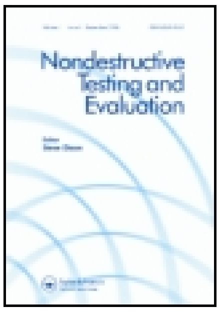

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Photoacoustic investigation of doped InP using open cell configuration

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PHOTOACOUSTIC INVESTIGATION OF DOPED InP USING OPEN CELL CONFIGURATION

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An open cell photoacoustic (PA) configuration has been employed to evaluate the thermal diffusivity of intrinsic InP as well as InP doped with tin and iron. Thermal diffusivity data have been evaluated from variation of phase of PA signal as a function of modulation frequency. In doped samples, we observe a reduced value for thermal diffusivity in comparison with intrinsic InP. We also observed that, while the phase of the PA signal varies linearly with the square root of chopping frequency for doped samples, the intrinsic material does not exhibit such behaviour in the experimental frequency range. These results have been interpreted in terms of the heat generation and phonon assisted heat diffusion mechanisms in semiconductors.

Keywords: Photoacoustic; InP; Thermal diffusivity

INTRODUCTION

The study of photothermal phenomena has in recent years become an active and important area of research in the field of material sciences. It has proven to be a valuable tool in evaluating thermal, optical and electronic properties of materials [1,2]. In particular, the photoacoustic (PA) technique, which is based on the thermal wave physics, has been shown to be an important method in the study of thermal parameters of materials as well as in the semiconductor industry [3]. In a PA setup periodic nonradiative deexcitation following an optical absorption generates thermal waves in the sample and its propagation through the coupling gas produces acoustic waves. These acoustic waves can be detected using a sensitive microphone thereby producing PA signal. Besides being an effective spectroscopic tool, laser-induced photothermal effect has been established as a sensitive method to study the dynamics of photoexcited carriers in semiconductors.

All the photothermal methods are based on the detection, by one means or other, of transient thermal waves that arise in the sample as a result of absorption of modulated light. The absorbed energy is converted into heat in the bulk as well as on the surface of the sample thereby producing pressure fluctuations in the ambient gaseous medium, which appears as

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the PA signal when detected by a suitable transducer. In the PA method, experimentally measured PA signal enables us to obtain information about the heat source generating the thermal waves. Thus the PA response also enables us to gather information regarding the thermal parameters, structural formations and inhomogeneities, in addition to the optical properties of the sample.

Thermal diffusivity depends on how heat diffuses through the sample and this diffusion of heat in semiconductors is an important transport property, which is of major consideration in semiconductor technology. The thermal diffusivity values of various semiconductors have been evaluated in the past by measuring PA signal as a function of modulation frequency [4,5]. The transport properties and lifetimes of the photoexcited carriers have also been studied using PA technique. A detailed discussion of the contributions of various factors to thermal flux in semiconductors under periodic optical excitation is given by Pinto Neto *et al.* [4,5]. In their work they studied the influence of carrier recombination and diffusion in PbTe and Si as well as thermal diffusivity of GaAs and Si. They observed contributions arising from both thermalisation and nonradiative recombination of photoexcited carriers in the bulk and surface of the material.

Analytical solution of the thermal flux generated in the semiconductors is given by Dramicanin *et al.* [6], who treated three distinct processes to generate heat, namely instantaneous thermalisation due to the electron–phonon interaction, and nonradiative recombination of photoexcited carriers in the surface and in bulk, respectively. The microphone version of PA technique can be employed in two ways, either in the reflection detection configuration (RDC) or in the transmission detection configuration (TDC). It is reported that, in the low frequency range (10–10,000 Hz), the TDC is more suitable than RDC to obtain the thermal and transport properties of the semiconductors [7]. The TDC, which is basis of design of the Open Photoacoustic Cell (OPC), is extensively used to measure both thermal and transport properties of a large number of samples such as GaAs, Si, Solar cells, $CdInGeS_4$, PbTe, etc. [4,5,8]. However, the effect of doping on the thermal and transport properties of semiconductor samples has not been studied in detail, except for a few reports [9,10]. Castro-Rodriguez [9] measured the effect of doping on the thermal transport of In doped CdTe thin films. Reich *et al.* [10] studied the effect of doping on the surface recombination velocity of Ge doped GaAs.

