Photothermal deflection studies of GaAs epitaxial layers

Nibu A. George, C. P. G. Vallabhan, V. P. N. Nampoori, and P. Radhakrishnan

Photothermal beam deflection studies were carried out with GaAs epitaxial double layers grown on semi-insulating GaAs substrates. The impurity densities in thin epitaxial layers were found to be the effective thermal diffusivity of the entire structure. © 2002 Optical Society of America

OCIS codes: 120.4290, 160.6000, 300.6430, 310.6870.

1. Introduction

The absorption of intensity modulated optical radiation by a sample leads to periodic heat generation, thereby causing excitation of thermal waves in the sample. Thermal wave physics is emerging as a valuable tool in the study of thermal properties of materials, especially in the semiconductor industry.1-5 Among the various methods for investigating these thermal waves the photothermal deflection (PTD) technique possesses some unique characteristics and advantages compared with the other approaches.5-7

The concept of light-beam deflection by thermally induced changes in the index of refraction of a medium has been known for a long time. However, only in did Boccara et al. 8 demonstrated the use of photothermal beam deflection as a valuable tool in material characterization. In subsequent years, theoretical and experimental contributions by Jackson et al.9 in 1981, Aamodt and Murphy10 in 1981, and Grice et al.11 in 1983 formed a strong basis for this technique. The PTD technique is essentially based on detection of the refractive-index gradient associated with a temperature gradient. Absorption of optical radiation (the pump beam) results in the generation of thermal waves in a solid sample, which eventually generates a temperature gradient in a gas or a liquid that is in contact with the sample’s surface. The refractive-index gradient associated with this temperature profile can be conveniently monitored by a second laser beam (the probe beam), which passes through the heated region, because the beam is deflected by the refractive-index gradient.

In the present paper we describe the use of the PTD technique for the thermal characterization of multilayer samples. Among the various experimental approaches to evaluating the thermal diffusion of solids by use of PTD, the strategy that we use in the present investigation is measurement of the signal phase as a function of pump–probe offset and fixed modulation frequency.6,12,13

2. Theory

The PTD technique can be employed in various detection configurations for the investigation of samples.6,7,14,15 Probe-beam deflection in the skimming configuration is one of the widely accepted simple approaches among the PTD measurement schemes. A schematic representation of the probe-beam skimming configuration is shown in Fig. 1. In this configuration the solid sample is irradiated by a focused laser beam and the resultant refractive-index gradient generated in the coupling fluid (a gas or a liquid) that is in contact with the sample surface is monitored with a low-power probe beam passing through this gradient. In this scheme it is assumed that the temperature distribution in the coupling fluid located close to the sample surface is the same as that which is located at the sample surface. The probe beam propagating through a spatially varying refractive-index gradient is deflected from its normal path, and the amount of deflection is determined by a number of thermal and optical parameters of the solid sample.

For a Gaussian beam propagating through a homogeneous medium, most of the beam power can be deduced from the analysis made by Mayo and Royce.16 The propagation of a light beam through a medium with a refractive-index gradient is expressed by the paraxial wave equation

\[ \frac{\partial^2 E}{\partial z^2} + \frac{1}{2} \left( \frac{\partial^2 E}{\partial r^2} + \frac{1}{r} \frac{\partial E}{\partial r} \right) = 0, \]

where \( E \) is the electric field amplitude, \( z \) is the propagation distance, \( r \) is the radial distance from the beam axis, and \( \nabla^2 \) is the Laplacian operator. The solution of this equation for a Gaussian beam is given by

\[ E(r, z) = E_0 e^{-r^2 / w^2} e^{-i k z}, \]

where \( E_0 \) is the amplitude of the incident beam, \( w \) is the beam waist, and \( k \) is the wave number. The deflection of the beam is given by

\[ \delta \approx \frac{\Delta n}{n_0} \frac{w^2}{L}, \]

where \( \Delta n \) is the refractive-index gradient, \( n_0 \) is the refractive index of the medium, and \( L \) is the distance between the sample and the detector.


\[ \frac{d}{ds} \left( n_o \frac{dr_s}{ds} \right) = V_n \frac{d}{ds} n(r, t) \]  

\[ n_o \frac{dr_s}{ds} = \phi(t) = \frac{1}{n_o} \frac{\partial n}{\partial T} \int_\text{path} V_n T(r, t) ds \]

where \( A \) is a complex integration constant, \( \delta \) is a spatial Fourier-transformed variable, and \( \beta_0 = (\delta^2 + jw/D_0)^{1/2} \)

Practically, the condition \( a = b = z = 0 \) cannot be achieved, and finite values of \( a, b, \) and \( z \) may result in a change in slope, especially when the sample possesses low thermal diffusivity. But, for samples with moderately high thermal diffusivity, Eq. (6) holds for finite values of \( a, b, \) and \( z \).

