High-carrier-density electron dynamics in low-temperature-grown GaAs

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(Received 2 January 1997; accepted for publication 12 April 1997)

Pump-probe differential transmission measurements examine high-carrier-density phenomena in as-grown and annealed GaAs samples grown at temperatures from 210 to 270 °C. We observe trap saturation and Auger recombination, and accurately model the measurements on annealed samples with a simple two level rate equation, allowing us to extract the trapped-electron lifetimes. © *1997 American Institute of Physics.* [S0003-6951(97)00524-X]

Low-temperature-grown GaAs (LT-GaAs) exhibits an ultrafast optoelectronic response that has been extensively studied and characterized for various material growth and annealing conditions.[1–3] However, much less is known about the trapping and recombination processes, particularly the trapped carrier lifetimes.[4–7] The characteristics of these processes are important technologically since recombination bottlenecks from trap states may limit the repetition rate at which devices made from LT-GaAs can operate.

In this investigation, we employ high-fluence pump pulses that create carrier densities sufficient to saturate the trapping states in LT-GaAs. For the annealed samples, a simple two-level rate equation accurately models the saturation dynamics and allows us to extract the free-electron and trapped-electron lifetimes. At extremely high carrier densities in some annealed samples, we observe that the initial free-electron lifetime decreases with increasing carrier density. This effect can be modeled by including an Auger recombination term in the rate equations. The Auger coefficient for these samples is many orders of magnitude larger than in standard GaAs. The results for the as-grown samples can not be adequately modeled with two-level rate equations, although from our measurements we may assess whether or not various physical processes are possibly responsible for the trapping dynamics.

The LT-GaAs samples studied are 1.5-μm-thick layers grown by molecular beam epitaxy at Lincoln Laboratory at temperatures of 210, 220, 230, 250, and 270 °C. *In situ* annealing was performed at 600 °C for 10 min on part of each wafer. All materials were found to be crystalline by electron diffraction. The layers were lifted off their GaAs substrates by etching and mounted on fused silica substrates in order to perform transmission measurements.

The laser source for the measurements is a 250 kHz Ti:sapphire regenerative amplifier producing 4 μJ, 80 fs pulses at 800 nm.[8] The probe pulses are obtained by splitting off ten percent of the output beam, focusing it into a 3 mm sapphire crystal to generate a single-filament white-light continuum, and filtering the continuum with a 10 nm bandwidth, 860 nm interference filter. The rest of the output beam is intensity modulated at 2 kHz by a mechanical chopper and used as the pump pulse source. The pump and probe beams are attenuated and focused on the sample with spot sizes of 100 and 80 μm, respectively. Pump-pulse energies range from 0.4 to 80 nJ and the probe-pulse energy is typically 40 pJ.

For each sample, we measure the differential transmission signal as a function of delay between pump and probe pulses for a range of pump fluences, typically from 5×10^{-6} to 1×10^{-3} J/cm^2. The dominant mechanisms through which the pump beam changes the transmission of the probe beam are band to band absorption bleaching and induced absorption from defect levels. Of these two signal components, the bleaching signal is predominant at high carrier densities, especially for the annealed materials. The 860 nm probe wavelength is chosen for two reasons: (1) The electron contribution to the absorption bleaching signal is a factor of twenty greater than the hole contribution[9] allowing us to isolate and examine the electronic response of the material. (2) Because we are probing states at the bottom of the conduction band, carrier cooling effects are evident in the rise time of the signal and not in its decay. This simplifies the modeling and analysis of the signal decay by allowing us to ignore a potentially variable parameter.

A set of normalized measurements for the 220 °C material, characteristic of those obtained for the annealed samples, is shown in Fig. 1. At low pump fluences, the differential signal decays exponentially, as expected, and the decay rate is independent of the pump fluence. As the pump

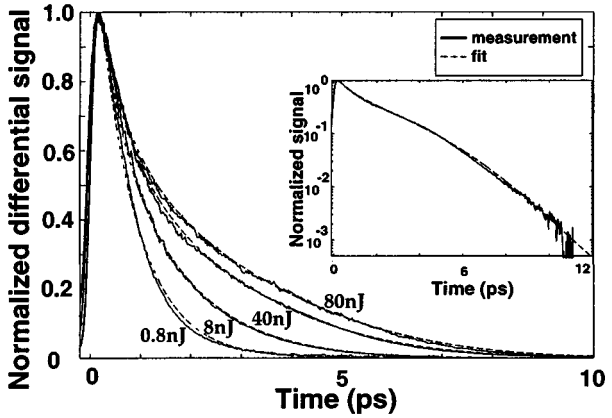

FIG. 1. The differential signal and the fit curves for the 220 °C-grown GaAs sample with pump-pulse energies of 0.8, 8, 40, and 80 nJ. (Fluences of 10^{-5}, 10^{-4}, 5×10^{-3}, and 10^{-3} J/cm^2.) Inset: 40 nJ curve on a log scale.