In this paper, we report the measurement of thermal diffusivity of pure InP and InP doped with tin and iron using an OPC. The thermal diffusivity determines the temperature distribution around a point heat source in a semiconductor. This quantity is given by

$$\alpha = \frac{k}{\rho C} \tag{1}$$

where k is the thermal conductivity, ρ the density and C is the specific heat capacity of the sample. Thermal diffusivity data is of vital importance in the context of semiconductor devices under actual operating conditions when they are subjected to a thermal load. InP is a key material for high speed electronic and optoelectronic devices. Hence, the measurement of α in pure and doped InP and a detailed analysis of heat diffusion process in this material when thermal loading influences their transport behaviour, have great physical significance, as far as device performance is concerned.

THEORY

As shown in Ref. [5], the heat generation phenomenon in semiconductors after irradiation with chopped optical beam arises due to three processes namely thermalisation, nonradiative bulk recombination and nonradiative surface recombination. If the excitation energy is greater than band gap energy, under certain experimental condition (frequency up to which thermal diffusion length becomes roughly five times the sample thickness), thermalisation is the dominating factor to PA signal generation in semiconductors [6]. It must be remembered here that the thermal diffusion length is inversely proportional to the chopping frequency, $\mu = \sqrt{2\alpha/\omega}$. For higher chopping frequencies, the contribution from nonradiative recombination of photoexcited carriers in bulk and on the surface of the sample becomes more dominant over the thermalisation component in the PA signal generation mechanism. In the earlier reported PA studies on semiconductors [4–6], it is shown that the sequence of the different thermal diffusion processes is as follows. In the thermally thick region of a semiconductor, the instantaneous thermalisation component comes first, followed by heat transfer due to the bulk recombination process and finally, the surface recombination process. However the heat generated in the first process has to diffuse through phonons only and hence it is slower. This means that in the lower chopping frequency range, pure thermal wave component dominates over the other two, whereas at high frequencies, the latter two mechanisms become significant. From one dimensional heat flow model of Rosencwaig and Gersho, one can deduce an expression for the PA signal generated due to the pure thermal wave component in the heat transmission configuration [11] as

$$\delta P = \frac{\gamma P_0 I_0 (\alpha_g \alpha_s)^{1/2}}{2\pi T_0 k_s f \sin\mathrm{h}(l_s \sigma_s)} e^{j(\omega t - \pi/2)} \tag{2}$$

where l_s is the sample thickness, γ the specific heat ratio and $\sigma_s = (1 + j)a_s$, $a_s = (\pi f/\alpha_s)^{1/2}$ is the thermal diffusion coefficient of the material. Other symbols have their usual meaning as defined in Rosencwaig and Gersho theory [12].

For an optically opaque and thermally thick sample ($l_s\sigma_s \gg 1$) the expression reduces to

$$\delta P = \frac{\gamma P_0 I_0 (\alpha_g \alpha_s)^{1/2} e^{-l_s\left(\frac{\pi f}{\alpha_s}\right)^{1/2}}}{\pi T_0 l_g k_s f} e^{j\left(\omega t - \frac{\pi}{2} - l_s a_s\right)} \tag{3}$$

This equation implies that, for a thermally thick sample, the amplitude of the PA signal decreases exponentially with the modulation frequency as $1/f \exp(-b\sqrt{f})$, where $b = l_s\sqrt{\pi/\alpha_s}$ and its phase varies as $\Phi_{th} = -(\pi/2) - b\sqrt{f}$. Thus the value for thermal diffusivity can be obtained from the slope of the plot between phase of the PA signal and square root of modulation frequency. For samples having relatively large thermal conductivity, evaluation of thermal diffusivity from phase data is a more reliable strategy as compared to evaluation from amplitude data of PA signal in the microphone version of PA setup [13].

EXPERIMENTAL

A schematic diagram of our experimental set up is shown in Fig. 1. The OPC used for the present study is a nonresonant PA cell under our experimental frequency range. The cross sectional view of the OPC is shown in Fig. 2. The cell has provision to illuminate the sample from both the rear side as well as the front side. In the present study, we have used the rear side illumination or the so-called heat TDC. The sample is fixed on the top of the air chamber of the OPC using vacuum grease at the edges and the irradiation is made on the exposed surface.

