

Photoacoustic study of the effect of doping concentration on the transport properties of GaAs epitaxial layers

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Abstract. We report a photoacoustic (PA) study of the thermal and transport properties of a GaAs epitaxial layer doped with Si at varying doping concentration, grown on GaAs substrate by molecular beam epitaxy. The data are analyzed on the basis of Rosencwaig and Gersho's theory of the PA effect. The amplitude of the PA signal gives information about various heat generation mechanisms in semiconductors. The experimental data obtained from the measurement of the PA signal as a function of modulation frequency in a heat transmission configuration were fitted with the phase of PA signal obtained from the theoretical model evaluated by considering four parameters—viz., thermal diffusivity, diffusion coefficient, nonradiative recombination time, and surface recombination velocity—as adjustable parameters. It is seen from the analysis that the photoacoustic technique is sensitive to the changes in the surface states depend on the doping concentration. The study demonstrates the effectiveness of the photoacoustic technique as a noninvasive and nondestructive method to measure and evaluate the thermal and transport properties of epitaxial layers. © 2003 Society of Photo-Optical Instrumentation Engineers. [DOI: 10.1117/1.1564101]

Subject terms: photoacoustics; semiconductors; thermal and transport properties.

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1 Introduction

In recent years, the laser-induced photoacoustic (PA) technique has been effectively employed to characterize semiconductor materials because of its versatility as a nondestructive and noninvasive method for the evaluation of material parameters.^{1–10} All the photothermal methods are based on the detection, by one means or other, of thermal waves generated in the sample after excitation with modulated optical radiation. In the simple and elegant PA technique these thermal waves produce density fluctuations in the specimen and the surrounding medium, which can be detected either by a sensitive microphone or by a piezoelectric transducer.

In the past, much work has been done in the characterization of both direct- and indirect-bandgap semiconductors using the PA technique.^{5–7} A detailed discussion of the contribution of various factors to the thermal flux in semiconductors under periodic optical excitation is given by Pinto Neto et al.⁶ Dramicanin et al.⁷ gave an analytical solution for various factors contributing to heat generation in semiconductors that has resulted in a major renaissance in the application of the PA effect to the characterization of transport properties of semiconductors. However, not much work has been done to study the influence of doping concentration on the thermal and transport properties of epitaxially grown semiconductor layers. Some of the recent investigations show that doping can definitely influence the

thermal diffusivity and surface recombination velocity of compound semiconductors.^{9–12}

In this paper, we present the results of our PA measurements on an epitaxial layer of GaAs doped with different concentration of Si, grown on a GaAs substrate by molecular beam epitaxy (MBE). Amplitude of the PA signal gives a clear picture of the various heat generation mechanisms in semiconductors. The phase of the PA signal is fitted with the theoretical model by taking the thermal diffusivity, diffusion coefficient, surface recombination velocity, and nonradiative recombination time as adjustable parameters to solve the heat diffusion equation.

2 Experimental Setup

A schematic representation of the open photoacoustic cell (OPC) used here is given in Fig. 1. Optical radiation from an argon ion laser at 488 nm (Liconix 5000) is used as the source of excitation, which is intensity-modulated using a mechanical chopper (Stanford Research Systems SR 540) before it reaches the sample surface. Detection of the PA signal in the cavity is made using a sensitive electret microphone (Knowles BT 1754). Details of the PA cell are explained elsewhere.¹³ The cell has flat response in the frequency range 40 to 4000 Hz. The phase of the photoacoustic signal is measured using a dual-phase digital lock-in amplifier (Stanford Research Systems SR 830), which is highly sensitive and can read a change of 0.01 deg in phase angle, which corresponds to a very small variation in

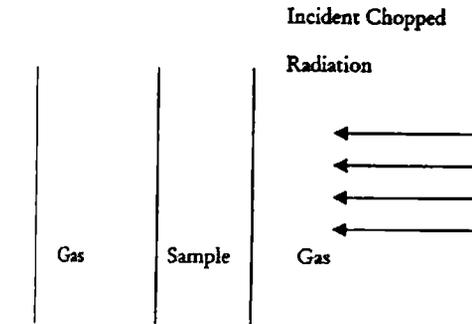


Fig. 1. PA cell geometry for the heat transmission configuration.

modulation frequency (~2 Hz), so that the data obtained have a high degree of accuracy. The laser power used for the present studies is 50 mW with a stability of ±0.5%. The samples are fixed on the OPC using vacuum grease at the top and the illumination by periodically modulated light is incident on the exposed portion of the sample.