3. Experimental

The thin films were grown by the molecular beam epitaxial method (Technical University of Eindhoven, Eindhoven, The Netherlands). All of the samples contained two epitaxial layers. The sample structure together with the specifications of each layer, including the growth conditions and dopants, are given in Table 1. For convenience we have labeled the samples arbitrarily 1, 2, 3, and 4.

Continuous-wave laser emission at 488 nm from an argon-ion laser (Liconix 5000) was used as the pump beam. The laser beam had a (1/e²) diameter of 1.2 mm. In all the measurements a laser power of 50 mW (±0.5%) was used. Carbon tetrachloride (CCl₄), which is the most suitable and most commonly used coupling fluid in photothermal deflection studies, was used as the coupling fluid to the sample. The significant parameters that make CCl₄ a good coupling fluid in the PTD technique are its low thermal diffusivity, \( \alpha = 7.31 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1} \) and its very high rate of change of refractive index with respect to temperature, \((dn/dT) = 6.12 \times 10^{-4} \text{ K}^{-1} \).

A schematic view of the experimental setup is de-
The pump-beam spot size at the sample surface was estimated to be $102 \mu m$. The mirror and cuvette to a height of $-10 \text{ mm}$ above the sample the lens ($L_t$, Fig. 2) were fixed on an arrangement one can accurately vary the pump-beam's position on the sample along the $x$ direction simply moving the translator in the $x$ direction; the mechanical chopper (Stanford Research System Model SR540) was placed in the pump-beam path to modulate the pump beam's intensity at the pump frequency.

A 3-mW He–Ne laser (Spectra-Physics) emitting at 632.8 nm was used as the probe beam to detect the strength of the refractive-index gradient generated in a direction orthogonal to the pump beam (z axis). A plastic fiber with a circular core of 1-mm diameter was used as a position-sensitive detector for the periodic deflection of the probe beam. The end of the fiber was firmly fixed upon an argon-ion laser at a distance of 15 cm from the sample. In other end of the fiber was coupled to a 0.25-m monochromator (McPherson) tuned to the probe beam wavelength. A photomultiplier tube was placed on the exit slit of the monochromator. The output from the photomultiplier tube was fed to a dual-phase lock-in-amplifier (Stanford Research Systems Model SR830) through an impedance-matching circuit. The entire experimental setup was laid on a moderately vibration-isolated table to protect the system from ambient vibrations. Measurements were carried out at a pump-beam modulation frequency of 10.6 Hz, and the distance between a probe beam height and the sample surface was as small as possible to produce a nondiffracted signal across the probe beam. A typical variation of the signal phase with the pump-probe offset for each sample is shown in Fig. 3. The maximum in the phase plot corresponds to zero offset. Identical signal profiles were observed for the four samples. In the present experiment the $p$-type substrate was grown upon the other surfaces of the GaAs epitaxial layers as well as of the substrate sides. Measurements were carried out by irradiating the thin-film sides as well as of the substrate sides of the four samples. In the present experiment the $p$-type substrate was grown upon the other surfaces of the GaAs epitaxial layers. The literature values of $\alpha$ of GaAs are significant influence on the PTD signal generated from the substrate. The literature values of $\alpha$ of GaAs.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$T$ (C°)</th>
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<tr>
<td>1 Si-doped GaAs (upper)</td>
<td>610</td>
</tr>
<tr>
<td>Si-doped GaAs (middle)</td>
<td>685</td>
</tr>
<tr>
<td>Semi-insulating GaAs (substrate)</td>
<td>400.0</td>
</tr>
<tr>
<td>2 Si-doped GaAs (upper)</td>
<td>610</td>
</tr>
<tr>
<td>Si-doped GaAs (middle)</td>
<td>685</td>
</tr>
<tr>
<td>Semi-insulating GaAs (substrate)</td>
<td>400.0</td>
</tr>
<tr>
<td>3 Si-doped GaAs (upper)</td>
<td>580</td>
</tr>
<tr>
<td>Si-doped GaAs (middle)</td>
<td>630</td>
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<tr>
<td>Semi-insulating GaAs (substrate)</td>
<td>400.0</td>
</tr>
<tr>
<td>4 Be-doped GaAs (upper)</td>
<td>610</td>
</tr>
<tr>
<td>Be-doped GaAs (middle)</td>
<td>695</td>
</tr>
<tr>
<td>Semi-insulating GaAs (substrate)</td>
<td>400.0</td>
</tr>
</tbody>
</table>

*Thickness of layer.
*Electron concentration.
*Substrate temperature at which the layers are grown.
*Hole concentration ($p$).

Table 1. Structure, Properties, and Growth Conditions of the Doped GaAs Epitaxial Layers on the Semi-Insulating GaAs Substrate.

