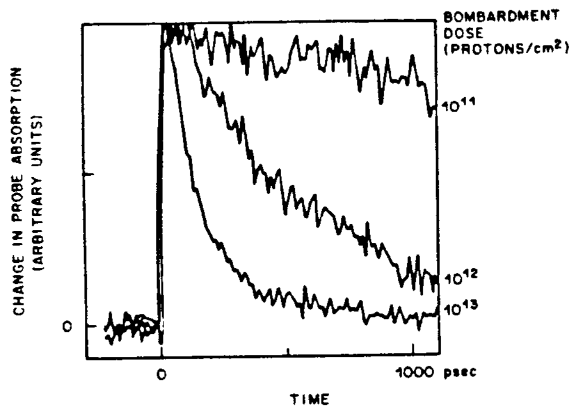

Fig. 4. Change in probe transmission as a function of time for annealed MQW samples.

Table 1. Recovery Time of Nonlinear Absorption for Proton-Bombarded MQW Samples[a]

Proton Dose (cm^{-2})	Recovery Time (psec)	
	Annealed	Unannealed
10^{10}	–	530
10^{11}	>3000	450
10^{12}	560	200
10^{13}	150	(27)
10^{14}	(33)	(<10)

[a] The unbombarded recovery time is 30 nsec. Values in parentheses refer to times measured in samples for which the excitonic nonlinearity is significantly reduced.

of the MQW structure. In this case we can expect that the damage tracks will not be continuous (range statistics indicate several hundred angstroms mean separation between damage centers), and hence higher doses may be possible without a significant change in the excitonic absorption.

We have performed a series of measurements of the absorption recovery times and the excitonic nonlinearities in proton-bombarded MQW structures.[17] The samples consisted of 80 periods of 102-Å GaAs layers, alternated with 101-Å $Ga_{0.71}Al_{0.29}As$ layers, grown by molecular-beam epitaxy over a GaAs substrate. The samples were bombarded with different doses of 200-keV protons. Some of the samples were annealed for 10 min at 300°C. The purpose of this gentle annealing is to eliminate shallow traps, which may anneal spontaneously, and thus ensure better long-term stability for these samples.

The recovery time of the excitonic absorption was measured using a conventional pump-probe scheme. We have used a tunable synchronously mode-locked LDS-821 dye laser, which produces 5–7-psec pulses spaced by 12 nsec. Figure 4 shows the change in probe transmission that is due to saturation by the pump, as a function of the delay between the pump and probe pulses, for an annealed sample bombarded with different proton doses. Table 1 summarizes the lifetime measurements for different proton doses in annealed and unannealed samples. As expected, annealed samples have longer lifetimes than their unannealed counterparts because of the removal of some damage centers. Values in parentheses refer to times measured in samples for which the excitonic nonlinearity is significantly reduced.

Figure 5 shows the (linear) absorption spectra of several unannealed samples with different proton-bombardment doses. It is observed that up to a dosage of about 10^{12} protons/cm^2 no significant change occurs. Higher bombardment doses cause both a decrease in the peak absorption and a broadening of the resonance. We believe that the slight change in the location of the peaks is not related to the bombardment and is the result of different well thicknesses in the various samples. These results are consistent with Eq. (5) with ~1000-Å spacing between damage centers. This is in reasonable agreement with the separation expected from range statistics given the accuracy with which we can determine L_d from the data (~a factor of 3). Similar spectra taken after annealing yield ~3000-Å spacing.