The samples used for the present investigation are Si-doped GaAs epitaxial layers grown on GaAs substrates of thickness 400 μm by MBE. The epitaxial layers have thicknesses of 10.25, 3, and 2 μm, and the respective carrier concentrations are 2 × 10¹⁴, 2 × 10¹⁶, and 2 × 10¹⁸ cm⁻³.

Theoretical Background

The transport properties of Ge-doped GaAs epitaxial layers have already been treated using the monolayer approximation.¹⁰ The differences between the two-layer approximation and the monolayer method for photothermal measurements are apparent only at high frequencies,¹⁴ viz., in the hundreds of kilohertz. Hence our semiconductor results can be explained in terms of thermal piston model proposed by Zwang and Gersho,¹⁵ according to which, the pressure fluctuations in the PA cell due to periodic heating of the sample are given by

$$P_0 \Theta = \frac{P_0 \Theta}{T_0 \sigma_g} e^{j\omega t} \quad (1)$$

where P_0 (T_0) is the ambient pressure (temperature), l_g is the length of the gas chamber, $\sigma_g = (1 + j)a_g$, where $a_g = (\alpha_g / \mu_g)^{1/2} = 1/\mu_g$, with μ_g the thermal diffusion length of the gas with thermal diffusivity α_g , and Θ the sample temperature fluctuation at the sample-gas interface ($x = 0$). Also, $\omega = 2\pi f$, where f is the modulation frequency. In the remaining sections we are considering the PA cell geometry for the heat transmission configuration shown schematically in Fig. 1. The temperature fluctuation Θ can be obtained from the solution of the thermal diffusion equation given by

$$\frac{\partial}{\partial x} \left(\frac{\partial T}{\partial x} \right) = \frac{Q(x,t)}{k_s} \quad (2)$$

where k_s (k_g) is the sample thermal diffusivity (conductivity) and $Q(x,t)$ is the heat power density generated in the

sample due to absorption of the intensity-modulated laser radiation. In semiconductors, if the incident energy is greater than the bandgap of the semiconductor, then thermal power density $Q(x,t)$ mainly arises from three different processes.

1. The thermalization component arises from fast non-radiative intraband transitions in the conduction band of semiconductors. This occurs mainly due to the electron-phonon interaction, which happens typically on the time scale of picoseconds. Hence this process can be taken as instantaneous for the modulation frequencies usually used in the photoacoustic experiment. The heat power density due to this process is denoted by

$$Q_D = \frac{\beta(E - E_g)}{E} I_0 \exp[\beta(x + l_s)] e^{j\omega t} \quad (3)$$

where β is the optical absorption coefficient for photons having energy E , incident at $x = -l_s$ with an intensity I_0 (W/cm²).

2. The second component is due to the recombination of the photoexcited carriers in the bulk of the material after they travel a finite distance $(D\tau)^{1/2}$, where D is the carrier diffusion coefficient and τ is the recombination time. The heat power density due to nonradiative bulk recombination is given by

$$Q_{NRR} = \frac{E_g}{\tau} n(x,t) \quad (4)$$

where $n(x,t)$ is the density of the photoexcited carriers.

3. The nonradiative recombination of the photoexcited carriers at the surface of the material also contributes to the total heat power density, and it is given by

$$Q_{SR} = E_g [v_0 \delta(x) + v_s \delta(x + l_s)] n(x,t) \quad (5)$$

where v_0 is the carrier recombination velocity at the heating surface and v_s is the surface recombination velocity at the sample-gas interface at $x = 0$.

