Determination of Absorption Coefficients and Thermal Diffusivity of Modulated Doped GaAlAs/GaAs Heterostructure by Photothermal Deflection Spectroscopy

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Abstract: We report in this study theoretical and experimental studies of photothermal deflection spectroscopy (PDS) for planar doped heterostructures AlGaAs/GaAs. The PDS spectra at T=300K are recorded by varying the wavelength of the excited radiation for different modulation frequencies. They show two different regions related to the fundamental electronic transitions from GaAs and Si doped AlGaAs. The DX levels have been observed approximately at 0.150 eV below the conduction band edge of Si doped Al0.33Ga0.67As (Eg=1.835 eV at 300 K). We give in this study a method of measuring the absorption coefficient of AlGaAs/GaAs heterostructures from the amplitude measurement of PDS signal. The optical absorption coefficient found was between 10 and 10^2 cm^{-1} in the energy range 1-2.5 eV for samples. The PDS data are compared with those given by conventional transmission technique. The evolution of PDS amplitude spectra by varying the modulation frequency from 5 to 100 Hz (usually called depth profile analysis) and using He-Ne laser probe beam shows that the PDS signal amplitude increases with decreasing the modulated frequency. A numerical procedure is applied to fit the measured amplitude of the PDS signal. The value of thermal diffusivity (D=0.27 cm^2 s^{-1}) shows a very good agreement with results given by spectroscopic ellipsometry.

Key words: Heterostructures, AlGaAs/GaAs, photothermal deflection spectroscopy

INTRODUCTION

Two dimensional GaAlAs/GaAs structures grown by molecular beam epitaxy (MBE) have been achieved using modulated doping techniques\(^1\). When a doped GaAlAs layer is grown on top of an undoped GaAs layer, a two dimensional electron gas (2DEG) can be formed at the interface\(^2\). This is due to the electron affinity difference between the two materials. The introduction of undoped “space layer” separates the electron in the channel and their parent donors. Thus it reduces considerably the coulomb effects between carriers and ionized impurities and produces an increase of the mobility of the 2DEG in the channel. The structure studied in this work is a two Si modulated doped GaAlAs/GaAs heterostructure elaborated according to the model proposed by Schubert et al.\(^3\).

In the present study we report results of photo thermal deflection spectroscopy (PDS)\(^4,5\). The aim of the PDS experiment is to determine the absorption coefficient of the thin layer structure in the visible spectral range. The absorption spectra show two maxima A (1.43 eV) and B (1.83 eV) respectively due to fundamental electronic transitions from GaAs and Ga\(_0.67\)Al\(_0.33\)As layers. A simplified one dimensional model similar to Rosencwaig and Gersho (R.G) theory\(^6\) is used for the theoretical analysis. It is solved for two layer transparent samples (\(\beta_1, l_1<1\) and thermally thin (\(\mu_1>l_1\)) it is possible to divide the PDS signal in two contributions. \(\beta_1\) and \(\beta_2\) are the absorption coefficient of the first layer and the second layers of the sample. The optical absorption coefficients of GaAs and Al\(_0.33\)Ga\(_0.67\) As compounds vary from 10 to 10^2 cm^{-1} in the energy range 1-2.5 eV. PDS measurements are compared with those obtained by conventional optical methods.

In this study we present also a method that enables one to evaluate the thermal diffusivity of the sample. This method is based on the analysis of both the measured amplitude of the PDS (recorded by varying the modulation frequency from 5 to 100Hz) and the theoretical data obtained by using an adequate model.

Theoretical model: Mandelis has presented a one dimensional theory of PDS\(^7\) using an analytical method similar to R.G theory\(^8\). As this theory is for thermally thick bulk samples, it is necessary to take into account the multiple reflection of the incident light in the film.

Figure 1 shows the one dimensional model used for the theoretical analysis. It is solved for three one dimensional layers of a high refractive medium, a thin-film sample and the backing material. The pumping beam is used to illuminate the sample at x=0 and –L in the Fig. 1.

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Fig. 1: Geometry of one-dimensional theoretical model for PDS

In a one dimensional geometry case, the expression of the PDS signal $S$ is given:

$$S = \frac{L}{n_0} \left( \frac{dT}{dz} \right) e^{i2\pi vt}$$  \hspace{1cm} (1)

where, $\frac{1}{n_0} \frac{dn}{dT}$ is the relative index change with temperature of the deflecting medium, $\frac{1}{n_s} \frac{dn}{dT}$ is larger for CCl$_4$ ($510^{-4}$ K$^{-1}$) than for air ($10^{-6}$ K$^{-1}$), $L$ the interaction length, $\nu$ the modulation frequency and $z_0$ the distance of the probe beam from the sample surface.