The nonlinear absorption spectra, which are shown in Fig. 6 for three annealed samples, are spectra of the change in probe transmission that is due to saturation by a pump beam of the same wavelength. Again we have seen no significant

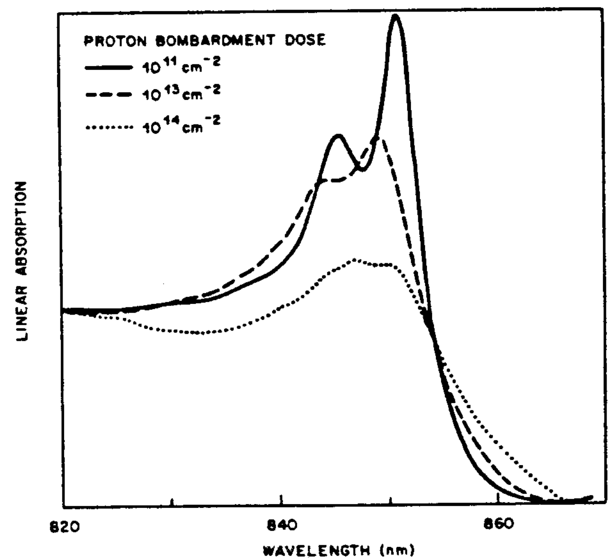

Fig. 5. Small-signal-absorption spectrum of unannealed MQW samples as a function of proton bombardment dose. The spectrum of a proton dose of 10^{11} cm^{-2} is indistinguishable from the spectrum of umbombarded material.

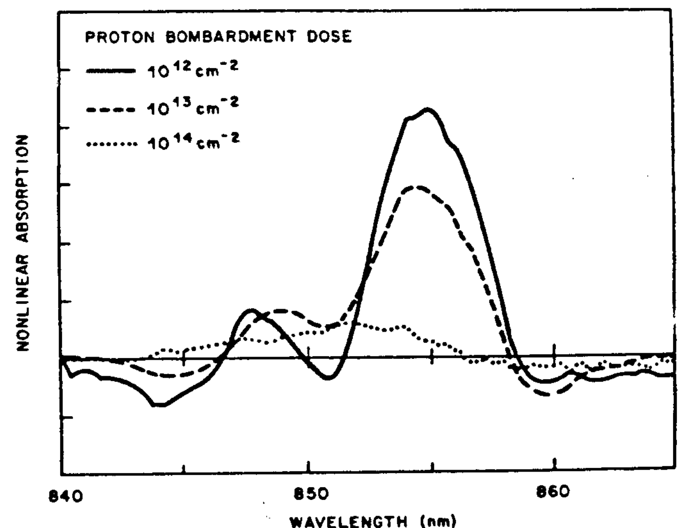

Fig. 6. Nonlinear absorption spectra for annealed MQW samples.

changes for proton dosage below 10^{12} protons/cm^2, a decreased response for 10^{13} protons/cm^2, and almost no response for proton dosage above 10^{14} protons/cm^2.

From this it is clear that a substantial reduction of the absorption recovery time can be induced by proton bombardment of MQW material before a significant change in the linear or nonlinear absorption characteristics occurs. A dose of 10^{13} protons/cm^2 seems to give a sufficiently short lifetime for mode-locking purposes (150 psec in annealed MQW material) without causing a major reduction in the nonlinearity.

4. MODE-LOCKING EXPERIMENTS

In our experiments we have used a commercial GaAs laser diode in an external linear cavity. The experimental setup is shown schematically in Fig. 7. The anamorphic prism pair was used to convert the elliptical output beam of the laser into a circular one. The MQW absorber was epoxied onto a highly reflecting mirror, and the GaAs substrate was removed by using a selective etch. An antireflection (A.R.) coating was then deposited over the etched surface. In the course of our investigations we have used a number of MQW samples, comprising between 40 and 80 quantum wells. The thicknesses of these wells were between 80 and 100 Å, corresponding to room-temperature excitonic absorption peaks in the range of 840–855 nm. The laser diode (Hitachi HLP1400) was modified by an A.R. coating on one facet. The quality of the coating could be estimated by measuring the output spectrum of the diode.[18] The residual reflectivity modulates the luminescence spectrum of the diode. From the modulation depth we found that the residual reflectivity, in some cases, was lower than 10^{-3}. As we shall see later, a low reflectivity is important for good mode locking.

