Large ultrafast optical nonlinearities in As-rich GaAs

S.D. Benjamin, A. Othonos and P.W.E. Smith

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The measurement of large ultrafast bandgap-resonant optical nonlinearities in As-rich samples of GaAs that have been grown at low temperatures is reported. Light-induced refractive index changes of magnitude greater than 0.1 and with picosecond response times have been observed. These materials appear to be promising candidates for the fabrication of compact, ultrafast all-optical devices.

To handle the terabit data rates that are predicted to be required in future communications and computer systems, it will be necessary to have components that respond at subpicosecond speeds. The only way currently known to achieve these speeds is by using all-optical devices [1].

The nonlinear materials that are of interest for all-optical devices typically fall into one of two categories [2]. The first category involves nonlinear effects that occur for light energies significantly less than the bandgap of the nonlinear material. These nonlinearities tend to be small, but they have extremely rapid response times of the order of 10^{-14}s. The second category corresponds to electronic transitions induced by light energies near or above the bandgap energy. These nonlinearities tend to be large for light close to the bandgap of the material, but to have relatively slow response times (~10^{-9} s) as the recovery depends on the recombination of the light-induced carriers.

In this Letter we report a study of a nonlinear material (As-rich GaAs) that can be tailored to provide subpicosecond recovery times [3, 4] due to rapid recombination of the light-induced carriers while maintaining a large nonlinearity for light energies near the band edge. In fact, we have measured in this material the largest known nonlinear optical index changes with picosecond response.

A series of As-rich GaAs (GaAs:As) samples were grown by molecular beam epitaxy (MBE) at substrate temperatures of 250, 300 and 350°C, along with an additional series of samples grown at the same temperatures, but annealed at 600°C for 1 h before being removed from the MBE growth chamber. The structure of the samples consisted of a thin AlAs lift-off layer, 0.1 µm of $Al_{0.3}Ga_{0.7}As$; 2.0µm of GaAs:As and 0.1 µm of Si doped (2 x $10^{18}/cm^3$) GaAs with all layers grown at 580°C except for the GaAs:As. The epitaxial layers were lifted off the substrates [5], cemented onto glass slides, and antireflection coated on one side to eliminate Fabry-Perot resonances.

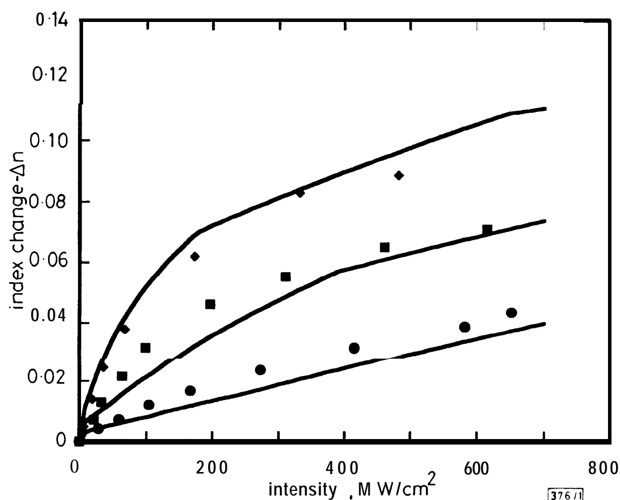

Fig. 1 *Index changes induced in 300 °C as-grown samples of GaAs:As by 3ps laser pulses of varying intensity and at a series of wavelengths that span the band edge of GaAs:As*

Theoretical curves were calculated from our rate-equation model of low-temperature-grown GaAs (see text)
—— theory
◆ 870nm
■ 875nm
● 880nm

The dependence of the index change in as-grown GaAs:As on the intensity of 3ps laser pulses is shown in Fig. 1 for a series of wavelengths that span the band edge of GaAs:As. The data points are obtained from Z-scan measurements [6] and represent the average index change induced in the material during the time the pulse is passing through the sample. The index changes saturate at high intensities as shown by the curves in Fig. 1. Index changes approaching -0.1 are observed for laser pulses at 870nm which is above the bandgap of the GaAs:As. Note that these changes are 2-3 times larger than those observed for normal high-temperature-grown GaAs [7]. The index changes measured are not caused by thermal effects because the Z-scan signature is opposite for a thermal (positive) nonlinearity and the laser pulse repetition rate was reduced to 5kHz to minimise any possible thermal problems. Also, from the known thermal parameters of GaAs, we calculate the thermal index change per pulse to be < 10^{4} for the highest fluences used in these experiments. When the pulse repetition rate is increased to 76MHz and the peak intensity is correspondingly reduced to maintain the same average power, no index change is detected by Z-scan. This shows that there is no measurable build up of index change over several pulses as would be expected if some of the measured index change were due to a slowly-responding photorefractive effect [8]. At the highest intensities, some of the measurement conditions are close to the limits of validity for the interpretation of Z-scan data. To verify our Z-scan data we made direct interferometric measurements [9] of the average index changes at 870nm and high fluences for all of the samples studied. The index changes measured in this way agree to within 25% with the Z-scan results.