From the above analysis, it is obvious that the solution to Eq. (2) depends on the density of photoexcited carriers, which obeys the carrier diffusion equation, namely,

$$\frac{\partial n}{\partial t} = D \frac{\partial^2 n}{\partial x^2} - \frac{n}{\tau} + \frac{\beta I_0}{h\nu} \exp(x + l_s) e^{j\omega t} - v_0 n(x,t) \delta(x) - v_s n(-l_s,t) \delta(x + l_s) \quad (6)$$

For 488-nm radiation from an argon ion laser we can assume that all the incident radiation is absorbed at the $x = -l_s$ surface, so that we can replace $\beta I_0 \exp[\beta(x + l_s)]$ in Eqs. (3) and (6) by $I_0 \delta(x + l_s)$. Since the thermal conductivity of the surrounding air is very small, we neglect the diffusion of heat into it. Then the solution of the coupled equations (2) and (6) leads to the expression for pressure fluctuations for the thermally thick sample as

$$\delta P = \frac{2\varepsilon l_0 P_0}{T_0 l_g \sigma_g k_s \sigma_s} \left[\frac{\varepsilon - 1}{\varepsilon} \exp(-l_s \sigma_s) + \frac{F \sigma_s}{D \gamma \tau} \left(\frac{1}{\sigma_s^2 - \gamma^2} + \frac{v \tau}{\sigma_s} \right) \right], \quad (7)$$

where $\sigma_s = (1 + j) a_s$, $a_s = (\pi f / \alpha_s)^{1/2} = (1 / \mu_s)$ with μ_s the thermal diffusion length of the sample, $\gamma = [(1 + j \omega \tau) / D \tau]^{1/2}$ is the carrier diffusion coefficient, $\varepsilon = E_g / h \nu$, $r = v / D \gamma$, $r_0 = v_0 / D \gamma$, and

$$F = \frac{1}{(1 + r_0)(1 + r) e^{\gamma l} - (1 - r)(1 - r_0) e^{-\gamma l}}. \quad (8)$$

In the experimental frequency range used here, $\omega \tau \ll 1$, so that F , r , r_0 become real constants independent of the modulation frequency. It is reported by Pinto Neto et al.⁶ that the OPC signal for a semiconductor sample in the thermally thick region is essentially determined by nonradiative recombination. Thus the expression for the pressure fluctuation in the experimental frequency range for which $\omega \tau \ll 1$ is given by

$$\delta P = \frac{2\varepsilon f_0 P_0 F}{T_0 l_g k_s D \gamma \tau \sigma_g} \left(\frac{1}{\sigma_s^2 - \gamma^2} + \frac{v \tau}{\sigma_s} \right), \quad (9)$$

and the phase of the OPC signal is given by

$$\Phi = \frac{\pi}{2} + \Delta \Phi, \quad (10)$$

where

$$\tan \Delta \Phi = \frac{(aD/v)(\omega \tau_{eff} + 1)}{(aD/v)(1 - \omega \tau_{eff}) - 1 - (\omega \tau_{eff})^2} \quad (11)$$

with $\tau_{eff} = \tau[(D/\alpha_s) - 1]$.

We took the thermal diffusivity, diffusion coefficient, surface recombination velocity, and relaxation time as adjustable parameters, and then we fitted the variable part of Eq. (10) to the experimentally obtained phase angle $\Delta \Phi$.

4 Results and Discussion

Log-log plots of the amplitude of the PA signal against chopping frequency for the samples under investigation are given in Fig. 2. The three different heat generation mechanisms are evident from the figure. In the low chopping-frequency range, thermalization is the dominating process in heat generation, followed by bulk and surface recombination of photoexcited carriers, respectively. Figure 3 shows the best theoretical fit to the experimentally obtained phase of the photoacoustic signal. The values obtained as the best fitting parameters for the theoretical model is given in Table 1. The fitting program follows essentially the least-squares method developed using MATLAB. The fitting analysis resulted in the following accuracy of the fitted parameters: thermal diffusivity $\pm 2\%$, diffusion coefficient $\pm 5\%$, nonradiative recombination time $\pm 3\%$, and surface recombination velocity $\pm 8\%$. It is seen from the figure that there is a minimum in the phase plot of all the specimens