Figure 8 shows the reflectivity spectrum of one such absorber–mirror assembly at low light intensity, where the absorption is unperturbed, and at higher light intensities. The power level of 2.5 mW corresponds to that in the laser under mode-locking conditions. The unsaturated reflectivity has two minima, which correspond to the light and heavy-hole excitons, with peaks at 839 and 846 nm, respectively, in this specific sample. The reflectivity at the heavy-hole peak is about 25% for low light intensities and increases to ~50% at 2.5-mW input power because of the saturation of the absorption line.

The laser with the absorber in it has a threshold current of approximately 70 mA, compared with 50 mA for the laser without the absorber. Figure 9 shows the output light intensity as a function of the drive current for three different configurations. The laser with the absorber often displayed some bistability near threshold. This behavior was usually associated with mode-locked operation and was seen when the

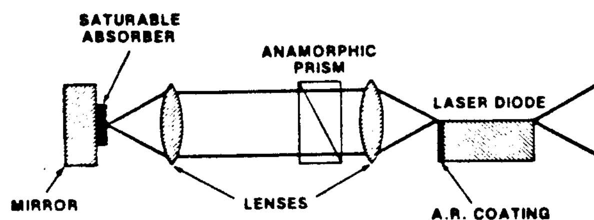

Fig. 7. Setup for mode-locking experiments.

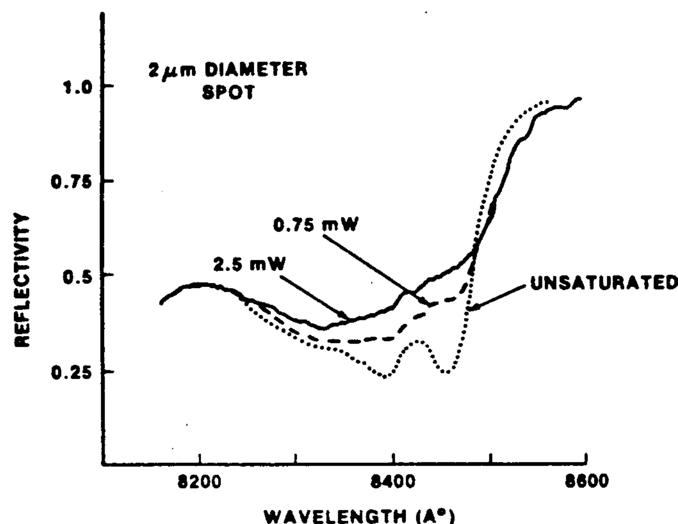

Fig. 8. Reflectivity spectrum of a mirror–MQW-absorber combination as a function of incident light intensity.

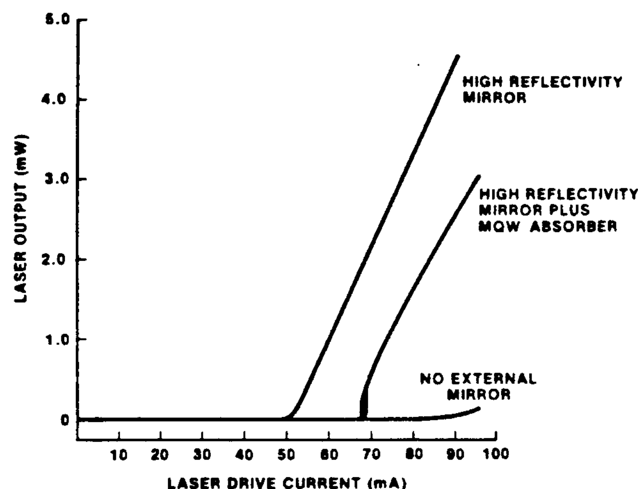

Fig. 9. Laser output characteristics for a Hitachi HLP 1400 laser diode with one facet A.R. coated (see text).

lasing wavelength was close to the peak of the exciton absorption. We found that we could tune the lasing wavelength by adjusting the lens–mirror and the lens–laser separations, thus using the chromatic aberrations in our cavity for wavelength selection.

