Fast nonlinear optical response from proton-bombarded multiple quantum well structures

Y. Silberberg and P. W. Smith
Bell Communications Research, Holmdel, New Jersey 07733

D. A. B. Miller and B. Tell
AT&T Bell Laboratories, Holmdel, New Jersey 07733

A. C. Gossard and W. Wiegmann
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

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Proton bombardment is shown to shorten the recovery time of the excitonic absorption in GaAs/GaAlAs multiple quantum well saturable absorbers. The response time can be reduced from 30 ns to 150 ps without affecting the absorption characteristics or the saturation energy.

Multiple quantum well structures (MQW's) made of GaAs/GaAlAs have recently been shown to have great potential for use as a nonlinear material in optical switching and signal-processing devices.[1] The nonlinearity associated with room-temperature exciton absorption can be saturated at powers and wavelengths compatible with semiconductor laser diodes. These structures have appreciable absorption even though their total thickness is of the order of 1 μm, and hence can be optically accessed to within 1 (μm).[2] Passive mode locking of semiconductor diode lasers, using MQW's as saturable absorber elements, has already been shown to produce a continuous train of 1.6-ps pulses,[2] and bistable optical devices having MQW's have been demonstrated.[3]

One of the most important parameters which characterizes a nonlinear optical element is the temporal response. The typical recovery time of the absorption in GaAs/GaAlAs MQW's is 30 ns.[1] This slow recovery time limits the time response of devices made from this material. The availability of MQW's with an appreciably faster response time would make possible a wide range of fast optical signal-processing applications. In this letter we report our studies on proton-bombarded MQW's. We have found that it is possible to reduce the recovery time by more than two orders of magnitude, down to 150 ps, without greatly affecting the other desirable characteristics of the material. Additional lifetime reduction can be obtained, but is accompanied by a reduction of excitonic absorption and a decrease in the nonlinear response.

The saturation of the exciton absorption on a picosecond timescale has been shown to be due to the screening of the excitons by light-induced free carriers.[1] The absorption recovery time is thus governed by the recombination rate of these free carriers. The exciton, which is generated through the absorption of a photon, is decomposed within ~ 0.3 ps into free carriers.[1,4] These free carriers screen against the generation of additional excitons. The elimination of these carriers is therefore necessary in order to restore the original absorption value. This is usually accomplished by natural recombination, which was measured to have a characteristic time of 30 ns in undoped GaAs/GaAlAs MQW's.[1] One strategy which may be used to reduce this recovery time is to focus the light tightly onto the MQW's, so that the carriers diffuse out of the interaction region at a rate faster than the recombination time. This method was used successfully to reduce the lifetime to less than 1 ns in the mode-locking experiment of Ref. 2. Obviously, tight focusing can be used only in a limited range of applications. Furthermore, it is limited to low light powers, in order to avoid total saturation of the absorber.

A more direct approach is to increase the recombination rate by introducing recombination centers into the lattice. This can be done by bombarding the material with high-energy ions as has been demonstrated in a variety of semiconductors.[5–8] Although it is well established that ion bombardment enhances the recombination rate, it is not clear that it can be used in MQW's without severely affecting the excitonic absorption feature. We therefore need to study the influence of ion bombardment on both the lifetimes and the excitonic absorption features.

When a MQW is used as a saturable absorber, thicknesses ~ 1 μm are typical. To ensure that the ion bombardment reaches through the entire thickness without excessive accelerating voltages, relatively light ions must be used: in this study we used hydrogen ions (protons). Previous studies of the effects of bombardment on excitonic absorption[9] have used heavy ions which tend to produce effectively continuous tracks of damage. Under the assumption that a damage track passing through a given excitonic volume will prevent an exciton from existing there, the excitonic absorption is proportional to $\exp(-FA_x)$, where F is the bombarding fluence in ions per unit area and A_x is the exciton cross-sectional area. For $\sim$60-Å exciton radius (appropriate for $\sim$100-Å-thick quantum wells[1]) the excitonic absorption would be strongly influenced at $F\sim 10^{12}/\text{cm}^2$. For protons however, the distance between damage centers on a given damage track can be so large that there may not even be one damage center within an exciton thickness. If we hypothesize that one damage center within an excitonic volume is sufficient to prevent an exciton from being formed there, and assume that the damage tracks are straight lines, then we can extend the probabilistic argument to show that the excitonic absorption α should decrease according to

$$\alpha = \alpha_0 \exp\{-FA_x[1-\exp(-L_z/L_d)]\}, \qquad (1)$$

where L_d (assumed constant) is the mean distance damage centers on a damage track, L_z is the exciton thickness, and α_0

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TABLE I. Absorption recovery time for proton-bombarded MQW's.