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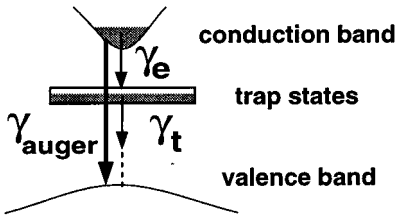

FIG. 2. A schematic depicting the processes modeled by the rate equations.

fluence is increased, the relaxation slows and becomes nonexponential as a result of the traps saturating or ''filling up''. For free-electron densities sufficiently larger than the trap density such that $N_{e\gamma_e} \gg N_{t\gamma_t}$, the decay actually becomes linear; dN_e/dt is independent of N_e and is determined solely by $N_{t\gamma_t}$. Here N_e represents the free-electron density, γ_e the unsaturated decay rate for an electron, N_t the trap density, and γ_t the rate an electron decays from the trapping state.

A simple rate equation model, pictured schematically in Fig. 2, accurately reproduces the measured signal for the annealed samples. The normalized rate equations are as follows:

$$\frac{df_e}{dt} = \frac{g(t)}{N_0} - \gamma_e f_e (1 - f_t) - \alpha f_e^3 N_0^2 \tag{1}$$

$$\frac{df_t}{dt} = \eta \gamma_e f_e (1 - f_t) - f_t \gamma_t . \tag{2}$$

In these equations $g(t)$ is the electron generation term (representing the pump pulse), N_0 is the initial carrier density $[\int_{-\infty}^{+\infty} g(t)dp]$, f_e is the fraction of injected electrons left in the conduction band ($N_e = f_e N_0$), f_t is the fraction of traps which are occupied, and η is the ratio of (N_0/N_t). The α coefficient is related to the Auger process, which we ignore now but will discuss later.

Three aspects of our modeling process should be mentioned. (1) Although the model fits the entire measured signal well, we are only interested in modeling the signal decay, so effects such as carrier cooling, which may be density dependent, do not have to be considered. (2) We can not predict from our measurements the fate of the trapped electrons—they may either recombine or decay to another trap state. (3) There is an induced absorption signal from defect levels present in the measurements evident as a negative signal with a decay time of several hundred picoseconds as shown Fig. 3. The ratio of the magnitude of the negative signal to the peak positive signal is 7% for low fluence measurements on the 210 °C material. This ratio decreases for higher fluence measurements and for samples grown at higher temperatures. Because the negative signal is small and practically constant on the relevant timescales ($1/\gamma_t \sim 10$ ps), we consider the negative signal level as the baseline to which the signal of interest decays. (Note that the extremely different timescales of the trapped-electron lifetime and the decay of the negative signal indicate that the induced absorption signal can not be from the same defects that are predominantly responsible for the electron trapping.)

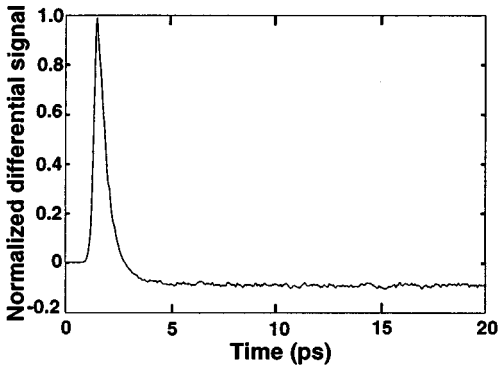

FIG. 3. A low-pump-fluence differential transmission signal from the 210 °C material showing the negative tail.

The model has three free parameters, neglecting the Auger term. For each sample, we determine γ_e by fitting the low-pump-fluence data with a single exponential. Then we select γ_t so that the only parameter which needs to be varied to fit all the curves for a given sample is η. This simple model fits the experimental measurements very well for all pump fluence levels over almost three orders of magnitude as can be seen in Fig. 1, leading us to believe it accurately depicts the basic physical processes occurring in this material. Table I shows the fitting parameters for the annealed materials. Surprisingly, the trapped electron lifetimes decrease with increasing growth temperature (decreasing defect concentration). This is contrary to the dependence expected if electrons and holes are recombining from different, spatially separated defects. The uncertainty in the values arise from the fact that we are fitting a series of curves for each sample and the modeled signal is relatively insensitive to the γ_t parameter, especially when γ_t is significantly larger than γ_e.