The output of the laser was monitored simultaneously by a fast photodiode connected to a sampling oscilloscope, by an optical multichannel analyzer for spectral measurements, and by a second-harmonic-generation autocorrelator for pulse-width measurements.

In our first attempt to obtain mode locking, we tried to implement the method of tight focusing for reducing the absorber recovery time. The MQW absorber used was not treated by photon bombardment. We found that good mode locking could be achieved only by using certain specific points on the MQW sample. It is probable that the spot size on the absorber was not small enough to reduce the response time to below 0.4 nsec and that the attainment of mode locking in this case was assisted by the presence of defects at these points that were causing fast carrier recombination. This conclusion

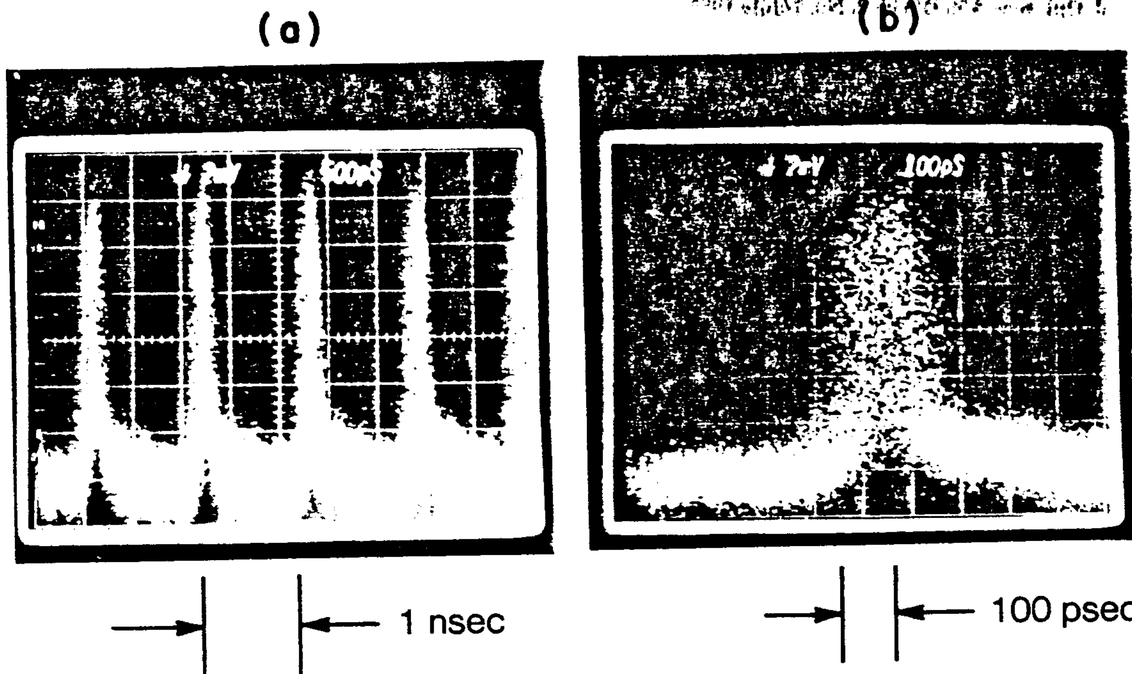

Fig. 10. Mode-locked laser output as observed with a fast photodetector (response time ~ 100 psec). (a) Pulse train, (b) expanded view of a single pulse.

is supported by the fact that these points were also characterized by higher laser thresholds.