Dose (proton/cm^2)	Recovery time (ps)	
	Annealed	Unannealed
10^{10}	...	530
10^{11}	> 3000	450
10^{12}	560	200
10^{13}	150	(27)
10^{14}	(33)	(< 10)

is the initial absorption value. For the MQW's, we may take L_z as the thickness of one GaAs well. If L_d is large compared with L_z, we expect that proton doses higher than $10^{12}/\mathrm{cm}^2$ will be possible without a significant change in the absorption characteristics.

Our MQW 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 1-μm $Ga_{0.71}Al_{0.29}As$ etch stop layer on a GaAs substrate. The samples were bombarded with different doses of 200-keV protons. Some of these samples were later annealed for 10 min at 300 °C, in order to eliminate very shallow traps, and thus to ensure a better long-term stability. The samples were then epoxied on sapphire disks, and the GaAs substrates were removed by a selective etch. An antireflection coating was evaporated over the samples to eliminate Fabry–Perot interferences.

Optical measurements were made using a tunable synchronously mode-locked LDS-821 dye laser. The output consisted of a train of 7-ps pulses spaced by 12 ns. The recovery time of the excitonic absorption was measured using a conventional pump-probe scheme at a wavelength of 848 nm. The results of these measurements are summarized in Table I. We observed a monotonic decrease in the recovery time, both in the annealed and unannealed samples. The values enclosed in parentheses in Table I signify a weak nonlinear response. In order to estimate the amount of damage to the excitonic feature, and the usefulness of these bombarded

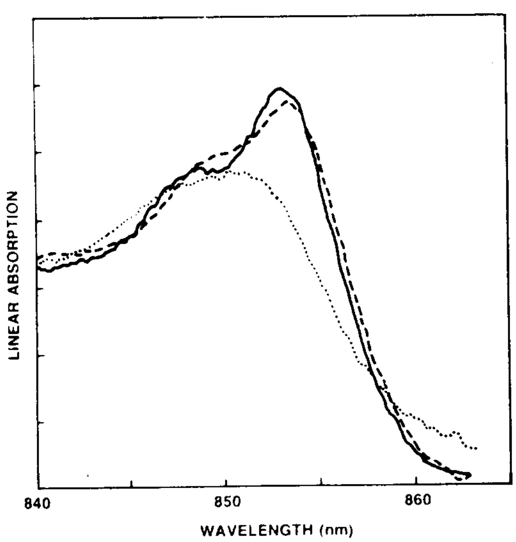

FIG. 1. Linear absorption spectra (in arbitrary units) for proton-bombarded MQW's. Solid line, $10^{12}/\mathrm{cm}^2$, dashed line, $10^{13}/\mathrm{cm}^2$, dotted line, $10^{14}/\mathrm{cm}^2$, all annealed.

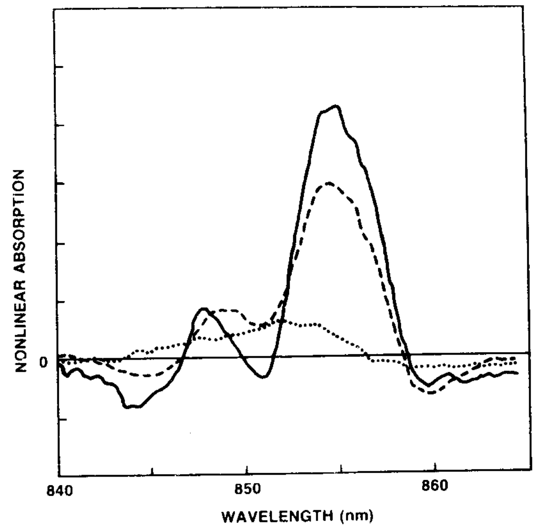

FIG. 2. Nonlinear absorption spectra (in arbitrary units) for proton-bombarded MQW's. Solid line, $10^{12}/\mathrm{cm}^2$, dashed line, $10^{13}/\mathrm{cm}^2$, dotted line, $10^{14}/\mathrm{cm}^2$, all annealed.

