Subpicosecond carrier lifetime in GaAs grown by molecular beam epitaxy at low temperatures

S. Gupta, M. Y. Frankel, J. A. Valdmanis, J. F. Whitaker, and G. A. Mourou
Ultrafast Science Laboratory, Room 1006 IST Building, University of Michigan, 2200 Bonisteel, Ann Arbor, Michigan 48109-2099

F. W. Smith and A. R. Calawa
Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts 02173-9108

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Epitaxial GaAs grown by molecular beam epitaxy (MBE) at low substrate temperatures is observed to have a significantly shorter carrier lifetime than GaAs grown at normal substrate temperatures. Using femtosecond time-resolved-reflectance techniques, a subpicosecond (<0.4 ps) carrier lifetime has been measured for GaAs grown by MBE at ~200°C and annealed at 600 °C. With the same material as a photoconductive switch we have measured electrical pulses with a full-width at half-maximum of 0.6 ps using the technique of electro-optic sampling. Good responsivity for a photoconductive switch is observed, corresponding to a mobility of the photoexcited carriers of ~120–150 cm^2/V s. GaAs grown by MBE at 200 °C and annealed at 600 °C is also semi-insulating, which results in a low dark current in the switch application. The combination of fast recombination lifetime, high carrier mobility, and high resistivity makes this material ideal for a number of subpicosecond photoconductive applications.

The development of ultrashort-pulse mode-locked laser systems has resulted in new techniques for the generation and detection of picosecond and subpicosecond electrical transients.[1-3] Among these, the use of semiconductor photoconductive switches are the most popular, because these devices can be used to efficiently generate signals and to generate and detect electrical transients in guided media or free space. Also, the semiconductor growth and processing techniques available for tailoring the properties of these materials enhance their versatility. The minimum attainable electrical pulsewidth from a photoconductive element is limited by a number of factors such as the laser pulsewidth, circuit parameters of the generation and detection site, and the carrier lifetime in the semiconductor. With the use of femtosecond lasers and photolithographically defined millimeter-wave co-planar structures, the limits to speed imposed by the first two factors can be reduced. To shorten the carrier lifetime of a semiconductor layer, impurity doping of the semiconductor,[4] growth of polycrystalline or amorphous material,[5] and damage by ion implantation[6] can be used. Earlier we reported that photoconductive switches based upon GaAs grown by molecular beam epitaxy (MBE) at low temperatures showed fast response (1.6 ps) and good responsivity in unoptimized structures.[7] In this letter we extend our earlier study[8] of the photoresponse of low-temperature (LT) GaAs using both a femtosecond transient reflectance technique and photoconductive switching measurements. From both experiments we have observed a subpicosecond carrier lifetime for LT-GaAs grown at ~200 °C.

The 2-μm-thick, (100)-oriented epitaxial films discussed here were grown by MBE at substrate temperatures of 400, 350, 300, 260, 200, and 190 °C. For all the growths an As_4 source was used, and the samples were mounted on the same Mo block using In solder. The growth rate was 1.0 μm/h, and the As/Ga beam-equivalent-pressure ratio was 10. Pieces of the LT-GaAs samples were annealed inside the growth chamber under an As overpressure, just after the completion of the growth, by raising the substrate temperature to 600 °C for 10 min.

A number of papers have reported novel material properties of as-grown and annealed LT-GaAs layers, especially those grown at ~200 °C.[9-11] For photoconductive-switch applications, the most relevant properties of both as-grown and annealed 200 °C LT-GaAs are that the materials are crystalline and yet contain a high density ($>10^{18}$ cm^{-3}) of point defects as As antisites, As interstitials, and Ga-related vacancies.[9,11] In addition to a high density of point defects, annealed 200 °C LT-GaAs grown in the Lincoln Laboratory MBE system also contains small (<5 nm) As precipitates[12] at densities of $\sim 3\times10^{16}$ cm^{-3}. The aforementioned point defects can act as recombination and trapping centers. Assuming simple Shockley–Read–Hall theory for the recombination mechanism of the photoexcited carriers, and using a density $N\sim10^{18}$ cm^{-3} for the deep levels, a capture cross section $\sigma\sim10^{-13}$ cm^2 (a typical value for deep levels in GaAs), and thermal velocity v_{th} at $T=300$ K, we estimate that the carrier lifetime $\tau=1/(N\sigma v_{th})$ in as-grown and annealed LT-GaAs to be less than 1 ps.

Although as-grown LT-GaAs is relatively conducting ($\rho\sim10$ Ω cm) at room temperature, annealed LT-GaAs is semi-insulating ($\rho\sim10^7$ Ω cm).[11] Despite the high density of point defects and As precipitates, the Hall mobility at room temperature in annealed LT-GaAs is relatively high (~1000 cm^2/V s).[11] Therefore, LT-GaAs grown at ~200 °C and subsequently *in situ* annealed has the desired properties of a fast photoconductor; namely, a short carrier lifetime, high carrier mobility, and high resistivity.

