Subband gap carrier dynamics in low-temperature-grown GaAs

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Measurements of the carrier relaxation dynamics in low-temperature-grown GaAs have been made with a femtosecond-resolution, time-resolved pump-probe technique using a subband-gap probe-beam wavelength. The transient absorption and index of refraction changes have been analyzed using a relaxation model with up to four different excited state populations. The carrier recombination time within the midgap trap states is found to be longer than the subpicosecond free-carrier trapping time. Two time scales are observed for the recombination rate, one of a few picoseconds and one of hundreds of picoseconds, indicating the presence of at least two different trap states for the free-carriers in this material. © *1997 American Institute of Physics.*
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Since its discovery,[1] low-temperature grown GaAs (LT-GaAs) has been the subject of sustained interest. This is primarily because its semi-insulating properties[2] and its very short carrier lifetime[3] have proven to be very useful in device applications,[4] and also because the roles of trapping and recombination are still unresolved and somewhat controversial. Measurements have demonstrated subpicosecond free-carrier relaxation times, which are attributed to the high densities of As related defects (especially As antisites) and, when annealed, As precipitates. Most recently, several new experiments have addressed the issue of separately resolving the free-carrier trapping time and the carrier recombination time in this material.[5–8]

A common means for determining the carrier relaxation dynamics is the transient differential transmission/reflection pump-probe technique. Figure 1 presents a typical result from such a measurement on a LT-GaAs sample grown at 210 °C and annealed. A bleaching of the absorption is observed near the zero time delay due to free-carriers photogenerated by the pump beam (the bandfilling effect),[9] and this recovers within a single picosecond. However, upon closer observation, in Fig. 1 inset, it is obvious that the system does not return to its ground state before several hundreds of picoseconds. It is very important to understand the nature of this long-lived excited state, since a slow carrier recombination time could severely limit the maximum repetition rate of optoelectronic devices made with this material.

Here we report results of a femtosecond-resolution, time-resolved pump-probe experiment on LT-GaAs using subband-gap photon energy for the probe beam. This experiment has a sensitivity targeted at the dynamics of subband-gap carriers, leading to new information on the carrier trapping and recombination times in the defect levels within the band gap. The sample investigated is a 1.5-μm-thick, molecular-beam epitaxial LT-GaAs film grown at 210 °C and *in situ* postannealed at 600 °C for 10 min. The film was removed from its native substrate by chemical etching and bonded to a fused silica substrate to facilitate transmission measurements.

The experiments were performed using a mode-locked Ti-sapphire oscillator operating at $\lambda = 810$ nm with a pulse width full width at half-maximum (FWHM) of 110 fs and an optical parametric oscillator operating at $\lambda = 1.56$ μm with a pulse width of 100 fs. The probe beam, which had 1 mW average power, an *s* polarization, and an incident angle of 15°, was focused down to a spot size of 50 μm onto the sample. The pump beam, with 18 mW average power, a *p* polarization, and an incident angle of 34°, was focused to 100 μm. Standard lock-in detection was used to measure the transient reflectivity (ΔR) and transmissivity (ΔT). The method described in Ref. 10 has been used to extract the induced absorption ($\Delta\alpha$) and index of refraction (Δn) changes from the measured ΔT and ΔR, taking into account the Fabry–Perot effect.

Results for the LT-GaAs sample grown at 210 °C and annealed are presented in Fig. 2. For semi-insulating GaAs (not shown), a pump-probe measurement with $\lambda_{pu} = \lambda_{pr} = 1.56$ μm yields only an instantaneous peak of small amplitude due to two-photon absorption. However, this two-photon absorption is too small to generate a significant amount of free-carriers. Figure 2(a) shows a pulse with a wider peak than the pump-probe cross correlation. This indi-