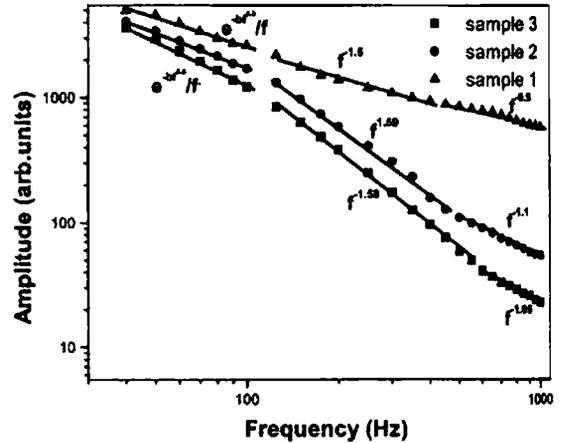


Fig. 2 Log-log plot of PA amplitude against chopping frequency.

under investigation. Many authors have attributed this change in shape as due to the change in heat generation mechanism in semiconductors from bulk nonradiative recombination to surface recombination of photoexcited carriers.⁶ From the present studies, it is seen that the frequency at which the phase data show a minimum changes with the concentration of dopant. This may be due to the increase in recombination centers with increase in doping concentration, which in turn enhances heat generation due to bulk recombination.

It is seen from Table 1 that the thermal diffusivity of the specimen under consideration is less than that of the earlier reported bulk GaAs sample.⁶ Thermal diffusivity is an important thermophysical parameter, which determines the distribution of temperature in systems where heat flow occurs. It was reported earlier that the thermal diffusivity in semiconductor films can deviate from the corresponding values in the bulk material.¹² It is seen clearly from Table 1 that the thermal diffusivity of the epitaxial layer decreases with increase in doping concentration. In semiconductors heat is transported by both phonons and charge carriers.

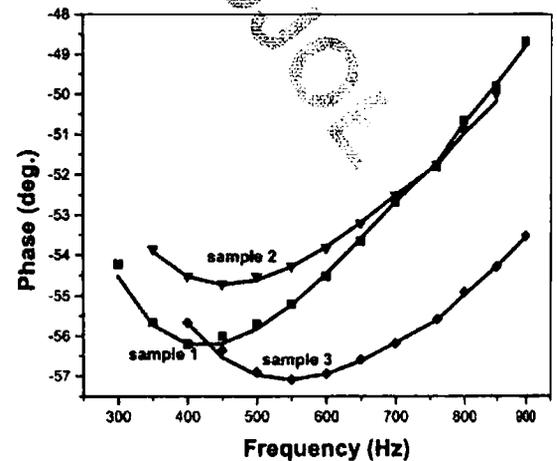


Fig. 3 OPC phase angle versus modulation frequency for the samples under investigation. The solid lines represents the fits of Eq. (10) to the data.

Table 1. Thermal and transport properties of GaAs epitaxial layers versus doping concentrations of Si.

Sample number	1	2	3
Thickness of epitaxial layer (μm)	10.25	3	2
Thickness of substrate (μm)	400	400	400
Concentration of silicon (cm^{-3})	2×10^{14}	2×10^{16}	2×10^{18}
Thermal diffusivity ($\text{cm}^2 \text{s}^{-1}$)	0.26	0.23	0.21
Surface recombination coefficient ($\text{cm}^2 \text{s}^{-1}$)	5.2	4.9	4.5
Surface recombination velocity (cm s^{-1})	415	476	525
Effective recombination time (μs)	11.2	9.8	7

The contribution to thermal conductivity from phonons is greater than that from carriers, especially for carrier concentrations less than 10^{20} cm^{-3} . The decrease in thermal diffusivity can be explained in terms of the dominant phonon contribution. Phonon scattering is a key source of carrier recombination processes and limits the performance of electronic and optoelectronic devices. Addition of the dopants, which can be considered as point defects, enhances the scattering of phonons, which results in a reduction of the phonon mean free path and consequently a decrease in thermal conductivity. It is shown in Ref. 16 that the thermal conductivity k is governed by the lattice thermal resistivity W through the relation $k = 1/W = AT^{-n}$, where $n = 1.25$ for GaAs, and A is a parameter that increases with increase in doping concentration. Since the thermal diffusivity and thermal conductivity are directly related to each other through $\alpha = k/\rho c$, where ρ is the density and c is the specific heat, the reduction in thermal conductivity with increased doping concentration directly leads to a lower value for the thermal diffusivity.