Optical radiation at 488 nm from an argon ion laser (Liconix 5000) is used as the source of excitation, which is modulated using a mechanical chopper (Stanford Research Systems SR 540). The laser beam has a spot size of 1.2 mm and is used without further focusing to avoid

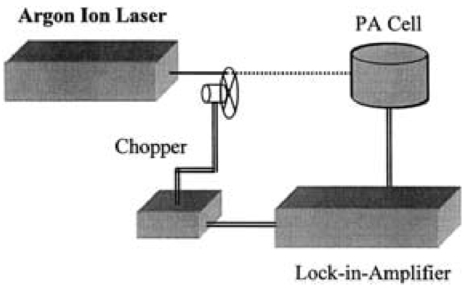

FIGURE 1 Experimental set-up.

lateral diffusion of heat. If the beam is unfocused, it spreads uniformly over the large surface area that in turn generates plane thermal waves, which propagate in one direction unlike hemispherical thermal waves that arise from a focused radiation [14]. These thermal waves produce a PA signal at the same frequency as the chopping frequency, on the rear side the sample, which is detected by a sensitive microphone (Knowles 1834). The phase of the PA signal is recorded using a dual phase lock-in-amplifier (Stanford Research Systems SR 830). The laser power used is 50 and 100 mW with a stability of $\pm 0.5\%$. As the doped samples have different surface qualities on its opposite faces, studies are done on both faces of the sample.

RESULTS AND DISCUSSIONS

The samples used for the present investigation are pure InP as well as InP doped with tin and iron. Intrinsic InP has highly polished surface whereas doped samples have different surface finishing such that one surface is highly polished while the other is roughened. All the samples are grown by liquid phase epitaxy (LPE) method. The undoped sample has a thickness of 520 μ and the InP samples doped with tin and iron have thickness of 374 and 350 μm, respectively. The doping levels of the tin doped InP and the iron doped InP are 10^{18} and $10^{17}\,cm^{-3}$, respectively. The Sn-doped sample is doped in the saturation region so that we can safely take the carrier concentration as equal to doping concentration [15], while the Fe-doped sample should be considered as semi-insulating.

Figure 3 shows the variation of phase of the PA signal as a function of chopping frequency for pure InP. This curve has a minimum due to the fact that, in the experimental frequency

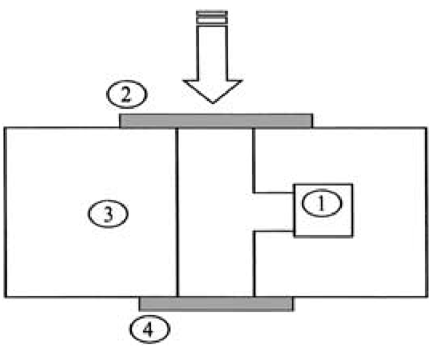

FIGURE 2 Cross-sectional view of the OPC: (1) the microphone; (2) the sample; (3) the acrylic body; (4) the glass window.

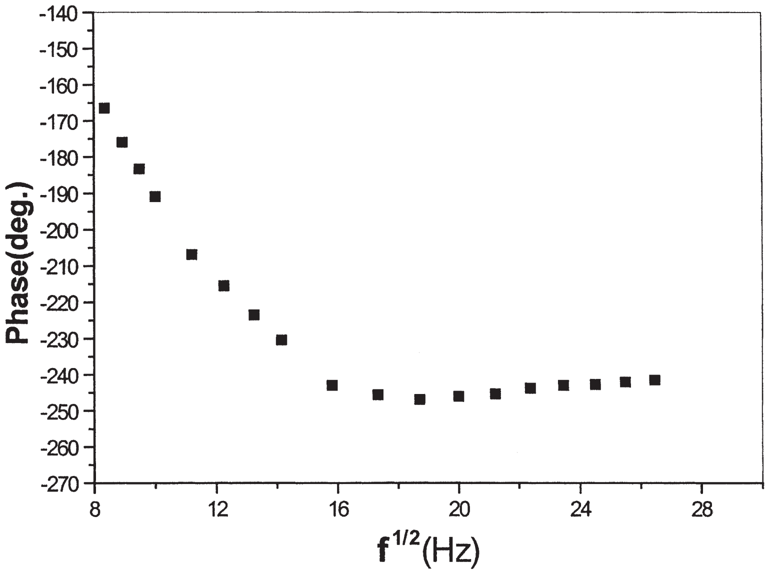

FIGURE 3 OPC phase versus square root of modulation frequency of a polished surface for pure InP.

range, both thermalisation and nonradiative recombination mechanism contribute to the PA signal generation in intrinsic InP. Thus, pure InP behaves in the same way as in reported earlier studies on other compound semiconductors [4–7]. But in the quasi thermally thick region, i.e. in the low chopping frequency range, thermalisation is the dominating source to heat generation mechanism in the semiconductors, as is evident from the linear nature of Fig. 4. The calculated value of thermal diffusivity from the slope of Fig. 4 agrees well with that of earlier reported value [16].