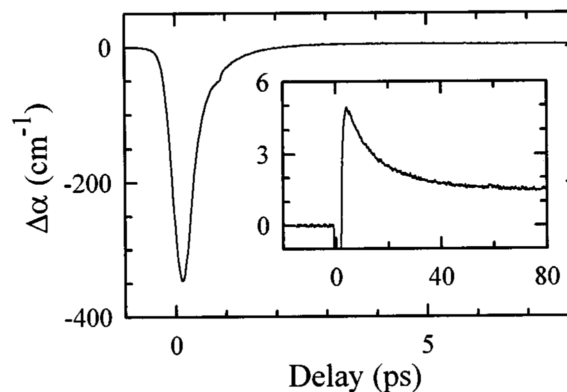

FIG. 1. Transient absorption change for an LT-GaAs sample grown at 210 °C and annealed, with $\lambda_{pu} = \lambda_{pr} = 810$ nm. Inset: the low-amplitude, long-duration relaxation tail observed with enhanced resolution.

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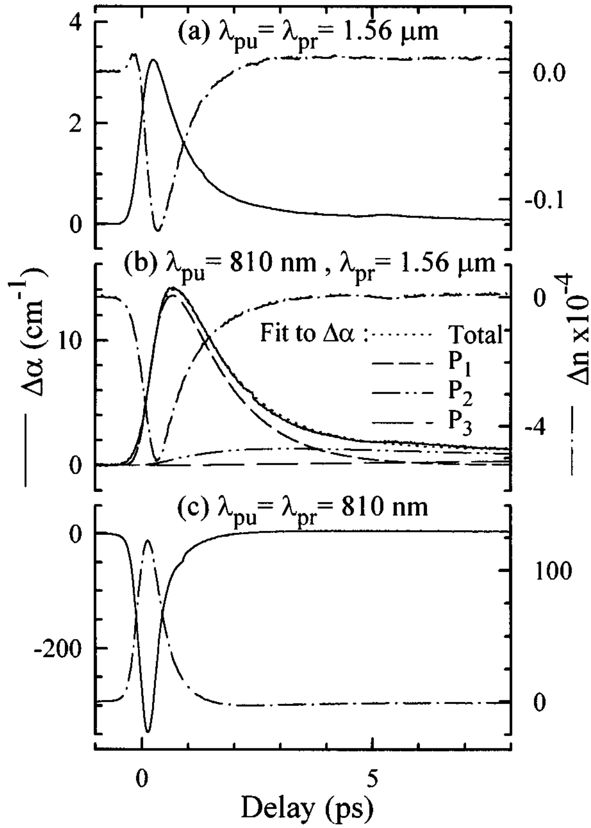

FIG. 2. Transient absorption and index of refraction change for an LT-GaAs sample grown at 210 °C and annealed. Also shown, in (b), calculated curves from the fitting procedure. P_1, P_2, and P_3 are the three individual excited-state population contributions to the signal (see the text).

cates that, for this sample, a significant number of carriers can be excited from the midgap defect states to the conduction band and/or from the valence band to the midgap defect states. Figure 2(b), when compared to Fig. 2(c), shows that a subband-gap probe-beam wavelength is more sensitive to the long lived excited states that are related to defects.

In Fig. 2(b), along with the experimental results, calculated curves for $\Delta\alpha$ are presented. These include contributions to the signal from three individual carrier populations, along with the total of these three. Fits were made using the following model

$$\left.\begin{array}{l}\Delta\alpha(t_d)\\ \text{or}\\ \Delta n(t_d)\end{array}\right\} = \frac{\alpha_0^{\text{pu}}/\hbar\omega_{\text{pu}}}{\int_{-\infty}^{\infty} dt I_{\text{pr}}(t)} \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt_1 R(t-t_1) \times I_{\text{pu}}(t_1) I_{\text{pr}}(t-t_d), \tag{1}$$

