Photoacoustic studies on n-type InP

Nibu A. George*
C. P. G. Vallabhan
V. P. N. Nampoori
P. Radhakrishnan
Cochin University of Science and Technology
International School of Photonics
Cochin 682022, India

Abstract. We discuss an open photoacoustic cell study on sulfur-doped n-type InP wafer. The thermal diffusivity of the sample is evaluated from the phase data associated with the photoacoustic signal as a function of the modulation frequency under heat transmission configuration. Analysis is made on the basis of the Rosencwaig-Gersho theory and the results are compared with those from earlier reported photoacoustic studies of semiconductors. Our investigation clearly indicates that the instantaneous thermalization process is the major heat diffusion mechanism responsible for the photoacoustic signal generation in an InP sample. © 2002 Society of Photo-Optical Instrumentation Engineers.

Subject terms: photoacoustic and photothermal science and engineering; indium phosphide; heat diffusion in semiconductors.

Paper 010144 received Apr. 25, 2001; revised manuscript received July 9, 2001; accepted for publication July 10, 2001.

1 Introduction
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 parameters of materials, especially in the semiconductor industry.1–3 Among the various methods used to study these thermal waves, the gas-microphone photoacoustic (PA) technique is the most powerful and commonly employed technique.4,5 Apart from the pure spectroscopic studies in the very beginning, the PA method has now grown to a multipurpose analytical tool for the evaluation of various thermal and optical parameters of different classes of materials. In many of the earlier reported papers, PA signal amplitude data as a function of the modulation frequency was mainly used for the thermal diffusivity measurements in semiconductors.1,6,7 However, the major renaissance in this field was made by Dramicanin et al.8 by analytically evaluating the expression for the distribution of the periodic thermal flux originating from three principal thermal sources, namely, the instantaneous thermalization component, the nonradiative bulk recombination, and the nonradiative surface recombination. The instantaneous thermalization is an aftereffect of the excited electron-phonon collision, whereas the bulk recombination arises as a result of the band-to-band recombination ofphotoexcited carriers that are diffused into the bulk of the semiconductor and the surface recombination is essentially governed by surface imperfections or trapping centers. The Dramicanin model is very useful for the analysis of PA signal amplitude and phase at the front and rear surfaces of semiconductor samples. Subsequently, in more recent years, using PA phase measurements in the heat transmission configuration, carrier transport properties such as carrier diffusion coefficient, carrier recombination velocity, and mean recombination time were evaluated together with the thermal diffusivity of a large number of semiconducting samples,9–17 namely, GaAs, CdTe, Si solar cells, CdInGaS4, InSb, GaSb, etc.

In this paper, we report the thermal diffusivity of sulfur-doped n-type InP sample measured using an open photoacoustic cell in the heat transmission configuration. The thermal diffusivity is defined by \( \alpha = k/(\rho C) \), where \( k \) is the thermal conductivity, \( \rho \) is the density, and \( C \) is the specific heat capacity of the sample. Here, \( \alpha \) is a significant thermophysical parameter that determines the heat diffusion in bulk as well as thin film samples. Being a widely used and important substrate material in the field of semiconductor technology, measurement of the thermal diffusivity and a detailed analysis of the heat diffusion processes of the sample have great practical significance.

2 Experimental Section
The cross-sectional view of the open PA cell (OPC) configuration used for this investigation is shown in Fig. 1. The cell has a provision to illuminate the sample from the rear side as well as the front side. In this study we used only the rear-side illumination or the so-called heat transmission configuration. The InP wafer is fixed to the top of the air chamber of the OPC using vacuum grease at the edges and the sample is irradiated on its surface facing the ambient. Modulated optical radiation at 488 nm from an argon ion laser (Liconix 5000) is used as the source of excitation. The original laser beam has a 1/e2 diameter of 1.2 mm and is used without further focusing to avoid lateral heat flow. The PA signal is produced in a small volume of air in between the sample and the microphone. The signal is then detected using a highly sensitive electret microphone (Knowles BT 1834). The phase of the signal as a function of the modulation frequency of the laser beam is recorded using a dual-phase digital lock-in amplifier (Stanford Research Systems SR830). Two different laser power levels, 50 and 200 mW,

*Current address: Delft University of Technology, Dept. of Applied Physics, Lorentzweg 1, 2628 CJ Delft, The Netherlands. E-mail: nibu@ws.tn.tudelft.nl.
with a stability of ±0.5% are used for the investigations. As the sample has different surface roughnesses on its opposite faces, studies were made with both faces for the laser irradiation.

