Ultrafast dynamics of nonlinear absorption in low-temperature-grown GaAs

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(Received 18 October 1995; accepted for publication 24 February 1996)

We present the results of a study of the subpicosecond dynamic behavior of optically induced absorption changes in low-temperature-grown GaAs. We show that the observed behavior is dominated by mid-gap trap states, and can be accurately modeled by the rate equations previously developed to describe quasi-cw results. Our data give the first approximate values for trap emptying times in this material. © *1996 American Institute of Physics.* [S0003-6951(96)04518-4]

Gallium arsenide that is grown at lower-than-normal growth temperatures has been shown to contain excess arsenic which gives this material some unique properties.[1] Of particular interest for ultrafast photonic applications is the fact that this material exhibits an enhanced light-induced refractive index change as large as 0.1 for light resonant with the band gap,[2,3] and that the carrier lifetime can be reduced from tens of nanoseconds in standard growth temperature GaAs to picoseconds or even subpicoseconds in annealed low-temperature-grown material.[1]

In previous publications[2,3] we showed that low-temperature-grown GaAs exhibits large light-induced absorption and refractive index changes, and that these changes can be accurately modeled by a set of rate equations which take into account the dominant role that traps play in the carrier dynamics. These previous studies, however, were done with pulses of the order of, or longer than, the key relaxation rates. Thus the detailed ultrafast dynamics were not revealed. In this letter, we present a study of ultrafast low-temperature-grown GaAs dynamics with a resolution of 150 fs. Previous measurements of the dynamics of the nonlinear absorption at lower carrier densities[1] exhibited a single exponential decay. We find that at higher carrier densities the dynamics are quite complex due to an induced absorption related to carriers in the traps which dominates the observed dynamics. This additional component of nonlinear absorption must be included for accurate modeling of ultrafast photonic devices based on low-temperature-grown GaAs. This study also allows us for the first time to estimate the trap emptying time in this material. This parameter is of crucial importance for ultrafast photonic device applications.[4]

We have developed a rate equation formalism to model the carrier dynamics in low-temperature-grown GaAs.[2,5] The processes included in the model are indicated in Fig. 1, which shows a band diagram with additional mid-gap states created by the excess arsenic. These mid-gap states, which have been ascribed[6] to EL2-like defects with concentrations as high as several times $10^{20}/\mathrm{cm}^3$, rapidly trap excited carriers. The assumptions in our rate-equation model are as follows: (a) N, the population of carriers at the bottom of the conduction band, contributes to absorption saturation; (b) both N and n, the population of carriers in excited states within the conduction band, contribute to the refractive index change; and (c) N_T, the population of carriers in the traps, contributes to the absorption but not to the index changes.

We describe the time dynamics of the carriers in low-temperature-grown GaAs by the following set of differential equations:

$$\frac{dN}{dt}=\frac{I\alpha}{h\nu}-\frac{N}{\tau_1}+\frac{n}{\tau_3}, \tag{1}$$

where α is the band-to-band absorption coefficient, I is the incident intensity of light with photon energy $h\nu$;

$$\frac{dN_T}{dt}=-\frac{I\alpha_T}{h\nu}-\frac{N_T}{\tau_2}+\frac{N}{\tau_1}+\frac{n}{\tau_4}, \tag{2}$$

where α_T is the absorption coefficient from the traps to excited states in the conduction band; and

$$\frac{dn}{dt}=\frac{I\alpha_T}{h\nu}+\frac{I^2\beta}{2h\nu}-\frac{n}{\tau_3}-\frac{n}{\tau_4}, \tag{3}$$

where β is the two-photon absorption coefficient. In all cases the decay times (τ) refer to the relaxation processes indicated in Fig. 1. The total absorption is given by

$$\alpha_{\mathrm{total}}=\alpha+\alpha_T+\beta I=\sigma N_0(1-N/N_0)+\sigma_T N_T+\beta I, \tag{4}$$

where σ is the cross section for band-to-band transitions, N_0 is the saturation carrier density,[7] and σ_T is the cross section for absorption from the traps to the (n) excited states in the conduction band.