4. Results and Discussion

Measurements were carried out by irradiation of the GaAs epitaxial layers. Almost identical signal profiles were observed for the four samples. In the present experiment the $p$-type substrate was grown upon the other surfaces of the GaAs epitaxial layers, as well as the substrate sides. Identical signal profiles were observed for the four samples. In the present experiment the $p$-type substrate was grown upon the other surfaces of the GaAs epitaxial layers, as well as the substrate sides.

Fig. 2. Schematic view of the experimental setup: M, mirror; C, chopper; L1, L2, lenses; Q, cuvette; S, sample; OF, optical fiber; MC, monochromator; PMT, photomultiplier tube.
The range of $\alpha$ values reported in the literature is 0.2–0.36 cm$^2$ s$^{-1}$, and the experimentally observed values also fall within this range. The large variation of thermal transport properties of semiconductor materials with the growth conditions, defects, etc., is similar to the heat conduction in dielectrics, where free carriers make the main contribution to heat transport in these materials.\textsuperscript{26,26}

Figure 4 shows the variation of PTD signal phase with pump-probe offset for sample 1 when the film side is facing the pump beam. Again, the thermal diffusivities of the films were evaluated from the slope of the plot, and the average values of the measured $\alpha$ are listed in Table 2.

The excitation photon energy, 2.54 eV, is much greater than the bandgap energy of GaAs (1.43 eV), and the entire energy is absorbed at the surface (about 1 $\mu$m) of the epitaxial layer itself. Consequently the samples are optically opaque at the excitation wavelength. Moreover, the fact that the entire energy is absorbed at the surface of the sample implies that heat is generated in the surface epitaxial layer but propagates through the entire structure. Nevertheless, the decrease of $\alpha$ values of the epitaxial layers compared with the bulk diffusivity value suggests some interesting but complex heat transport mechanisms in these samples. The increase in the number of scattering centers that resulted from doping of GaAs with either Si or Be and the consequent reduction of the phonon mean free path do not seem to be

<table>
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<th>Sample Number</th>
<th>Film-Side Illumination</th>
<th>Substrate-Side Illumination</th>
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</thead>
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<tr>
<td>1</td>
<td>0.193</td>
<td>0.212</td>
</tr>
<tr>
<td>2</td>
<td>0.187</td>
<td>0.212</td>
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<tr>
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<td>4</td>
<td>0.160</td>
<td>0.206</td>
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the only reasons for the noticeable reduction in the diffusivity values of the epitaxial layers. Recently a number of research papers reported the experimental observation of substantial reduction (as much as 50%) in lattice thermal conductivity in semiconductor thin films, especially when the thin-film thickness was of the order of the phonon’s mean free path. But the exact values of the phonon’s mean free path in the samples investigated here are not available for detailed analysis. According to the kinetic theory the phonon’s mean free path in the bulk sample $\lambda_{\text{bulk}}$ can be evaluated from the relation

$$\lambda_{\text{bulk}} = \frac{3k_{\text{bulk}}}{Cv},$$

where $k_{\text{bulk}}$ is the bulk thermal conductivity, $C$ is the volumetric heat capacity, and $v$ is the speed of sound in the material. For bulk GaAs, $k_{\text{bulk}} = 0.46 \text{ W/(cm/}°\text{C)}$, $C = 0.33 \text{ J/(g/}°\text{C)}$, and $v \approx 4.0 \times 10^5 \text{ cm/s}$, which lead to the estimation of a phonon mean free path of approximately $100 \text{ nm.}$ This value is smaller than those of the surface layer thicknesses 200 and 250 nm, of the samples. However, the estimated value of the phonon’s mean free path need not be strictly true, because there is a large discrepancy between the experimentally observed value of the phonon’s mean free path and that evaluated theoretically; hence it is hard to analyze the observed thermal diffusivity data without knowing the exact value of $\lambda_{\text{bulk}}$. For example, in a recent paper Ju and Goodson measured the effective phonon mean free path in Si as 300 nm, whereas that evaluated by the kinetic theory is only 43 nm. But an important point to be mentioned is that the thicknesses of the thin films are too small compared with the thermal diffusion length and hence the tabulated thermal diffusivity value may be the effective diffusivity of the thin films and that of the substrate. However, a general conclusion that can be drawn from the tabulated thermal diffusivity values is that, for samples 1–3, all doped with Si, the trend is for a decrease in thermal diffusivity with a decrease in doping density and an increase in thickness of the second (under) epitaxial layer. Also, for sample 4, which is doped with Be, the diffusivity value is smaller than that of the corresponding $n$-type sample (sample 1).

This study is supported by the Netherlands University Federation for International Collaboration, The Netherlands. The authors thank J. H. Wolter and J. E. M. Haverkort, Technical University of Eindhoven, Eindhoven, The Netherlands, for providing the sample.

References