Nonlinear optical absorption and temporal response of arsenic- and oxygen-implanted GaAs

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(Received 4 September 1998; accepted for publication 8 February 1999)

We have measured the nonlinear optical absorption of arsenic and oxygen implanted epitaxial GaAs for a range of ion doses and annealing temperatures. The response time, \( \tau_A \), and a parameter, \( M_{\text{max}} \), which characterizes the performance of the structures as modulators, are both reduced by implantation, and correspondingly the nonbleachable losses are increased. We show that similar combinations of \( (\tau_A, M_{\text{max}}) \) can be achieved using either ion species and various combinations of dose and annealing temperatures. Furthermore, the data were all located on a well-defined curve in the \( (\tau_A, M_{\text{max}}) \) plane, provided amorphization, which occurs at high implant doses, was avoided. We deduce that there exists a limit to the modulation if a specific response time is required.

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Semiconductor saturable absorbers have become popular for applications in passive mode locking of solid-state lasers and all-optical switching.\(^2\) In both cases it is beneficial to have fast response times \( (\tau_A \approx 1 \text{ ps}) \) while retaining as much modulation as possible. There are two main approaches to the generation of a short \( \tau_A \) in semiconductors. One involves growth at low temperature (LT) using molecular beam epitaxy (MBE);\(^3\) the other is via ion implantation\(^4,5\) of high temperature grown material. LT-MBE growth has been used extensively for the fabrication of semiconductor saturable absorber mirrors (SESAMs) for passive mode locking,\(^1\) and more recently ion-implanted SESAM devices were also demonstrated.\(^6\) In this letter we present a comprehensive study of the nonlinear optical absorption modulation and response times of arsenic As- and oxygen O-implanted GaAs saturable absorbers annealed under different conditions. We show that similar combinations of modulation and response time can be realized using either ion species for various combinations of dose and annealing conditions, demonstrating that a rather general relation exists between modulation and response time in annealed ion implanted GaAs.

Our test structures were grown using metal organic chemical vapor deposition (MOCVD) and consisted of a Bragg reflector followed by 500 nm of GaAs. The Bragg mirror was centered at \( \lambda = 830 \text{ nm} \) and had a typical reflectivity of greater than 99% over a range of 70 nm. After growth the samples were implanted with 700 keV As ions or 250 keV O ions with doses ranging from \( 8 \times 10^{10} \) to \( 1 \times 10^{15} \text{ cm}^{-2} \) and annealed at 500, 600, or 700 °C for 20 min under arsine overpressure, or remained unannealed. For both ion species the displacement distribution lay solely inside the 500 nm GaAs layer. After implantation and annealing we applied an 830 nm antireflection (AR) coating to the samples to minimize Fabry–Perot effects. All optical measurements were done in reflection.

We performed two types of measurements using 830 nm, 80 MHz, 100 fs pulse trains. First, we measured the fluence dependent reflectivity \( R(F_P) = P_{\text{out}}/P_{\text{in}} \) using single beam excitation. Note that, due to the AR coating, changes in \( R(F_P) \) were dominated by absorption bleaching in the GaAs layer causing the reflectivity to increase. Since the underlying Bragg mirror is nonabsorbing for wavelengths above 750 nm it does not contribute to the nonlinear response but merely acts as a 100% reflector. Typical traces of reflectivity versus pulse fluence are shown in Fig. 1 for unimplanted as well as \( 8 \times 10^{13} \text{ cm}^{-2} \) O-implanted, 500 °C/20 min annealed samples. The experimental data were fitted numerically using a model for a traveling wave, two level saturable absorber.\(^7\) The fit is excellent for the most interesting fluence range whereas the disagreement found for fluences higher than 0.5 mJ cm\(^{-2}\) is mainly attributed to two photon and free carrier absorptions [(TPA) and (FCA)], which were not included in the model. The latter effects also account for the incomplete bleaching achieved in unimplanted GaAs. The numerical fits identified both the maximum modulation depth, \( \Delta R \) and the nonbleachable losses, \( \Delta R_{\text{ns}} \), consistently between samples, neglecting TPA and FCA. Figure 1 clearly shows that the modulation of the ion-implanted sample is smaller compared with the unimplanted case, and that \( \Delta R_{\text{ns}} \) has increased. In general we have found that \( \Delta R_{\text{ns}} \) increases and \( \Delta R \) decreases for higher doses or lower annealing temperatures. For instance, in Fig. 1 the O-implanted sample has \( \Delta R = 0.39 \) and \( \Delta R_{\text{ns}} = 0.26 \) which compares with the values for the unimplanted sample where \( \Delta R_{\text{GaAs}} = 0.59 \) and \( \Delta R_{\text{ns, GaAs}} = 0.1 \). We attribute these changes to deep levels.
created by the implantation, which give rise to additional transitions to states high in the bands. We expect that these transitions are difficult to bleach due to the large density of states high in the bands. The deep level transitions also contribute to the linear absorption resulting in a smaller linear reflectivity $R_{\text{lin}}$. However, our data show that any change in $R_{\text{lin}}$ was smaller than the uncertainty of $\pm 0.05$, arising from small differences in the AR coatings of different samples. This explains the slighter higher $R_{\text{lin}}$ of the O-implanted sample compared to the unimplanted case. Since $R_{\text{lin}} + \Delta R + \Delta R_{\text{ms}} \approx 1$ any increase in $\Delta R_{\text{ms}}$ primarily corresponded to a reduction of $\Delta R$.