Light resonant with the band gap will excite electrons to the bottom of the conduction band (N). After some time,

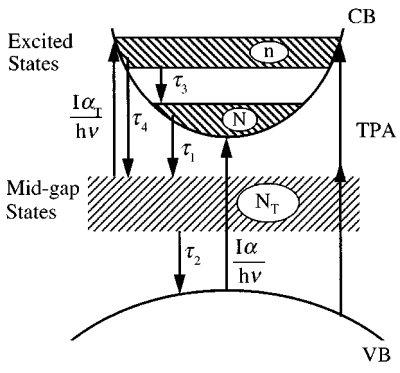

FIG. 1. Band diagram of low-temperature-grown GaAs showing the key excitation and decay processes in the rate equation model. α is the band-to-band absorption coefficient, and α_T is the absorption coefficient due to carriers in the midband states. CB and VB refer to the conduction and valence bands, respectively. The two-photon excitation process is designated by TPA.

 0003-6951/96/68(18)/2544/3/$10.00

these carriers will be trapped in the mid-gap states (N_T) from where they can be optically excited to upper states (n) in the conduction band. This process gives rise to an additional absorption mechanism in low-temperature-grown GaAs, and can produce a substantially larger carrier concentration in the conduction band than is possible with just band-to-band absorption. This is because although the population at the bottom of the conduction band (N) saturates at $N_0 = 1.9 \times 10^{18}/\text{cm}^3$ (see Ref. 2), for sufficiently intense pump light, the populations in the mid-gap states (N_T), and in the upper states (n) can continue to increase throughout the duration of the pump pulse. For the conditions appropriate for the data reported in Ref. 2, our model indicates that the population (n) can reach a value of $1.3 \times N_0$ during the 3 ps excitation pulse, and that this results in an index change of approximately two times the maximum change observed for normal GaAs with a low concentration of mid-gap states—in good agreement with our experimental results. For sufficiently intense pump light, the population (n) is also enhanced by two-photon absorption [Eq. (3)].

A series of low-temperature-grown GaAs samples were grown by molecular beam epitaxy (MBE) at substrate temperatures of 250, 300, and 350 °C, and lightly annealed during the growth of a normal-growth-temperature cap layer. The structure of the samples consisted of a thin AlAs lift-off layer; 0.1 μm of $Al_{0.3}Ga_{0.7}As$; 2.0 μm of low-temperature-grown GaAs; and 0.1 μm of Si-doped ($2 \times 10^{18}/\text{cm}^3$) GaAs cap layer. All layers were grown at a normal growth temperature of 580 °C except for the low-temperature-grown GaAs. The epitaxial layers were lifted off from the substrates,[8] cemented onto glass slides, and antireflection coated on one side to eliminate Fabry–Perot resonances.

Experiments were performed using 150 fs pulses from a mode-locked Ti-sapphire laser tunable around the band edge of the sample material. The absorption was probed with a variable delay after an intense saturating pump pulse centered at zero delay. Initial experiments with a relatively weak saturating pulse exhibit a fast rise followed by a single exponential decay of absorption saturation (due to band filling) providing a direct measure of the trapping time, τ_1. At higher pump intensities the dynamics are quite complex as shown in Fig. 2 by our experimental pump-probe measurements at $\lambda = 870$ nm of absorption dynamics of low-temperature-grown GaAs. The *absorption saturation* due to band filling is present; however, it is offset early on by an *increase in absorption* that decays away on a time τ_2 and causes the negative dip seen in Figs. 2(a) and 2(b). This additional absorption process is optical excitation of carriers from the trap levels back into the conduction band. The dynamics of this additional absorption can be described as follows. Near zero delay, two-photon absorption of the pump places carriers high in the conduction band where they are then rapidly trapped in a time τ_4 comparable to the resolution of our experiment. This leads to a rapid increase in the trap related absorption as evidenced by the rapid decrease in transmission near zero delay (falling edge of sharp peak). The related increase in absorption due to carriers in the traps then decays away as the traps empty with a time τ_2. The samples in Fig. 2 were chosen to illustrate the absorption dynamics for the cases $\tau_1 > \tau_2$ and $\tau_1 < \tau_2$.