Fig. 2 shows the dependence of the absorption on light intensity for a series of wavelengths using the same sample of as-grown GaAs:As. The data points represent the average absorption during the passage of the 3ps pulses through the sample.

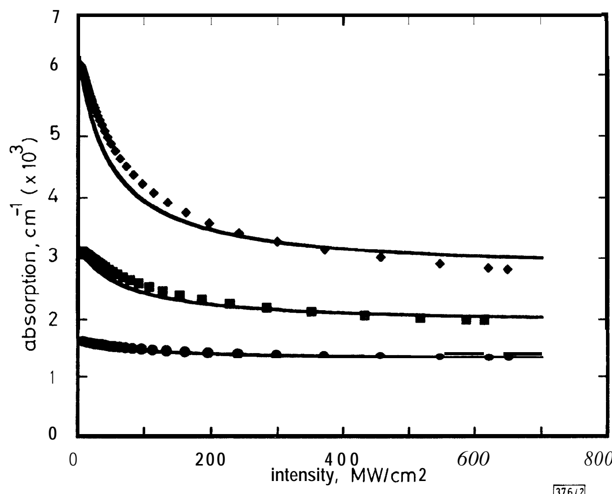

Fig. 2 *Absorption coefficient of 300 °C as-grown samples of GaAs:As as measured using 3ps laser pulses of varying intensity and at a series of wavelengths that span the band edge of GaAs:As*

Theoretical curves were calculated from our rate-equation model of low-temperature-grown using the same parameters that were used to fit the refrective index data of Fig. 1 (see text)

We used a standard pump-probe time resolved differential transmission spectroscopy arrangement with 150fs pulses tuned to 865nm to measure carrier lifetimes of 1.4ps for 250°C and 300°C as-grown samples and 3.3ps for the annealed samples. The samples grown at 350°C had much lower response times (60 – 90ps). These pump-probe measurements at high fluences showed a growth of absorption saturation near the band edge after the termination of the pump pulse. This behaviour is consistent with the rate equation model described below.

We have developed a simple phenomenological rate equation model to explain the observed index and absorption saturation. The model assumes that carriers in mid-gap states related to the presence of excess As can be excited by the incident light to levels high in the conduction band; that there is a contribution to the absorption proportional to the number of carriers in these mid-gap states; that a steady state population distribution is reached

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during the pump pulse; and that the index changes are caused by all carriers in the conduction band, and are given by the theory of Banyai and Koch [10] for GaAs which takes account of exciton effects, band filling, and bandgap renormalisation.

Our rate-equation model has been fitted to the experimental data as shown in [1, 2]. Relatively good agreement with the experimental measurements is found for the following conditions: decay rates for carriers from bottom of conduction band to mid-gap = measured absorption recovery time (1.4ps); decay rate for carriers high in the conduction band (dominated by decay to mid-gap states [11]): 0.3 1 ps; decay rate from mid-gap states to valence band: 2.8ps. The measured variation of unsaturated absorption with wavelength is accounted for by changes in both the transition probability and the saturation carrier density: the average saturation carrier density is set to the value found by Banyai and Koch [10] for GaAs near the band edge (1.3×10^{18}). Further details of this model will be described in a subsequent publication. It should be noted that with these assumptions, the model predicts absolute values of both axes of the plotted curves. There are no additional adjustable fitting parameters.

Samples that are annealed at 600°C for 1 h exhibit even larger index changes, as high as -0.13 at 870nm and high intensities. Also the index change saturates much more easily, so that large index changes are obtained at the lower intensities. These are both desirable effects but are offset by a substantial increase in loss ($\sim 2000 cm^{-1}$) in the annealed samples. This increase in loss appears to vary as $1/\lambda^4$ and is likely due to Rayleigh scattering from the As precipitates that form during annealing. A high density of large precipitates would also be expected to form in the thin Si doped cap layer [12]. A major part of the loss increase may be due to scattering from this surface layer. The as-grown samples were subjected to a short anneal at 580°C during the growth of the Si doped cap layer and these samples also appear to exhibit some loss (-500 cm-*) due to scattering.

One figure of merit [13] which is useful in evaluating nonlinear materials for optical device operation (W) is a measure of the phase change that can be obtained in an absorption length. $W > 1$ is required for good device performance. The loss in the annealed samples is so large that W is much less than unity for all wavelengths around the band edge. Our as-grown samples, however, have W of the order of unity or better, in the region just below the bandgap where the absorption is small but the index change is still substantial. A precise evaluation of W in the low absorption region is not presented due to the difficulty of measuring the absolute absorption in these very thin samples.

In conclusion, we have reported measurements of light-induced nonlinear index and absorption changes in arsenic-rich GaAs. They are in good agreement with a simple phenomenological rate equation model which includes the effects of mid-gap states. These changes recover in times that can be as short as ~1ps, and are, to our knowledge, the largest nonlinear index changes ever observed with picosecond response. We believe that GaAs:As is a promising material for the fabrication of ultrafast all-optical devices.

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