In the second, pump-probe measurement we determined the time resolved differential reflectivity for different pump fluences using pump-probe delays as long as 300 ps. As an example, Fig. 2 shows the normalized differential reflectivity, $dR$, for O-implanted samples for different doses and annealing conditions compared with the unimplanted case. The pump-excited carrier density in the probed volume was approximately $2.5 \times 10^{18}$ cm$^{-3}$ at a pump fluence of 28 $\mu$J cm$^{-2}$. As in the first measurement, the dominating mechanism causing the reflectivity change is absorption bleaching in the GaAs layer. Contributions to $dR$ originating from changes in the real part of the complex refractive index of GaAs are negligible for our structures. This was confirmed through modeling assuming typical index changes $<0.1$. In all samples we observed a multieponential response corresponding to carrier thermalization and cooling within the first several 100 fs, and trapping and recombination via deep-level traps introduced by ion implantation. As is apparent from the sample data in Fig. 2, for increasing implantation dose or decreasing annealing temperature we obtained faster capturing-related signal decay indicating that the density of deep-level traps is higher for these conditions, as would be expected. Furthermore, the fastest signals decay with effectively a single exponential with time constants $<200$ fs and are dominated by carrier capturing, masking even the cooling process. Signals dominated by fast capturing evolve to negative differential reflectivity, i.e., photoinduced absorption (PIA), leveling off at a $dR$ value, usually a few percent or less, below zero. This is then followed by very slow (several 100 ps) evolution to $dR = 0$ (see inset in Fig. 2). This has been similarly observed in LT-GaAs, and attributed to slow recombination of captured carriers which provide additional transitions from midgap to high levels in the bands, after bleaching from band filling has ceased due to fast and complete capturing. In contrast to the case of LT-GaAs, we did not observe any slow residual bleaching attributable to completely filled traps even at excitation densities $>1 \times 10^{19}$ cm$^{-3}$. Further studies are needed to clarify the recombination dynamics. The nature and density of deep level traps in ion implanted GaAs is a research topic of current interest. Recently, it has been shown that, similar to LT-GaAs, ionized As antisites ($A_{\text{Ga}}^{\text{Ga}}$) are present in implanted and annealed GaAs. These deep level traps may also play a role in our samples.