3 Methodology and Results

As indicated earlier, the heat conduction in a photoirradiated semiconductor sample is mainly contributed by three factors. If the excitation energy is more than the bandgap energy of the semiconductor material, then under certain experimental conditions, the photogenerated carrier recombination processes (surface and bulk) will also contribute to the heat transport. The usually reported order of various thermal diffusion mechanisms in the thermally thick regime of a semiconductor is that the instantaneous thermalization component comes first, followed by the heat transfer by the bulk recombination process, and finally the surface recombination mechanisms. This means that the instantaneous thermalization component will dominate the other two mechanisms in the lower range of modulation frequencies, whereas at very high frequencies the latter two mechanisms will predominate. However, the terms describing the contributions from carrier recombination processes are not discussed here since these mechanisms are found to be absent in this case.8 From the 1-D heat flow model of Rosencwaig and Gersho, one can arrive at the expression for the PA signal generated by the pure thermal wave component in the heat transmission configuration as18

\[
\delta P = \frac{\gamma P_0 I_0 (\alpha_s \alpha_t)^{1/2}}{2 \pi \Gamma_1, \sigma_1} \exp[j(\omega t - \pi/2)].
\]  

(1)

If the sample is optically opaque, i.e., if the excitation energy is greater than the bandgap energy, and thermally thick (i.e., \( l_s \alpha_s \gg 1 \)), then Eq. (1) reduces to

\[
P \approx \frac{\gamma P_0 I_0 (\alpha_s \alpha_t)^{1/2}}{\pi \Gamma_1, \sigma_1} \frac{e^{-l_s (\pi f / \alpha_s)^{1/2}}}{f} \times \exp[j(\omega t - \pi/2 - l_s \alpha_s)],
\]

(2)

where \( l_s \) and \( \alpha_s \) are the sample thickness and the thermal diffusion coefficient, respectively, and the balance of the notation has the usual meaning, as defined in Rosencwaig-Gersho theory.18 Equation (2) implies that for a thermally thick sample, the amplitude of the PA signal decreases exponentially with the modulation frequency according to \((1/f) \exp[-l_s (\pi f / \alpha_s)^{1/2}]\), while the phase \( \phi \) decreases linearly with \(-l_s (\pi f / \alpha_s)^{1/2}\). The preceding equation holds only when the contributions from the nonradiative recombination processes are negligible. Hence, the thermal diffusivity \( \alpha_s \) of the sample can be obtained either from the amplitude plot or from the phase plot. Though in many reports the amplitude behavior is used to evaluate the thermal diffusivity of solid disklike samples, the necessary condition for employing this approach is that the detector (microphone) should have flat response over the frequency range of interest. Otherwise, complicated normalization procedures are required as reported by Nikolic et al.9 However, the nonflat response of the microphone will not affect the phase data and hence it seems to be a simpler and more reliable strategy. But, a major drawback of the phase method is that the phase plot will not obey a linear relationship, as predicted by Eq. (2) when thermoelastic bending of the sample is a dominant mechanism, which is usually observed with samples having very low thermal diffusivity.19

Figure 2 shows the variation of the PA signal phase with the square root of the modulation frequency for two different pump powers. Sulfur-doped n-type InP sample grown by the LEC method (Sumitomo Electric Industries, Japan) has a carrier concentration of \( 4 \times 10^{18}/\text{cm}^3 \). One surface of the 350-μm-thick wafer is polished and etched while the other surface is preetched. From Fig. 2, we can identify a linear portion that satisfies Eq. (2). The deviation from the straight-line fit in the low-frequency region is obviously due to the fact that the sample is thermally thin in this regime. From the slope of the plot, we calculated the thermal diffusivity of the sample as 0.401 (±0.005) \( \text{cm}^2/\text{s} \). The measured value for the n-type InP is slightly less than the thermal diffusivity 0.4569 \( \text{cm}^2/\text{s} \) (\( k = 0.68 \\text{W cm}^{-1} \text{K}^{-1} \), \( \rho = 4.79 \text{ g cm}^{-3} \), \( C = 3.017 \text{ J g}^{-1} \text{K}^{-1} \)) of pristine InP (Refs. 20–22). In doped samples, the impuri-
ties and the associated free carriers can impede heat transport. These scattering processes are complicated by phonon dispersion, which results in a reduction in the phonon group velocities, and consequently a reduced thermal conductivity (diffusivity).