More reliable mode-locked operation was obtained with proton-bombarded samples. Mode locking could be achieved at almost any point on the sample. The samples used were bombarded with 10^{13} protons/cm^2 and later annealed for 10 min at 300°C. According to Table 1, these samples have a 150-psec absorption recovery time. Figure 10 shows the photodetector response to a mode-locked output. The repetition rate of 1 GHz reflects the round-trip time in the cavity, which was typically 15 to 30 cm long. In the longer cavities we observed a tendency to settle into double pulsing, i.e., two

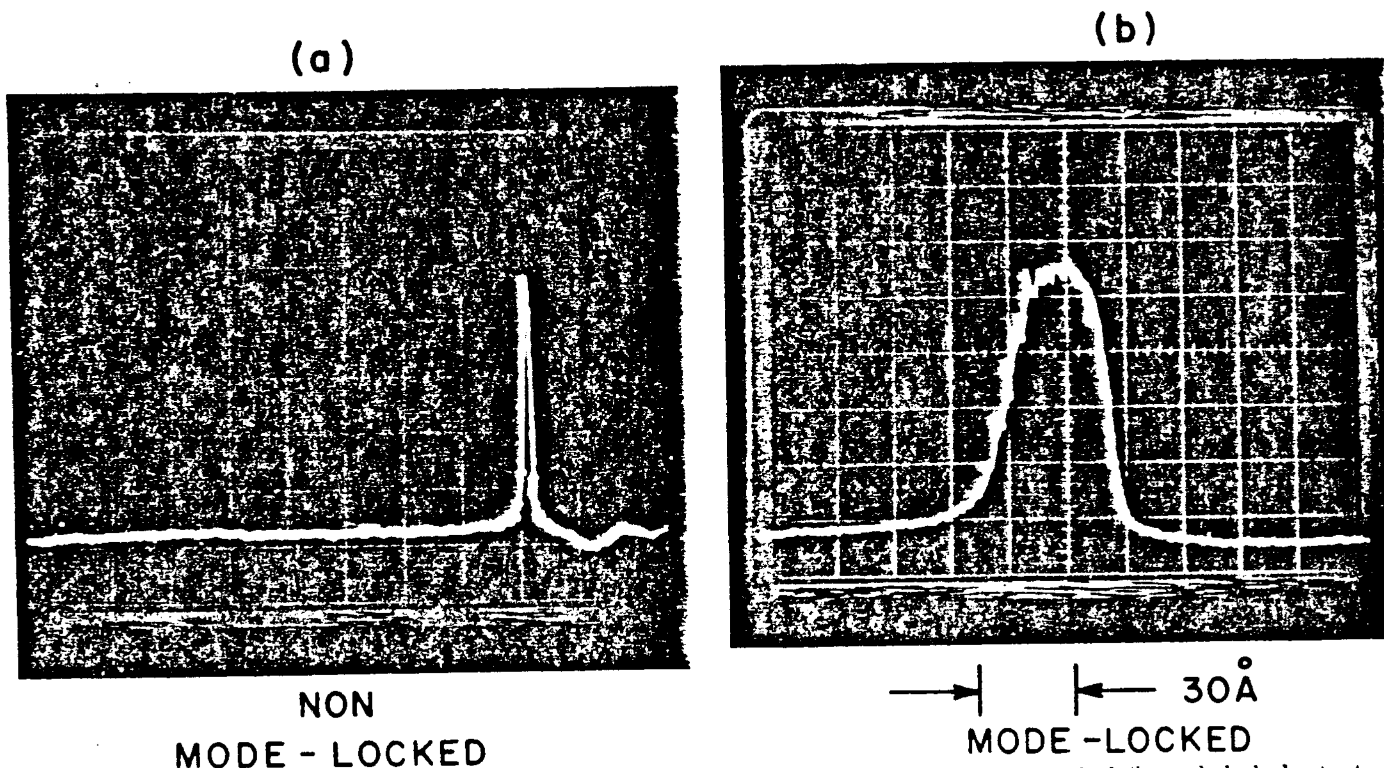

Fig. 11. Mode-locked laser spectrum observed with an optical multichannel analyzer. (a) Non-mode-locked, (b) mode-locked output. Both spectra are shown with a full-scale range of 200 Å.