For the 210 and 270 °C annealed materials, the initial decay rate of the signal increases at very high carrier concentrations as shown in the inset of Fig. 4. Rate equations can not model this effect unless a carrier-density dependent decay term is included. Two physical processes potentially able to explain the observed signals are Auger-assisted trapping and Auger recombination. Incorporating Auger-assisted trapping into the model does not enable us to fit the data. The process turns on too slowly as a function of carrier density and it can not produce a significant change in the initial decay rate because the electron traps rapidly fill and quench the process. Adding an Auger recombination term ($\alpha f_e^3 N_0^2$) enables reasonable measurement fits as seen in Fig. 4. The Auger coefficients are estimated to be 10^{-25} and 10^{-27} cm^6/s for the 210 and 270 °C materials, respectively, both terms orders of magnitude larger than in standard GaAs. A possible explanation for the increased Auger rates is that the large concentration of defects in LT-GaAs reduces the

TABLE I. Fitting parameters.

Material	270 °C	250 °C	230 °C	220 °C	210 °C
$\tau_e = 1/\gamma_e$	8 ps	2.3 ps	1.5 ps	0.75 ps	0.61 ps
$\tau_t = 1/\gamma_t$	0.5–3 ps	2–4 ps	1.5–2.5 ps	5–9 ps	15–25 ps

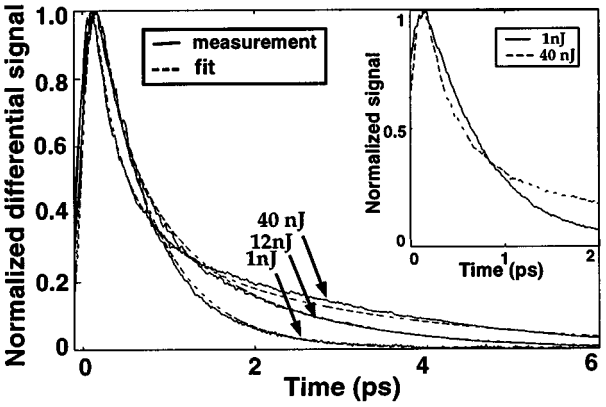

FIG. 4. The differential signal and the fit curves for the 210 °C-grown GaAs sample with pump-pulse energies of 1, 12, and 40 nJ. The inset shows more clearly the increased initial decay rate in the high-fluence measurement.

crystallinity of the material and relaxes the momentum conservation requirement for carrier scattering. This relaxation creates a much wider range of momentum states into which the excited electrons may scatter, drastically increasing the Auger rate.

The signals from the as-grown samples are more complex and are not possible to fit with a two-level rate equation. Qualitatively, the signals are similar to those of the annealed samples; at low pump fluences, the signal decays exponentially to a negative level and at higher pump fluences the signal decay slows down and develops a long positive tail. If a model similar to the one previously discussed can be loosely applied to as-grown LT-GaAs, it appears as if the trapped carrier lifetime also decreases with increasing growth temperature. Recently, Siegner *et al.*[6] proposed that this effect may be explained if the dominant recombination pathway is via trapped electrons recombining with free holes. We believe this is not likely to be a dominant process. In their analysis, Siegner *et al.* assume the negative signal, with a time constant of 100's of picoseconds, is indicative of induced absorption from the primary trapping states; not necessarily a valid assumption in view of our results for the annealed materials. However, even if we regard this as a valid assumption and use rate equations to model the proposed process, we find they can not fit the data. If the rate equation parameters are chosen to fit the signals for low and moderate pump fluences, the rate equations can not fit the much slower decay rates observed at high fluences. This is because at high carrier densities both the free-carrier decay rate and the trapped-carrier decay rate are proportional to the number of free carriers so the trapping bottleneck is not effective.

In conclusion, we have examined electron trapping dynamics under high-carrier-density conditions in a series of LT-GaAs samples. For the annealed materials, fitting the results with a two-level rate equation model allows us to measure the free- and trapped-electron lifetimes. The results for the as-grown material are qualitatively similar but require a more complex model for curve fitting.

Support for this research was provided by the Air Force Office of Scientific Research, USAF, under Grant No. DOD-G-F49620-95-1-0227. This work was also supported by the National Science Foundation through the Center for Ultrafast Optical Science under STC PHY 8920108. T. Sosnowski is an IMRA America scholar.

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