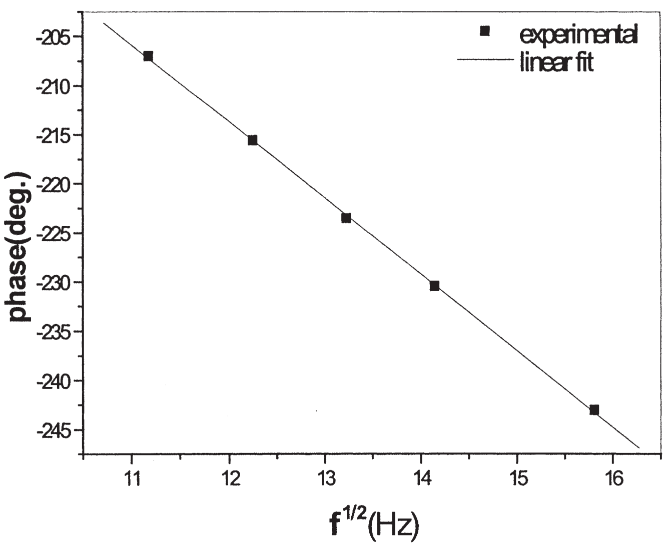

FIGURE 4 OPC phase versus square root of modulation frequency of a polished surface for pure InP in the quasi thermally thick regime.

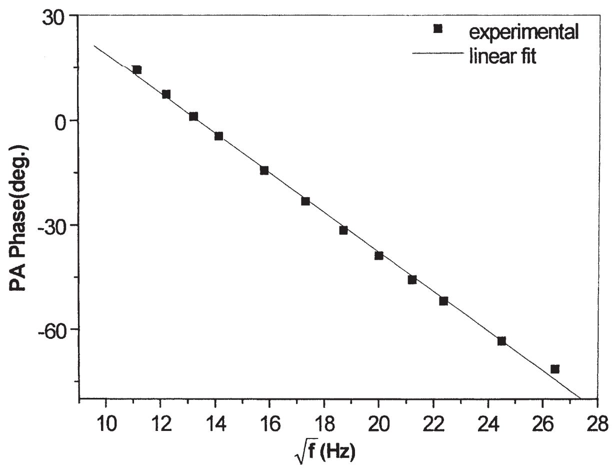

FIGURE 5 OPC phase versus square root of modulation frequency of a polished surface for InP doped with tin.

Figures 5 and 6 show the plots of phase of the PA signal, as a function of square root of modulation frequency in the thermally thick region of InP doped with tin and iron respectively. In both cases the polished surface of the sample is exposed to chopped optical radiation. It is obvious that both the plots show a linear relation between the phase of the PA signal and square root of chopping frequency. This linear relation implies that, thermoelastic bending of sample has no significant influence on the PA signal generation of doped samples. This is due to the fact that our samples have a relatively high thermal diffusivity and a thickness of only 400 μm. The temperature gradient in the sample is thus too small to produce thermoelastic bending. The absence of a minimum in the phase plot of the doped samples implies that, the contribution from nonradiative recombination to the PA signal is negligible [6,13].

In a steady state transport experiment, as in the case of PA effect, electrons and holes are continuously created at high energy by the absorption of light and they lose energy through thermalisation. In general, the photothermal signal from the semiconductor depends not only on how heat is carried away by each quasi particle system but also on how energy and momentum is distributed between them. It is known that for bulk semiconductors, the

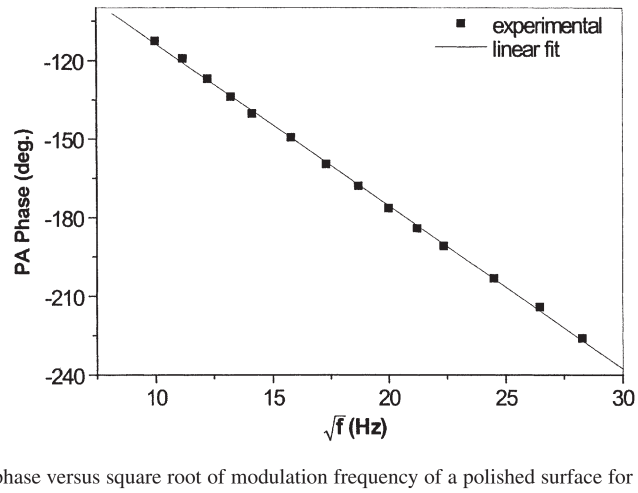

FIGURE 6 OPC phase versus square root of modulation frequency of a polished surface for InP doped with iron.

