Subpicosecond carrier dynamics in low-temperature grown GaAs on Si substrates

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This letter describes time-resolved differential reflection measurements on low-temperature grown GaAs on (100) Si substrates. The carrier recombination depends sensitively on growth and anneal conditions. The differential reflectance signals of samples annealed at 600 °C are dominated by an exponential subpicosecond transient, which can be as short as 370 fs. Optical microscopy and atomic force microscopy show that the films are comparably smooth or smoother than other GaAs material grown on Si. X-ray diffraction indicates tensile strain in the films, which is explained by the different thermal expansion coefficients of GaAs and Si. © *1999 American Institute of Physics.* [S0003-6951(99)01643-5]

Low-temperature grown GaAs (LTG-GaAs) is well known for its subpicosecond photocarrier lifetime.[1] This property together with its high resistivity and high electron mobility makes it one of the best photoconductive materials with subpicosecond response times available today. Photodetectors operating at frequencies up to 560 GHz[2,3] and sources of THz radiation such as photomixers[4] have been based on LTG-GaAs. We have investigated the growth of LTG-GaAs by molecular beam epitaxy (MBE) directly on silicon (Si) substrates. The use of silicon substrates is important because Si has three times the thermal conductivity of GaAs. This is needed for high power applications such as photomixer devices.[5] Other motivations are the prospect of integrating LTG-GaAs with Si-based electronics and larger substrate sizes.

About a decade ago the growth of GaAs on Si was investigated in great detail.[6] The main problems associated with heteroepitaxy of GaAs on Si are the different crystal structures, the lattice mismatch of 4% and the different thermal expansion coefficients. The lack of inversion symmetry in the GaAs crystal structure can lead to the formation of antiphase domains (APDs). The formation of APDs can be suppressed for example by growing on off-axis (100) Si substrates. Due to the lower growth temperature the strain resulting from thermal mismatch will be less problematic for LTG-GaAs on Si compared to regular GaAs on Si. A few groups have reported growth of LTG-GaAs on Si.[7–9]

Several researchers used pump–probe techniques to investigate the carrier dynamics in LTG-GaAs grown on GaAs.[1,10,11] Annealed LTG-GaAs material shows subpicosecond photocarrier lifetimes for anneal temperatures up to approximately 600 °C.[1,10] If the anneal temperature is 700 °C or higher the lifetime exceeds 1 ps.[11] There is one report on the carrier dynamics of LTG-GaAs grown on Si.[7] Our study extends these results to different wavelengths close to the band edge of the LTG-GaAs film and investigates the effect of growth and anneal conditions in more detail.

For this study thin films of LTG-GaAs have been grown by MBE on off-axis (100) silicon substrates, miscut by 4° towards the [011] direction. The structural quality of the films has been characterized *in situ* by reflection high energy electron diffraction (RHEED) and *ex situ* by optical microscopy, atomic force microscopy (AFM) and double crystal x-ray diffraction (DCXD).

The procedure for synthesis of LTG-GaAs on Si consists of the following steps: After a cleaning procedure,[12] the substrates are transferred into the MBE system where the native oxide is desorbed by increasing the substrate temperature to 800 °C and exposing the Si substrate to the Ga beam and the Si beam.[12,13] We nucleate the GaAs growth at a substrate temperature of 400 °C and grow 100 nm. This layer is then annealed for 10 min at 600 °C followed by growth of 25 nm of GaAs at 600 °C. At this point, the RHEED pattern is streaky but usually shows no evidence of surface reconstruction. This procedure is known as the two-step nucleation method. This initial layer serves as a template for the LTG-GaAs layer. The growth is interrupted while the substrate temperature is lowered to temperatures between 220 and 280 °C as measured with a thermocouple touching the back of the substrate. 900 nm of LTG-GaAs are deposited at this low temperature. The RHEED pattern during the LTG-GaAs growth is spotty indicating a rough, crystalline growth front.