The temperature distribution in the deflecting medium $T_f(z)$ is complex and given by the equation (2):

$$T_f(z) = Q \exp(-\sigma z)$$  \hspace{1cm} (2)

where, $\sigma = (1+i)(2\pi v/2D)$ is the thermal diffusion length and $D$ the thermal diffusion constant. Here the index $I$ take the subscripts, $f$ and $b$ for the film sample, fluid and backing material respectively. $Q$ is the complex temperature rise above the average temperature on the sample surface and its expression is obtained by solving the one dimensional heat equation in the different environments sample, fluid and packing material and assuming a continuity of the temperature and the heat flux at the different interface. In the case of homogeneous optically absorbing sample and for a sample made of one layer the expression of $Q$ is given by:

$$Q = \frac{\alpha_1}{n_0} \left( \frac{d\bar{n}}{dT} \right) e^{-\alpha z}$$  \hspace{1cm} (3)

where, $\alpha$ is the optical absorption coefficient, $K$ the thermal conductivity and $R$ the refraction factor:

$$\alpha = \frac{K}{\bar{n} \mu}$$

$Q$ is a complex parameter depending on the optical and thermal proprieties of the sample.

The amplitude $A$ and the phase $\phi$ of the PDS signal are finally given:

$$A = \frac{\sqrt{2}}{n_0} \left( Q_f^2 + Q_i^2 \right)^{1/2} e^{\phi/2}$$  \hspace{1cm} (4)

$$\phi = \tan^{-1} \left( \frac{Q_i}{Q_f} \right) - \frac{z_0}{\mu} - \frac{\pi}{4}$$  \hspace{1cm} (5)

where, $Q_f$ and $Q_i$ are the real and imaginary part of $Q$ respectively.

Saadallah introduced a general form for the expression of the periodic temperature surface of a semi conducting multilayer sample, deposited on a substrate and heated using a modulated light beam. It showed a good agreement with the experimental data when the sum of the thickness of all layers is much smaller than the thickness of the substrate.

McGahan introduced an analytical expression for the surface temperature based on Green’s functional treatment of heat condition equation. In this work we used the expression of $T_f$ given by equation (4) of ref$^{[9]}$ in the many layer case, determined without any assumption on the value of the layer thickness. This expression was obtained by resolving the heat equation in the different media with suitable boundary conditions. It showed a very good agreement with data obtained using GaAs and AlGaAs/GaAs hetero structures. Table 1 lists the physical constants used in our calculations.

**Samples and experimental setup:** The samples used in this study are GaAs compound and three AlGaAs/GaAs modelled doped hetero structure grown by molecular beam epitaxy on semi-insulating GaAs (001) substrate and elaborated according to the model proposed by Schubert et al.$^{[13]}$. Heir structures are illustrated in Fig. 2. Two delta doping monolayer were incorporated. The first planar doping $\delta$Si ($N^{\delta}_s = 2 \times 10^{12}$ cm$^{-2}$) is introduced in a plateau (Sample $S_1$, $x=0.33$), in a narrow QW (Sample $S_2$, $x=0.24$), or in a barrier (Sample $S_3$, $x=0.41$) as it is shown in Fig. 3.
Table 1: Physical constants used in theoretical calculations

<table>
<thead>
<tr>
<th>Material</th>
<th>Thermal conductivity K (W/cm K)</th>
<th>Density ρ (g/cm³)</th>
<th>Specific heat C (J/g K)</th>
<th>Thermal diffusivity D=K/ρC (cm²/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs</td>
<td>0.55</td>
<td>5.36</td>
<td>0.33</td>
<td>0.31</td>
</tr>
<tr>
<td>AlₓGa₁₋ₓAs</td>
<td>0.55 - 2.12 x + 2.48x²</td>
<td>5.36 - 1.6 x</td>
<td>0.33 + 0.12x</td>
<td>0.31 - 1.23 x + 1.46x²</td>
</tr>
<tr>
<td>CCl₄</td>
<td>1.0310⁻³</td>
<td>1.59</td>
<td>0.67</td>
<td>7.4810⁻⁴</td>
</tr>
</tbody>
</table>

Fig. 2: Schematic cross section of sample

Fig. 3: The self consistent calculation of the conduction band structure, the levels energy and the corresponding wave functions, for samples S₁ (a), S₂ (b) and S₃ (c)

The second planar δ(Si) (N²Dₑ=10¹³ cm⁻²) is placed in AlₓGa₁₋ₓAs at 150Å from the cap layer in order to compensate the electrical charge needed for surface depletion.

The sample is heated using a 1000W halogen lamp placed behind the slot of a monochromator. Light coming out of the monochromator is chopped using a mechanical chopper. The optical absorption is generated in the sample a thermal wave which propagates until the CCl₄ fluid and creates a temperature gradient causing a refractive index gradient, hence a deviation of the probe laser beam.

Fig. 4: PDS spectra of 375µm GaAs compound

This deviation is measured by using a position photodetector attached to a look in – amplifier. The PDS spectra are normalized by to the flux of incident photons.