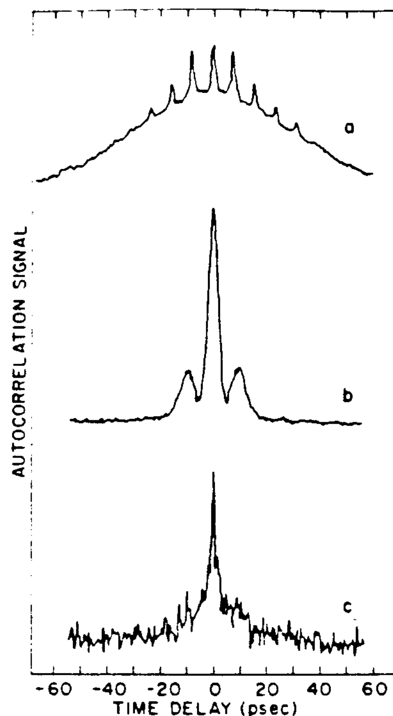

Fig. 12. Autocorrelation traces of mode-locked laser output pulses (see text).

pulses per round-trip time. This tendency was stronger at higher drive currents. Stable pulsing as seen in Fig. 10 could be obtained for periods of hours. Mode-locked operation was characterized by a significant spectral broadening. Figure 11 shows the spectra of a non-mode-locked and a mode-locked output. The former is probably single-mode operation, and it is observed either when the lasing wavelength is tuned beyond the absorption range of the MQW material or when the power level is high enough to saturate the absorber completely. The mode-locked spectrum is very wide, typically 30 Å, and often displays a structure that can be related to the diode-laser modes. This is a remarkable phenomenon, since the antireflection coating over the diode facet was of extremely good quality. Although mode locking could be obtained over a range of wavelengths within the absorption range of the MQW material, short pulses were obtained only when the laser was tuned to a specific wavelength, which was about 5 nm longer than the room-temperature exciton peak. We have some indications that the absorption peak at the light spot actually shifts by this amount to a longer wavelength when the laser is operating because of heating of the MQW absorber at that point. A local temperature increase of ~20°C would explain this shift.

Figure 12 shows several autocorrelation traces obtained from our mode-locked laser. The upper trace (Fig. 12a) shows a typical result that is obtained when the laser is not tuned to the optimal wavelength. Relatively broad pulses, with a FWHM of 20–50 psec, are typical. The autocorrelation signal is characterized by a multipeaked structure, with the peaks separated by 8 psec. This time can be identified as the transit time in the diode, and it reflects the substructure of the spectrum that is due to the diode modes. Better mode locking is obtained when the wavelength is tuned to the optimal point, just above the room-temperature exciton peak. Figure 12b shows a trace often obtained in these circumstances. The triple-peak autocorrelation is probably caused by a satellite pulse, which trails the main pulse. The FWHM of this main pulse, assuming a Gaussian pulse shape, is 3.3 psec. The shortest pulses are obtained with the unbombarded material close to laser threshold. In Fig. 12c we show the shortest pulse detected so far, with a FWHM of 1.6 psec.

5. CONCLUSIONS

In this paper we have described the results of our current studies of semiconductor diode mode locking. Clearly, much work remains to be done.

The recent observations of an ultrafast component to the exciton absorption saturation recovery[19] suggest that it may be possible to improve mode locking by utilizing this fast response along the lines proposed by Haus and Silberberg.[13] The fact that our narrowest mode-locked pulses are still ~5× broader than the minimum possible with the observed spectral bandwidth suggests that further pulse narrowing may be possible—perhaps by using some soliton pulse-shaping mechanisms.[20–22]

We believe that the photon-bombardment technique that we described to reduce the MQW absorber response time has broader applicability than the mode-locking applications discussed here. Proton-bombarded MQW material is a versatile nonlinear-optical material that should permit the development of low-power bistable optical devices and other all-optical switching elements that can be driven with laser diodes.