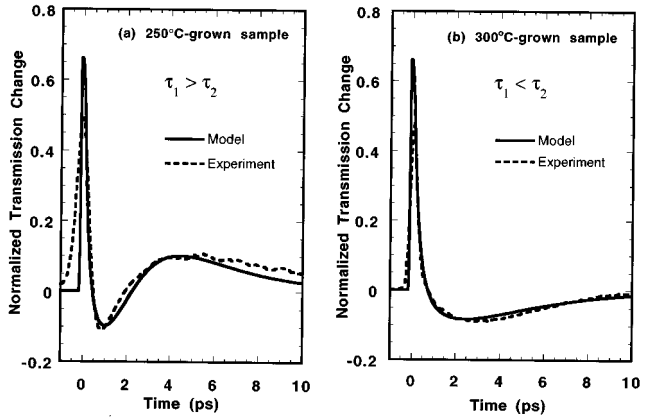

FIG. 2. Absorption dynamics of lightly annealed low-temperature-grown GaAs after excitation by an intense 150 fs pulse at 870 nm. The solid lines are the dynamic behavior predicted by our rate-equation model. (a) Sample grown at 250 °C where $\tau_1 > \tau_2$. (b) Sample grown at 300 °C where $\tau_1 < \tau_2$.

The solid lines show a fit of our rate equation model to the experimental data. The two-photon absorption coefficient β was directly measured for these samples and was found to be 35 cm^2/GW. We also measured the absolute value of the trap absorption in these samples[9] and found $\sigma/\sigma_T \sim 50$. The value of N_0 was taken from the fit to the quasi-cw (continuous wave) data in Ref. 2 to be $1.9 \times 10^{18}/\text{cm}^3$. With these parameters, the rate equations were fitted to the experimental measurements of absorption dynamics with τ_2, τ_3, and τ_4 as fitting parameters. For the 250 °C-grown sample, a good fit was obtained with $\tau_1 = 3.0$ ps, $\tau_2 = 1.4$ ps, $\tau_3 = 100$ ps, and $\tau_4 = 0.60$ ps [Fig. 2(a)]. For the 300 °C-grown sample, we found a good fit with $\tau_1 = 1.4$ ps, $\tau_2 = 3.0$ ps, $\tau_3 = 100$ ps, and $\tau_4 = 0.31$ ps [Fig. 2(b)]. In each case the fit specified is unique since a good fit cannot be obtained if any of the fitting parameters are varied by more than 10%. The rapid τ_4 relaxation times indicate that the trapping times for excited carriers are significantly shorter than those for carriers near the band edge. The long τ_3 times can be understood as a manifestation of the phonon bottleneck effects in low-temperature-grown GaAs reported in Ref. 10.

Table I compares, for the 300 °C-grown sample, the time constants found from the rate equation fits for the 150 fs data with the quasi-cw (3 ps) data fits reported in Ref. 2. It can be seen that very good agreement is obtained, even though for the 150 fs case two-photon absorption effects play an important role, and no assumptions of steady-state carrier concentrations under optical excitation are made. This gives us confidence in the validity of our model, and in the accuracy of

TABLE I. Comparison of the time constants used to fit rate equation model to experimental data for lightly annealed 300 °C grown GaAs excited at 870 nm with (a) 3 ps pulses (quasi-cw) as reported in Ref. 2, and (b) 150 fs pulses as reported in this letter.

3 ps experiments	150 fs experiments
$\tau_1 = 1.4$ ps	$\tau_1 = 1.4$ ps
$\tau_2 = 2.8$ ps	$\tau_2 = 3.0$ ps
$\tau_3 = \infty (\gg \tau_4)$	$\tau_3 = 100$ ps
$\tau_4 = 0.31$ ps	$\tau_4 = 0.31$ ps

the decay times found from the fits to the experimental data.

In conclusion, we have shown that traps play a dominant role in the ultrafast absorption dynamics of low-temperature-grown GaAs, and have shown how our measurements of absorption dynamics provide for the first time estimates of the trap emptying times. We have verified the validity of our rate equation model for this material, and have reported variations of key parameters with growth conditions. Future work will clarify the dependence of these parameters on material growth and annealing conditions, and indicate how the material can be optimized for ultrafast photonic switching applications.

We would like to thank J. Harbison and his colleagues at Bellcore who provided the low-temperature-grown GaAs samples used for these experiments, and S. Dzioba at Bell Northern Research Ltd. for the AR coatings. This work was supported in part by the Natural Sciences and Engineering Research Council of Canada, and the Ontario Laser and Lightwave Research Centre.

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