Using the results of the modulation and pump-probe measurements, we have characterized the different samples by plotting their modulation behavior normalized to that of unimplanted GaAs, using the parameter $M_{\text{max}} = (\Delta R/1 - R_{\text{lin}})(1 - R_{\text{lin,GaAs}}/\Delta R_{\text{GaAs}})$, against an effective recovery time $\tau_A$. For consistency with previous work, $\tau_A$ was defined as the delay after which $dR$ had decayed to 1/e times its peak value. Note that annealing was very effective in removing implant-induced changes in $R_{\text{lin}}$, and hence for our annealed samples $M_{\text{max}} \approx \Delta R/\Delta R_{\text{GaAs}}$. However, $M_{\text{max}}$ was introduced for generality since a reduction in $R_{\text{lin}}$ (even if $\Delta R_{\text{max}}$ remained unaffected after implantation), should be reflected by a poorer assessment of modulator performance, and this would be masked if the potential effect of $R_{\text{lin}}$ were omitted. Samples implanted with different ions, doses, and annealed under different conditions are, therefore, represented as points in the ($\tau_A$, $M_{\text{max}}$) plane. This is shown in Fig. 3 for the complete set of annealed samples. The unannealed samples are not shown because, although extremely fast, their $M$ value was very low and varied due to in situ annealing under high excitation, making them less suitable for applications.

There is a clear tendency for the data to fall on a well-defined curve relating the achievable modulation to the speed of recovery. Unimplanted GaAs is located on the right hand side of this graph while increasing the implant dose, for a particular annealing condition, progressively moves the data to the left (shorter $\tau_A$ and smaller $M_{\text{max}}$). On the other hand, increased annealing temperatures, at a particular implanta-
tion dose, moves the points back to the right (towards longer $\tau_A$ and larger $M_{\text{max}}$). Generally more than an order of magnitude higher dose is required using an O-implant compared with As to achieve the same combination of response time and modulation depth. Figure 3 underlines the fact that a specific $(\tau_A, M_{\text{max}})$ combination can be achieved by using a range of different combinations of ion species, doses, and annealing conditions. This suggests that the types of defects in annealed As- and O-implanted GaAs are quite similar and the projected curve defines a rather general relationship between $\tau_A$ and $M_{\text{max}}$.

There are, however, outlying cases. First, for As doses above $1 \times 10^{13} \text{cm}^{-2}$, annealed at $500^\circ\text{C}/20$ min, $M_{\text{max}}$ drops rapidly while $\tau_A$ becomes pulse width limited, increasing the uncertainty on the location of the point As, $1 \times 10^{13} \text{cm}^{-2}$, $500^\circ\text{C}/20$ min. Second, As doses above $1 \times 10^{14} \text{cm}^{-2}$ actually reverse the general trend, such that while $M_{\text{max}}$ decreases $\tau_A$ increases. This is apparent in the lines for As, $600^\circ\text{C}/20$ min and $700^\circ\text{C}/20$ min, the latter indicating also the irreversibility of the change even at higher annealing temperatures. The departure most likely indicates a change in the irreversibility of the change even at higher annealing temperatures. The uncertainty on the location of the point As, $1 \times 10^{13} \text{cm}^{-2}$, $500^\circ\text{C}/20$ min and $700^\circ\text{C}/20$ min, the latter indicating also the irreversibility of the change even at higher annealing temperatures. The departure most likely indicates a change in the irreversibility of the change even at higher annealing temperatures.

In summary, we have studied nonlinear absorption modulation and the recovery time of a large set of As- and O-ion implanted GaAs samples. We showed that both $\tau_A$ and $M_{\text{max}}$ decrease through ion implantation with the decrease in $M_{\text{max}}$ being due to increasing nonbleachable losses. It was found that, for doses below amorphization, the data from all annealed samples lie on a well-defined curve in the $(\tau_A, M_{\text{max}})$ plane suggesting that the residual defects after annealing are of similar nature and that the curve defines the achievable $(\tau_A, M_{\text{max}})$ performance of ion implanted GaAs. Oxygen, being the lighter ion, can be implanted at higher doses than As without amorphization, creating more point defects and shorter $\tau_A$ while preserving highest possible modulation. For the $8 \times 10^{14} \text{cm}^{-2}$ O-implanted, $600^\circ\text{C}/20$ min annealed sample we measured a $\tau_A < 400 \text{fs}$ at 80% of the modulation of unimplanted GaAs. Overall, we have shown that ion implantation is a useful and extremely versatile process for producing ultrafast absorption modulation devices.

M. J. Lederer acknowledges the support of ElectroOptic Systems Pty, Ltd. and the Australian Government for the award of an APA Industry scholarship. H. H. Tan acknowledges the fellowship awarded by the Australian Research Council. The work at ETH Zürich was supported by the Swiss National Science Foundation.