Moreover, the linear dependence of the phase data on the square root of the modulation frequency is an important result that must be analyzed in detail. First, this observation implies that the thermoelastic bending is not a contributing factor to the PA signal. This may be due to the fact that the InP sample possesses moderately high thermal diffusivity and the sample has sufficiently large thickness. Then the heat generated at the irradiating surface is transmitted quickly to the other side without leaving a considerable temperature gradient along the thickness of the specimen. But in the case of materials with low thermal diffusivity, the thermoelastic bending may dominate and in such cases appropriate corrections have to be incorporated in the calculation.10

Note also that in many OPC studies involving a variety of semiconductors, more complex phase behavior with a minimum in the phase plot is reported.9–17 This nonlinear behavior of the phase data is attributed to surface and bulk recombination processes of photogenerated carriers in those materials. However, our observation indicates that for this sample, the photogenerated excess carrier recombination processes do not contribute to the PA signal in any significant manner in the frequency range of our investigations. To confirm this we used two different incident laser powers, 50 and 200 mW, and the corresponding phase plots are shown in Fig. 2. However, both plots yield the same value of diffusivity, which supports our arguments. Apart from this, it is a well-known fact that the surface roughness of semiconductor samples has a pronounced influence on the carrier recombination process. To check whether the surface roughness has any influence on the PA signal phase data, we used two different configurations. In the first case, the polished side of the sample was illuminated and the PA signal was detected at the rough side, and in second case, the sample was turned upside down and its rough side was illuminated and the PA signal was detected at the polished side. But the results obtained (Fig. 3) again lead to the same diffusivity value and this also confirms that throughout the frequency range of our investigations, the instantaneous thermalisation component is the major contributing factor to OPC signal generation from the present sample. If the photogenerated carrier recombinations (surface and bulk) are contributing to the PA signal, then its influence should be visible within the frequency range of our investigations.9–17

4 Conclusion

We measured the thermal diffusivity of n-type InP from the phase data of the OPC signal under the heat transmission configuration. In this configuration, the pure 1-D heat flow model of Rosencwaig and Gersho can be used for the thermal diffusivity measurements of solid disklike materials having moderately high diffusivity values. The results of our investigation show that in the case of semiconductor samples, this simple and direct approach can also be applied for the thermal diffusivity measurements, provided the contributions from photogenerated carrier recombination are negligible.

Acknowledgments

This work is supported by Netherlands University Federation For International Collaboration (NUFFIC), the Netherlands. The authors would like to thank Prof. J. H. Wolter and Dr. J. E. M. Haverkort, Technical University of Eindhoven, the Netherlands, for providing the sample. One of the authors (NAG) wishes to acknowledge the Cochin University of Science and Technology for providing financial assistance.

References


Nibu A. George received his MSc degree in physics with a specialization in laser physics from Pondicherry Central University, India, in 1997. He is currently a senior research fellow working toward his PhD degree at the International School of Photonics, Cochin University of Science and Technology, India. His research interests include optothermal effects in liquid crystals, semiconductors, organic dyes, polymers, and ceramics.

C. P. G. Vallabhan received his MSc and PhD degrees in 1965 and 1971, respectively, from Kerala University and did postdoctoral research at Southampton University in the United Kingdom. He was also a visiting professor at the Fraunhafer Institute, Freiberg, Germany, and the Eindhoven University of Technology, the Netherlands. He is currently a professor at the International School of Photonics, Cochin University of Science and Technology. His research interests include lasers, laser matter interactions, opto-electronics, and molecular and solid state physics. He has published more than 200 research papers in these areas. He is a fellow of the Optical Society of India, president of the Photonics Society of India, and vice president of the Indian Laser Association.

V. P. N. Nampoori received his MSc and PhD degrees from M. S. University, Baroda, in 1974 and 1978, respectively. He is currently a professor at the International School of Photonics, Cochin University of Science and Technology. His research interests include thermal lens spectroscopy, fluorescence spectroscopy, nonlinear optics, fiber optics, and laser-produced plasma. He is the author or coauthor of more than 200 research papers.

P. Radhakrishnan received his MSc degree in physics from Kerala University in 1977, and his PhD degree from Cochin University of Science and Technology in 1986. He was a lecturer with the Cochin College from 1979 to 1988. He is currently a professor with the International School of Photonics, Cochin University of Science and Technology. His research interests include laser technology, laser spectroscopy, and fiber optic sensors. He is a member of the executive committee of the Photonics Society of India.