TABLE I Thermal diffusivity value of doped and undoped InP

Sample	Thermal diffusivity in $cm^2 s^{-1}$	Reported value
InP (Pure)	0.443 ± 0.007	0.44
InP:Sn (*n*-type)	0.388 ± 0.004	–
InP:Fe (Semi-insulating)	0.374 ± 0.006	–

thermal contribution from phonons is much larger than that from carriers. It means that, in semiconductors, most of the heat is carried away by phonons rather than photoexcited carriers. Thus, in the low chopping frequency range, heat generated due to thermalisation component from electron–phonon interaction dominates over nonradiative recombination processes. Introduction of a dopant into the host lattice creates point defects in the lattice, which in turn enhances the phonon scattering mechanism and consequently also the heat diffusion process and thus the PA signal generation mechanism. This behaviour of the thermalisation component should exhibit the linear behaviour of phase plots of PA signal for doped samples, as in Figs. 5 and 6.

From the Table I it is seen that, the thermal diffusivity value of doped samples are less than that of intrinsic InP. This is because lattice thermal conductivity arises from interaction of phonons and scattering of phonons with lattice. Introduction of a dopant into the host lattice produces more scattering centres, which in turn reduces the phonon mean free path. A reduction of the phonon mean free path reduces the lattice thermal conductivity and hence the value of thermal diffusivity. The lattice thermal conductivity is related to doping and temperature through the relation $(1/W) = AT^{-n}$; where W is lattice thermal resistivity. For InP, $n = 1.55$ at 300 K [17]. Hence, at constant temperature, the lattice thermal conductivity is directly related to A and it is a parameter, which decreases with doping. Thus doped samples show a reduced value for thermal conductivity. From Eq. (1), it is clear that thermal conductivity and thermal diffusivity are directly related so that lowering of value of one reduces the other. This explains our experimental observation that a doped sample shows a reduced value of the thermal diffusivity.

It is also seen from Table I that thermal diffusivity of *n*-type $10^{18}\,cm^{-2}$ Sn-doped InP is slightly larger than that of $10^{17}\,cm^{-2}$ Fe-doped semi-insulating InP. In semiconductors having carrier concentration below $10^{20}\,cm^{-3}$, phonon assisted heat diffusion dominates over heat diffusion by photoexcited carriers. Fe doping results in midgap energy levels, which are known to create a large lattice relaxation around them, which are effective scattering centres for phonons. We find that the deep Fe centre is much more effective in reducing the thermal conductivity than the shallow Sn donor.

Many studies on semiconductors using OPCs [4–7] show a minimum in the phase plot of PA signal. We also observe this minimum for pure InP. The nonlinear behaviour in phase data of the signal is attributed to bulk and surface recombination components. At higher chopping frequencies this contribution becomes more visible in comparison with the slower heat diffusion processes from pure thermal wave component. The phase plots we obtained here for the doped samples reveals that, in the experimental frequency range, it is the thermalisation component, which dominates over nonradiative recombination contribution to PA signal. In order to check the influence of photoexcited carriers to PA signal, we followed the same experimental procedure but at a higher power level (100 mW). But the results are identical (not shown). In order to investigate the influence of surface finishing on thermal diffusivity of doped samples, we have repeated the experimental procedure with roughened surface facing chopped optical radiation. But that also yielded the same values for thermal diffusivity indicating the absence of influence of photoexcited carriers in heat diffusion mechanism in doped samples of carrier concentration less than $10^{20}\,cm^{-3}$. If photoexcited carriers also contribute to heat diffusion mechanism, then its influence should be visible in the frequency range used here.

CONCLUSION

In conclusion, we studied the PA signal from pure InP and InP doped with tin and iron under heat transmission configuration using OPC. The thermal diffusivity is obtained for all the samples. It is seen that doping reduces the thermal diffusivity value. It is also seen from our investigation that nature of dopant influence the heat diffusion processes. The present investigation also confirms that for carrier concentration less than $10^{20}\,\mathrm{cm}^{-3}$, phonon assisted heat diffusion mechanism dominates over carrier assisted heat diffusion processes. The present investigation shows that under heat transmission configuration, the pure one-dimensional model of Rosencwaig and Gersho can be used to measure thermal diffusivity of semiconductors having moderately high thermal diffusivity values. Evidence has been obtained for heat diffusion by nonradiative recombination of photoexcited carriers in pure InP.

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