To measure the photoexcited carrier lifetime of as-grown and annealed LT-GaAs, an all-optical pump-probe experiment[6,13] was done using 100-fs laser pulses from a balanced colliding-pulse mode-locked (CPM) dye laser ($\lambda = 620$ nm). In this experiment, the pump beam generates a hot electron-hole plasma, which thermalizes to a quasi-equilibrium distribution in ~ 1 ps[14] through the processes of carrier-carrier and carrier-LO phonon scattering. The average pump power of ~ 5 mW, focused to a 20-μm-spot on the sample generates an initial photoinjected carrier density of $\sim 1.0 \times 10^{18}$ cm^{-3} at the surface, assuming an absorption coefficient of 4×10^4 cm^{-1}. The dominant effect of this injected carrier population is to induce absorption nonlinearities ($\Delta\alpha$) through the mechanisms of bandfilling, band-gap renormalization, and free-carrier absorption.[15] The $\Delta\alpha$ can be related to changes in the refractive index (Δn) by the Kramers–Kronig relations. Following the analysis of Bennet *et al.*,[15] and assuming that a quasi-equilibrium distribution has been reached in about a picosecond, we estimate an initial $\Delta n \sim 5.8 \times 10^{-4}$. For such small changes in Δn, the time evolution of $\Delta n(t)$ will essentially follow the decay in the photoexcited carrier population resulting from trapping/recombination at deep levels, surface recombination and diffusion of nonuniformly generated carriers. For timescales of less than ~ 80 ps, as in our case, the primary contribution to the change in $\Delta n(t)$ is due to trapping/recombination at deep levels, as the other processes occur on a slower time scale.

Figure 1(a) shows the normalized transient reflectivity data for two unannealed LT-GaAs samples grown at 200 and 260 °C (inset). Figure 1(b) is a plot of the observed signal decay times for a series of unannealed LT-GaAs epilayers plotted as a function of the LT-GaAs growth temperature, where the decay time is obtained from the initial $1/e$ slope of reflectivity transient. For comparison, a typical range of carrier lifetime for Cr-doped semi-insulating GaAs substrates is also shown in Fig. 1(b). From the measured peak amplitudes of the reflectivity for the LT-GaAs epilayers and the Cr-doped GaAs substrates, we calculate that Δn is in the range of $4.0\text{–}6.0 \times 10^{-4}$, in close agreement with the value calculated above. The carrier lifetime observed for LT-GaAs grown at temperatures between 260 and 400 °C is much lower than that of conventional, high-quality, MBE-grown 580 °C GaAs, for which we measure a carrier lifetime of ~ 1 ns. This decreased carrier lifetime is due to the larger defect densities incorporated at the lower growth temperatures. For the samples grown at 200 and 190 °C, the initial decay time is measured to be less than 0.4 ps. For such short times, intraband carrier dynamics also affect the measured signal, and thus this measurement gives only an approximate value of the carrier lifetime in these samples. This dramatic reduction in the carrier lifetime is believed to result from the large deep-level density due to $\sim 1\%$ excess As incorporation, as mentioned previously, which is not observed in the samples grown at temperatures of 260 °C or above.[11]

Two additional features are observed in the reflectivity data of the unannealed 190 and 200 °C LT-GaAs layers.

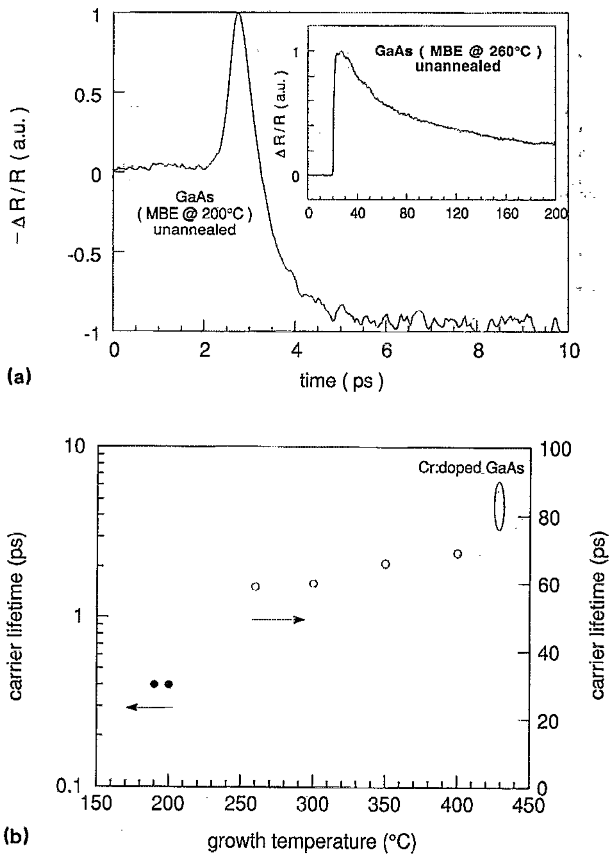

FIG. 1. (a) Normalized transient reflectivity of unannealed, 200 °C LT-GaAs layer. The peak $\Delta R/R$ amplitudes are between 4.0 and 6.0×10^{-4}. (The inset shows the same measurement for GaAs grown by MBE at a higher temperature of 260 °C.) (b) Carrier lifetime as a function of growth temperature for a number of unannealed LT-GaAs layers and Cr-doped semi-insulating substrates. Note the different time scales used in the figure.