Other future work will involve the development of integrated designs, including laser, waveguide, and absorber all on the same substrate, and the extension of these mode-locking techniques to quaternary semiconductors lasers operating in the 1.3–1.5-μm wavelength region, which is of great importance for optical communications.

APPENDIX A: PROBABILITY OF FINDING NO DAMAGE CENTER WITHIN AN EXCITONIC VOLUME

In the case previously considered by Aspnes *et al.*[23] of bombardment by heavy ions in a three-dimensional semiconductor, the damage tracks formed have damage centers that are close together. Hence, if a damage track passes through a given exciton cross section, there will certainly be a damage center within the exciton volume; this leads to a simple planar problem whose solution is well known.[23] In our case, we cannot assume that the damage centers are closely spaced compared with the exciton thickness because we have used light ions (protons) in order to guarantee adequate penetration depth. Furthermore, it is not correct to assume that the resultant damage centers are in a uniform, isotropic, statistical distribution; the mean separation of damage centers along the length of the track is largely set by the mass of the ion, whereas the mean lateral separation of damage tracks is set by the dosage. Another way of expressing the lack of statistical uniformity is to note that the positions of damage centers are partially correlated; the probability of finding a second damage center can be higher directly below a first center because the centers tend to form along the approximately straight damage tracks.

Consequently, we will derive a new statistical argument from first principles. We will assume that (1) the lateral positions of the damage tracks are independent with a density

per unit area F equal to the bombarding flux, (2) the damage tracks are straight and perpendicular to the surface, (3) the positions of damage centers along a given track are independent with constant mean separation L_d, (4) the exciton volume can be approximated by a cylinder of thickness L_z and an area A_x perpendicular to the surface.

Consider an arbitrarily small volume Δv of cross-sectional area Δa and thickness Δl arbitrarily small. The probability p_v of forming a damage center in Δv is

$$p_v = p_a \times p_l, \tag{A1}$$

where $p_a = \Delta a \times F$ is the probability of a damage track's passing through area Δa and $p_l = \Delta l/L_d$ is the conditional probability that a damage center is formed within a length Δl given that a damage track passes through area Δa. Here we use the standard relation of conditional probability $p(B) = p(A) \times p(B \text{ given } A)$.

The probability $(1 - p_{Lz})$ of finding no damage center in a column of length L_z under the cross-sectional area Δa (still given that a damage track passes through area Δa) is then given by

$$(1 - p_{Lz}) = (1 - p_l)^{L_z/\Delta l}, \tag{A2}$$

i.e., the probability of finding no damage center in length Δl, $1 - p_l$, and no damage center in the next length Δl, and so on for all $L_z/\Delta l$ lengths Δl that go to make up the column of length L_z. (p_{Lz}) is the probability of finding at least one damage center in the column of length L_z given that a damage track passes through area a.] In the limit of small Δl,

$$(1 - p_{Lz}) = \exp(-L_z/L_d). \tag{A3}$$

Analogously to Eq. (A1), we can now write for the probability $p(aL_z)$ of finding at least one damage center in the column of length L_z under area Δa

$$p_{aLz} = p_a \times P_{Lz}. \tag{A4}$$

Therefore, since there are $A_x/\Delta a$ such columns in the exciton volume, we obtain for the probability P_x of finding no damage centers in an exciton volume

$$P_x = (1 - p_{aLz})^{A_x/\Delta a}, \tag{A5}$$

i.e., in the limit of small Δa

$$P_x = \exp\{-FA_x[1 - \exp(-L_z/L_d)]\}, \tag{A6}$$

from which one obtains Eq. (5) in Section 3.

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