The films appear specular and featureless to the naked eye as well as under an optical microscope. Phase contrast Nomarski microscopy does not reveal any roughness. The rms roughness as determined by AFM in a 2 μm by 2 μm scan is typically around 2.5 nm with a peak to peak variation

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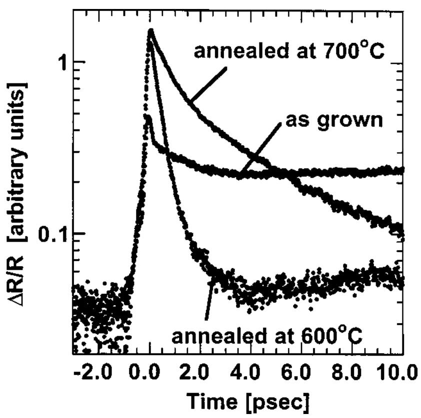

FIG. 1. Time-resolved differential reflection signals from samples grown at 240 °C. The samples are as-grown and annealed at 600 and 700 °C.

below 30 nm. This is about an order of magnitude higher than the roughness we find on LTG-GaAs grown on GaAs substrates; but it is among the best roughness values reported for GaAs grown on Si.[14]

DCXD shows that the lattice constant of the GaAs film normal to the interface is 0.2% shorter than the GaAs bulk value. The full width half maximum (FWHM) of the peak from the GaAs film is approximately 1000 arcsec. We believe that the tensile strain is due to the different thermal expansion of GaAs and Si. The GaAs film contracts more than the Si substrate leading to the tensile strain. In contrast LTG-GaAs films on GaAs substrates are strained compressively due to the excess As in the films.

The response time of the material was measured by time-resolved differential reflection. Our experimental setup is a pump–probe arrangement using short laser pulses generated by a mode-locked Ti:sapphire laser. The laser pulses are centered at a wavelength of 858 nm. Their repetition rate is 105 MHz and their duration is 135 fs. Pump and probe pulses are polarized orthogonal to each other. The pump beam is modulated with an acousto-optical modulator (AOM) at 9.0 MHz. Additionally the pump and the probe beam are modulated with a mechanical chopper at 2.5 and 1.8 kHz, respectively. The signal is measured using lock-in detection. The average powers are typically 10 mW for the pump beam and the 0.35 mW for the probe beam. The $\Delta R/R$ signals are independent of probe power for the power levels used. The spot size on the sample is about 50 μm.

We estimate that each pump pulse generates approximately $2\times10^{17}\,\mathrm{cm}^{-3}$ carriers. The wavelength of 858 nm is chosen because it is about 16 meV larger than the band edge of GaAs at room temperature. Therefore carrier cooling within the bands by optical phonon emission should not affect the measured signals. The measured changes in reflection are on the order of 10^{-4}.

Different samples from the same wafer underwent rapid thermal annealing in a forming gas (90% N_2, 10% H_2) ambient for 30 s at anneal temperatures of 400, 500, 525, 550, 575, 600 and 700 °C. The samples are capped with a GaAs wafer during the anneal to avoid outgasing of As. Samples from four wafers grown at 220, 240, 260 and 280 °C were measured.

Figure 1 shows signals measured on the wafer grown at 240 °C. The three traces are from an as-grown sample and

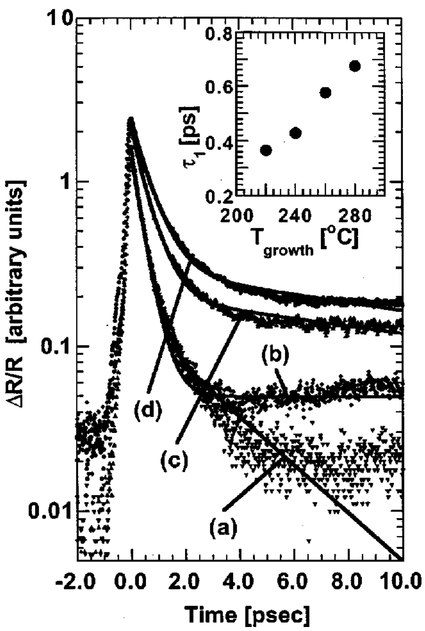

FIG. 2. Time-resolved differential reflection signals from samples grown at different temperatures of: (a) 220 °C, (b) 240 °C, (c) 260 °C and (d) 280 °C. All samples are annealed at 600 °C. Each set of data is fit with the sum of two exponentials. The inset shows the dependence of the initial transient decay time τ_1 on growth temperature.