where t_d is the time delay, $I_{\text{pu}}(t)$ and $I_{\text{pr}}(t)$ are the pump and probe pulse profile, respectively, α_0^{pu} is the ground state absorption coefficient at the pump wavelength, and $\hbar\omega_{\text{pu}}$ is the pump beam photon energy. $R(t)$ is the material response function, given by

$$R(t) = a_{\text{ins}}\delta(t) + a_1 P_1(t) + a_2 P_2(t) + \dots, \tag{2}$$

where a_i are amplitude factors and $\delta(t)$ is the Dirac delta function, accounting for an instantaneous contribution to the response. $P_i(t)$ are the impulse response functions obtained by solving the coupled relaxation equations

$$\begin{aligned} \frac{\partial P_1(t)}{\partial t} &= -\frac{P_1(t)}{\tau_1} + \delta(t), \\ \frac{\partial P_2(t)}{\partial t} &= -\frac{P_2(t)}{\tau_2} + \frac{P_1(t)}{\tau_1}, \end{aligned} \tag{3}$$

and so on.

Then, $P_i(t)$ represents successive excited-state populations, decaying with the relaxation time τ_i. Here we did not attempt to fit the data with both the electron and hole densities, since that requires a precise knowledge of the $\Delta\alpha$ and Δn caused by each carrier type at each of their different excited states. Then, in terms of carrier densities, each $P_i(t)$ is composed of both electron and hole densities at a given stage of their relaxation, when they have similar relaxation times. Nevertheless, this phenomenological model allows one to extract *every* component contributing to $\Delta\alpha$ and Δn that has a different relaxation time. Also, from the results, it does appear that we often can, from qualitative arguments, ascribe a given $P_i(t)$ to a particular carrier type (i.e., as being dominated by a particular carrier type).

The values obtained from this fitting procedure are presented in Table I. Values preceded by the symbol "≈" indicate a more uncertain value due to the relatively small contribution of this excited state population to the signal and/or due to a relaxation time much longer than the scanned

TABLE I. Parameters obtained from the fitting procedure of the results for an LT-GaAs sample grown at 210 °C and annealed. Values of 178 and 12 000 cm^{-1} were assumed for the ground state absorption of the pump beam (α_0^{pu}) at $\lambda_{\text{pu}} = 1.56$ μm and 810 nm, respectively. The a_i associated to $\Delta\alpha$ are in $\times 10^{-17}$ cm^2, those for Δn are in $\times 10^{-21}$ cm^3. The relaxation times (τ_i) are in picoseconds.

			P_1		P_2		P_3		P_4	
		a_{ins}	a_1	τ_1	a_2	τ_2	a_3	τ_3	a_4	τ_4
$\lambda_{\text{pu}} = \lambda_{\text{pr}} =$	$\Delta\alpha$				74	0.73	4.31	7.3		
1.56 μm	Δn	0.51			−3.55	0.72	0.24	10	0.07	>300
$\lambda_{\text{pu}} = 810$ nm	$\Delta\alpha$		0.01	0.37	10.8	1.2	0.78	9.5	0.30	≈138
$\lambda_{\text{pr}} = 1.56$ μm	Δn		−1.49	0.13	−3.28	0.75			0.03	≈440
$\lambda_{\text{pu}} = \lambda_{\text{pr}} =$	$\Delta\alpha$		−441	0.14	−66.3	0.58	2.75	≈10	0.82	>300
810 nm	Δn		106	0.39	−4.0	1.5	−0.43	≈30	0.07	>300

time window. The symbol ''>'' indicates that, while the data has a long relaxation time signal, no precise value for this relaxation time can be obtained from the fitting procedure.