The diffusion coefficient of a semiconductor is a very important quantity, which determines the distance traveled by photoexcited carriers before their recombination. It is seen from the values obtained for the diffusion coefficient that it is not the ambipolar transport but the diffusion of minority carriers that essentially determines the signal generation. This means that at an incident power of 50 mW, the photoinduced carrier population is determined by the impurity concentration. It is also seen from the data that the diffusion coefficient decreases with increase in doping concentration. The diffusion coefficient is directly related to the mobility of carriers through the Einstein relation $D = kT\mu/e$, where k is the Boltzmann constant, T is the temperature, e is the carrier charge, and μ is the carrier mobility. At a constant temperature, the diffusion coefficient is essentially determined by the mobility of photoexcited carriers. The mobility of holes decreases with increase in doping concentration, which results in a reduced value of the diffusion coefficient.

Incorporation of dopants has a strong influence on surface recombination. The dopants act as scattering centers, which lead to a deterioration in the transport properties of the photoexcited carriers. One of the effects of incorporation of dopants is the generation of macrosteps (terraces), and the presence of macrosteps gives rise to striation. It was reported that the surface recombination velocity of the photoexcited carriers increases with increase in doping concentration.

tration of Ge on an epitaxial layer of GaAs,¹⁰ which agrees with our experimental result. It can be understood from the relation $\nu = \sigma v_{th} N_{st}$ (where σ is the capture cross section for the photoexcited carriers, v_{th} is the thermal velocity of the photoexcited carriers, and N_{st} is the number of trapping centers per unit area) that the surface recombination velocity is directly proportional to the density of surface trapping centers. The number of trapping centers for the photoexcited carriers at the surface of the epitaxial layer increases with doping, leading to an increase in the surface recombination velocity, which is in agreement with our experimental result.

The doping concentration influences the recombination time of the photoexcited carriers. It is important to point out that the photoacoustic signal is very sensitive to the carrier lifetime, so that the proper choice of this parameter is a significant step in the simulation process. The total carrier lifetime depends on various recombination processes. In the indirect-bandgap semiconductors like Si, nonradiative recombination is the dominant process, whereas in the direct-bandgap semiconductors like GaAs, radiative recombination dominates. Hence the evaluation of nonradiative recombination in GaAs and the study of variation of the nonradiative recombination time with doping have great physical significance, especially with respect to design and fabrication of semiconductor light sources. It is seen from our experiment that the recombination time decreases with increase in doping concentration. This is because the recombination time is directly related to the mobility of the photoexcited carriers. Since the mobility of carriers decreases with increase in doping concentration, the recombination time also decreases with increase in doping concentration. Our values for the recombination time are well within the range of earlier reported values for doped samples.⁶

In conclusion, we have demonstrated in this paper the capability of the PA technique in general and OPC detection in particular to study the thermal and transport properties of photoexcited carriers in layered semiconductor structures. We have investigated the influence of doping on the thermal and transport properties of the epitaxial layer of GaAs doped with Si of various concentrations, using thermal wave transmission and detection technique. From the analysis of experimental data, it is obvious that the thermal diffusivity of epitaxial layers decreases with increase in doping concentration. It is likewise seen that the diffusion coefficient of the minority carriers decreases with increase in doping concentration, which is due to the reduction in the mobility of carriers with doping. Doping also influences the surface recombination velocity and the nonradiative recombination time. The surface recombination velocity of the photoexcited carriers increases with increasing doping concentration, whereas the nonradiative recombination time decreases. This paper shows that the PA technique in the transmission detection configuration is a simple and effective method for the study of thermal and transport properties in semiconductors.

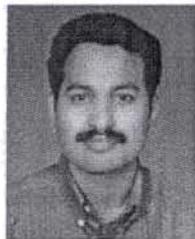
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