First, the initial change in the reflectivity is negative for times less than 1 ps after the pump laser pulse is incident on the sample surface. (Note that for this sample $-\Delta R/R$ is plotted.) In contrast, all the LT-GaAs samples grown at higher temperatures show positive $\Delta R/R$. The second interesting feature is the crossing of the zero axis and the subsequent slow recovery ($\sim 10\text{–}15$ ps) of the signal for times greater than 1 ps after the pump beam is incident on the sample. The transient reflectivity signal observed for LT-GaAs grown at 260 °C or above show no significant difference between the as-grown and annealed samples. However, samples grown at 190 and 200 °C and annealed, both show a similar initial $\Delta R/R$ as in Fig. 1(a), but $\Delta R/R$ is positive and has no slow component, as shown in inset of Fig. 2.

These results lead us to conclude that the carrier lifetime in LT-GaAs is primarily determined by the growth temperature and not by the annealing, and that the As precipitates present in the annealed material are not necessary to achieve the fast recombination times. In general, crystalline GaAs has been observed to exhibit a positive $\Delta R/R$, although an initial $\Delta R/R$ that is negative has been observed for sputtered polycrystalline GaAs films.[13] The 200 and 190 °C LT-GaAs layers used in this work are

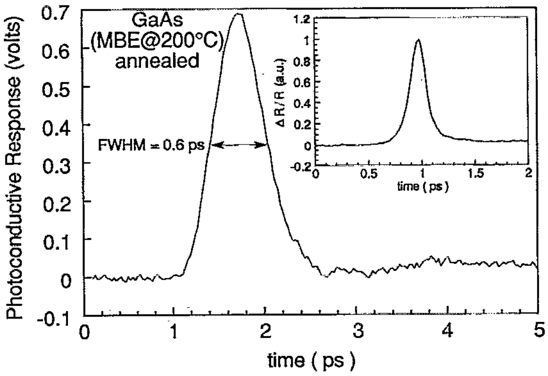

FIG. 2. Photoconductive switch response of annelaed, 200 °C LT-GaAs layer, as measured by external electro-optic sampling. A 10 V bias was applied across a 10-μm gap in the 50 Ω co-planar waveguide transmission line. (Inset) Transient reflectance measurement of the same epitaxial layer.

thought to be crystalline, as inferred from the *in situ* reflected high-energy diffraction pattern observed during growth. At present we do not have a clear understanding of why the initial $\Delta R/R$ is negative in the as-grown samples.

We believe that the slowly recovering component of the signal for times greater than 1 ps after the pump beam is incident on the sample is not indicative of the carrier lifetime, as inferred from the transient reflectance measurements on annealed samples and the photoconductive switching measurements to be discussed below. Similar behavior has also been observed for low temperature, MBE-grown $In_{0.52}Al_{0.48}As$ that is lattice-matched to InP.[16] It is possible that this slow transient in $\Delta R/R$ arises from reemission of carriers from near band-edge states into the conduction and/or valence bands and the subsequent fast recombination of the free carriers.

Photoconductive switch measurements were made using a 50 Ω co-planar waveguide transmission line on an annealed 200 °C LT-GaAs epitaxial layer. The electrodes were defined photolithographically using 500-Å/3500-Å Ti/Au metallization. A 10-μm gap was included in the center electrode of the transmission line. (For the unannealed LT-GaAs layers, the low dark resistivity resulted in poor photoconductive switching.) An electrical signal was launched by shorting the gap in the biased transmission line with pulses from the CPM laser, and this signal was measured using a standard external electro-optic sampling technique.[17] The results of this measurement are shown in Fig. 2. The pulse has a nearly symmetric shape and a full width at half maximum (FWHM) of 0.6 ps. The pulse width is limited by the electro-optic system response time of $\sim$0.3 ps.[17] From the $1/e$ decay time of the electrical pulse in Fig. 2, the carrier lifetime in the annealed 200 °C LT-GaAs material is estimated to be less than 0.4 ps.

The mobility μ of the photoexcited carriers in LT-GaAs can be estimated from an expression for the integrated photocurrent.[5] For the parameters of our experiment, we estimate $\mu = 120$–150 cm^2/V s. This calculated mobility is significantly lower than the Hall mobility cited above for 200 °C LT-GaAs. Large differences between mobilities derived from transient and steady-state measurements are common in materials with large density of traps,[18] and arise from the different trapping/detrapping and recombination kinetics of the picosecond photoconductive transient conditions and the Hall effect measurement conditions.

In conclusion we have demonstrated that MBE-grown GaAs exhibits a subpicosecond (<0.4 ps) carrier lifetime for growth temperatures near 200 °C. After annealing, this material has significantly better responsivity when used as a photoconductive switch. Therefore annealed LT-GaAs is an ideal photoconductive material for the generation of subpicosecond electrical pulses[1,2] and for subpicosecond sampling gates in guided-wave and terahertz-beam systems.[1,3]

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