samples, we measured the linear and the nonlinear absorption spectra, which are shown in Figs. 1 and 2 respectively for annealed samples. The nonlinear absorption signal is the change in the probe transmission induced by the pump beam. The pump beam intensity was 0.5 mW, and it was focused to a 50-μm spot on the sample. We have taken care to eliminate that part of the nonlinear response which originates from a thermal shift of the absorption line rather than the saturation of absorption. The thermal component was removed by measuring the difference between two nonlinear spectra; the two spectra were taken with the probe pulse adjusted to arrive at the sample slightly ahead, then slightly behind, the pump pulse. The thermal contribution has a response time much longer than the 12-ns repetition rate of the laser, and thus has not decayed between the pulses. By taking the difference we therefore subtract the contribution of the slow thermal background and are left with the net nonlinear saturation spectrum.

Annealed samples which were bombarded by doses of up to $10^{12}/\mathrm{cm}^2$ have linear and nonlinear spectra which are indistinguishable from those of unbombarded samples, although their recovery time was substantially reduced. For a proton dose of $10^{13}/\mathrm{cm}^2$ there is some broadening of the exciton peaks, and a certain decrease of the nonlinear response. At a dosage of $10^{14}/\mathrm{cm}^2$ the excitonic absorption peak is smeared considerably, and the nonlinear response has almost vanished. The degradation of the excitonic feature with bombardment dose seems to be more pronounced for the unannealed samples, but not very much so. The lifetimes, on the other hand, are considerably shorter for the unannealed samples. It may be concluded that the damage centers removed by annealing have more effect on carrier lifetime than on the excitonic absorption. It may therefore be advantageous to relax the annealing conditions in order to obtain faster response times than 150 ps while still retaining the advantage of long-term stability.

Using Eq. (1) and the absorption spectra versus dose data, we estimate that if the sample is not annealed, the average distance between recombination centers along the proton track is approximately 1000 Å, and this distance increases to about 3000 Å after annealing. Note that part of the effect of the bombardment is to broaden the excitonic absorption peaks. This may be due to a reduction of exciton lifetime by the damage. For example, an inhomogeneous local field caused by the recombination centers could field ionize the excitons, leading to Stark broadening as has recently been observed with externally applied fields of $\sim 10^4$ V/cm parallel to the MQW layers.[10]

In conclusion, we have demonstrated that proton bombardment is an effective way to decrease the recovery time of the nonlinearity of MQW's, and that the response time can be reduced to less than 150 ps without substantially affecting the saturation energy. Note that since the recovery time has been reduced by more than two orders of magnitude, the saturation intensity in cw experiments has been increased by the same factor. Such fast-response material should find widespread use for a variety of optical switching and signal-processing applications.

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[1]D. S. Chemla, D. A. B. Miller, P. W. Smith, A. C. Gossard, and W. Wiegmann, IEEE J. Quantum Electron. **QE-20**, 265 (1984).
[2]Y. Silberberg, P. W. Smith, J. Eilenberger, D. A. B. Miller, A. C. Gossard, and W. Wiegmann, Opt. Lett. **9**, 507 (1984).
[3]H. M. Gibbs, S. S. Tarng, J. L. Jewell, D. A. Weinberger, K. Tai, A. C. Gossard, and W. Wiegmann, Appl. Phys. Lett. **41**, 221 (1982).
[4]W. H. Knox, R. L. Fork, M. C. Downer, D. A. B. Miller, D. S. Chemla, C. V. Shank, A. C. Gossard, and W. Wiegmann, in *Ultrafast Phenomena IV*, edited by D. A. Auston and K. B. Eisenthal, Proc. Fourth Int. Conf. Monterey, June 1984 (Springer NY, 1984), pp. 162–165.
[5]P. R. Smith, D. H. Auston, A. M. Johnson, and W. M. Augustyniak, Appl. Phys. Lett. **38**, 47 (1981).
[6]A. G. Foyt, F. J. Leonberger, and R. C. Williamson, Appl. Phys. Lett. **40**, 447 (1982).
[7]P. M. Downey and B. Schwartz, Appl. Phys. Lett. **44**, 207 (1984).
[8]R. B. Hammond, N. G. Paulter, and R. S. Wagner, Appl. Phys. Lett. **45**, 289 (1984).
[9]D. E. Aspnes, S. M. Kelso, C. G. Olson, and D. W. Lynch, Phys. Rev. Lett. **48**, 1863 (1982).
[10]D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus (unpublished).