Certain significant trends are noted in the data of Table I. For example, the relaxation times of $P_1(t)$ extracted from the $\Delta\alpha$ and Δn data at $\lambda_{pu}=810$ nm and $\lambda_{pr}=1.56$ μm, (that is, 0.37 and 0.13 ps, respectively) are remarkably close to those found from Δn and $\Delta\alpha$, respectively, for $\lambda_{pu}=\lambda_{pr}=810$ nm. This suggests that the two different values of τ_1 for $P_1(t)$ are representative of the relaxation contributions of the two different carriers. We know for $\lambda_{pu}=\lambda_{pr}=810$ nm that the $\Delta\alpha$ response arises from the bandfilling effect[10] and, since the conduction band density of states is much smaller than that of the valence band, that the signal is mainly due to free electrons. For this pump-beam wavelength, the carriers are also photoexcited with an excess kinetic energy of ≈ 100 meV. Thus, the relaxation time of 0.13 ps can be directly associated with the cooling of electrons, and $\tau_1=0.37$ ps can be interpreted as the hole cooling time.

A corresponding similarity between relaxation times for $P_2(t)$ extracted using the Δn and $\Delta\alpha$ for different sets of pump-probe wavelengths is also observed. In this case, we note that the amplitude (a_2) associated with Δn for both $\lambda_{pu}=\lambda_{pr}=1.56$ μm and $\lambda_{pu}=810$ nm, $\lambda_{pr}=1.56$ μm remains essentially constant, meaning Δn scales with the density of generated carriers. This contribution (a_2) to Δn can then be assumed to be due to free-carriers,[10] and since the electron effective mass is about 7 times smaller than that of a hole, Δn should mainly be due to the free electrons. Therefore, these relaxation times, $\tau_2=0.7$ and 1.2 ps can be associated with the electron and hole trapping times, respectively. This assignment is also confirmed by the fact that $\Delta\alpha$, when $\lambda_{pu}=\lambda_{pr}=810$ nm, is still dominated by the free-electrons, even after the carriers have cooled.[11] As for the case of carrier cooling, results extracted using $\Delta\alpha$ at one set of pump and probe wavelengths have been correlated with information gathered from Δn at a different set of wavelengths.

The next populations [$P_3(t)$ and $P_4(t)$] exist within midgap defects, and the associated time constants are related to trapped carrier lifetimes. A relaxation time of around 10 ps has been observed in saturation experiments[7] and attributed to the trapped electron and hole recombination. This time agrees with the value obtained here for τ_3. The presence of $P_4(t)$ indicates that there is, at the least, one other kind of trapping center with a much longer recombination time present in this LT-GaAs sample.

The trapping and recombination mechanism and the nature of the defects responsible for it are still controversial. Arsenic-precipitate defects are believed to lead to recombination as soon as electrons and holes are trapped at the defect.[6] Point defects, and especially As antisites, are believed to yield a fast trapping time but a slower recombination time.[8] The latter seem to apply to the behavior observed here. The $P_3(t)$ population could result from electrons trapped at As antisites, while $P_4(t)$ could be due to a shallow trap (As complex), and weak coupling of the defect to the GaAs matrix would explain the long τ_4 relaxation time.

Whether such a long recombination time would severely limit the repetition rate of an optoelectronic device would depend on how many carriers each recombination channel (i.e., each set of trapping states) can handle and on how much the saturation of one channel by carriers would affect the characteristics of the material. For the former, saturation experiments[7] have indicated that the first recombination channel can handle a high number of carriers while the second saturates easily. Therefore, due to the low density of carriers trapped in the long-lived trapping states, only small perturbations to the material characteristic are expected on this >100 ps time scale. The possibility of the existence of a high density of trap states having a lifetime of ~ 10 ps still raises uncertainty regarding ultrahigh-repetition-rate pulse detection.

To summarize, the results of the analysis of subband-gap pump-probe data using a model that includes different excited-state populations show four different time scales for the carrier relaxation dynamics. The first is related to the hot carrier relaxation within the band. The second, having $\tau \leqslant 1$ ps, is associated with the free-carriers being trapped into midgap states. The last two, around ten ps and several hundreds of ps, are attributed to carrier recombination within the midband-gap trapping states. This indicates the presence of at least two sets of trapping states for the electrons and the holes.

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