samples annealed at 600 and 700 °C. These traces illustrate the trends we find on the samples from all four wafers. As-grown samples and samples annealed at lower temperatures (approximately up to 525 °C) show differential reflection signals $\Delta R/R$ that consist of a subpicosecond transient response on top of a term that is essentially constant on the 10 ps time scale investigated. Both signal components are of comparable magnitude. The exact dependence of the two components on anneal and growth temperature appears to be complicated. Some $\Delta R/R$ signals also contain additional terms and the constant term depends on pump power in a nonlinear fashion. Here we will not discuss samples annealed at these low temperatures in more detail. If the anneal temperatures are increased further up to 600 °C, the magnitude of the constant term decreased and the magnitude of the transient response increases. The subpicosecond transient dominates the $\Delta R/R$ signals of the samples annealed at 600 °C. These samples are discussed in more detail in the next paragraph. The $\Delta R/R$ traces of samples annealed at 700 °C have decay times in excess of 1 ps. Sign reversal of the $\Delta R/R$ signals was not observed in this experiment.

We now focus on the samples annealed at 600 °C because they may prove useful for subpicosecond photoconductive applications. Figure 2 shows the $\Delta R/R$ signals from samples grown at different temperatures and annealed at 600 °C. A least square fit with the sum of two exponentials is also shown for each set of data. Table I summarizes the fit

TABLE I. Fit parameters for fits shown in Fig. 2.

Sample	T_{growth} (°C)	τ_1 (ps)	τ_2 (ps)
a	220	0.37	3.0
b	240	0.43	∞
c	260	0.58	19
d	280	0.68	18

parameters. The first exponential term accounts for at least 86% of the signals initial amplitude in all cases. The inset in Fig. 2 shows the time constants τ_1 of the first exponential term as a function of growth temperature. The time constant τ_1 monotonically increases from 370 to 680 fs with growth temperature. The best fit for the sample grown at 240 °C is achieved when τ_2 approaches large values compared to the time scale of the measurement. For that reason we let τ_2 approach infinity. The time constant τ_2 does not show a clear trend with growth temperature.

Judging from the differential reflection measurements, anneal temperatures around 600 °C seem best suited to achieve subpicosecond carrier lifetimes with lower growth temperatures leading to shorter lifetimes. This is similar to LTG-GaAs grown on GaAs substrates[1,10,11] despite the presumably high dislocation density in material grown on Si and the reversed sign of the strain in the LTG-GaAs film. We observe a similar trend with growth temperature as Frankel *et al.*[7] We find however in contrast to their results a strong dependence on anneal conditions. The different results are most likely due to the different wavelengths used in the two experiments. Frankel *et al.* used a wavelength of 620 nm (2 eV) which is well above the band gap of LTG-GaAs and their results will therefore include effects of carrier cooling within a band. Our experiment is performed at a wavelength of 858 nm, only a few meV above the band gap, and will mainly be sensitive to carrier recombination or trapping. The long tails seen in our samples annealed at 600 °C indicate that a fraction of the photogenerated carriers relaxes on a much longer time scale. This is not desirable for devices requiring a short lifetime. Fine tuning of anneal and growth conditions should lead to improved results because the relaxation appears to be strongly dependent on these conditions.

In conclusion we have grown LTG-GaAs films on Si substrates. The films show very good surface morphology, they appear specular and featureless under an optical microscope, and AFM reveals 2.5 nm rms roughness. X-ray diffraction indicates tensile strain in the films which is explained by the different thermal expansion coefficients of GaAs and Si. Pump–probe experiments demonstrate that carrier recombination is very sensitive to anneal and growth conditions. When annealed at 600 °C, a subpicosecond transient dominates the time-resolved differential reflection signals. However, the signals also contain a smaller slow component. LTG-GaAs grown on Si is promising for high speed, high power devices where a higher thermal conductivity is needed and for integration of a high speed photoconductor with Si based circuitry.

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