RESULTS AND DISCUSSION

The Fig. 4 shows the PDS amplitude spectra (curve S(E)) of a thick GaAs compound (375µm) used as a substrate for the elaboration of our heterostructures, at the modulated frequency f=13Hz. The thermal diffusion length µₛ for this sample GaAs is large compared to his thickness at this modulated frequency (µₛ = 871µm). So we considerate the sample as thermally thin. The PDS spectrum has been normalized by the saturated intensity just before the onset of a sudden decrease.

The phase information is obtained from both calculations and experiments. The phase spectra, however, indicate only a constant value because the sample is thermally thin. Therefore the α dependences of the phase shift contain little information.

The optical absorption coefficients α were derived from the PDS amplitude spectrum by comparison with the theoretical analysis.

For a GaAs sample of thickness L=375 µm and for the modulated frequency f=13Hz, we plot the theoretical PDS amplitude calculated from Eqs (4) and (5) of ref[9] (by setting l₁=l₂=0) as a function of the optical absorption coefficient α. The theoretical curve S(α) in Fig. 5 shows the dependence of the normalized amplitude of the PDS signal on the optical absorption coefficient. In the low- α range the deflection amplitude increases with increasing α; but in the large -α range the signal saturates to a constant value. This saturation occurs for optically opaque and thermally thin (µₛ>L>1/α) sample.
From the measured PDS spectrum we determine the absorption coefficient $\alpha_0 (E)$ by corresponding the theoretical normalized curve $S(\alpha)$ and the experimental curve $S(E)$ for each value of excitation energy $E$ (Fig. 5). The results obtained appear in Fig. 7 (curve $\alpha_c$), and they can be compared with that deduced from optical measures.

We report also in Fig. 7 the absorption curve $\alpha_c(E)$ deduced from PDS measure and experimental results obtained by Casey. From spectroscopic ellipsometry measurement.

We notice a good agreement between these spectra for $1.3\text{eV} < E < 1.45\text{eV}$. That shows the best choice of the model of calculation used.

It is possible to see that photo deflection spectroscopy gives the absorption spectra over the fundamental gap. To make a comparative study with classical optics it would be necessary to use much thinner samples, but it would be difficult to obtain sufficiently good optical polishes.
Table 2: Values of thermal diffusivity, thermal diffusion lengths calculated from the frequency \( f = 13 \)Hz and thickness \( d_i \) of each layers constituting the sample

<table>
<thead>
<tr>
<th>Layers</th>
<th>Thickness (( \mu m ))</th>
<th>Thermal diffusivity ( cm^2/s )</th>
<th>Thermal diffusion length (( \mu m ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capsule GaAs</td>
<td>0.050</td>
<td>0.31</td>
<td>871</td>
</tr>
<tr>
<td>( Al_{0.33}Ga_{0.67}As )</td>
<td>0.25</td>
<td>0.063</td>
<td>390</td>
</tr>
<tr>
<td>GaAs</td>
<td>375</td>
<td>0.31</td>
<td>871</td>
</tr>
</tbody>
</table>

We report on the Table 2 values of thermal diffusivity, thermal diffusion lengths calculated from the frequency \( f = 13 \)Hz and thickness \( d_i \) of each layer constituting the sample. The thermal diffusion lengths \( \mu_i = (D_i/\pi f)^{1/2} \) for use modulated frequency (Table 2) is large compared to the thickness of each layer. So we considerate the sample as thermally thin.

Yasuhiro et al.\(^{[16]}\) treated the multi-layer model in which the incident light was assumed to be entirely absorbed at the surface of the sample. The approximate formula of the PDS signal in this case indicates the possibility of investigating two optical absorption coefficients of the first and second layers of the sample. So the PDS signal is the sum of two contributions coming respectively from GaAs and Si doped \( Al_{0.33}Ga_{0.67}As \) layers:

\[
\mu = \frac{1}{2a_i} f_i (1 - \sum \alpha_i l_i) = s_1 + s_2
\]

To determine the absorption spectra of \( S_1 \), we adjust the corresponding PDS spectrum to the absorption spectrum of GaAs substrate. We obtain thus absorption spectra of the sample \( S_1 \) represented in Fig. 9.

Figure 10 shows the PDS amplitude spectra of the sample \( S_1 \) and for different modulated frequencies (13Hz, 53Hz and 93Hz). It is pointed out that the shift of the absorption curves is due to the difference in the thickness of thermal active layers at the sample which has an influence on the photo thermal deflection signal since \( \mu > L_s \).

The procedure for finding the thermal diffusivity coefficient of the sample is as follows. First the PDS spectrum of the sample was recorded by varying the modulated frequency from 10 to 100Hz. The excitation radiation, used was 632.8nm He-Ne. For this frequency range and this excitation energy where the sample is thermally thin (\( \sigma_s d << 1 \)) and optically opaque (\( \alpha > 1 \)) the equation (3) can be written as:

\[
A = \frac{L_s}{a_i} \frac{d}{dT} \frac{l_0 (1 - R_s)}{2 K_g (b + g)} \sqrt{D} \exp(-z_c \sqrt{\frac{\pi f}{D_i}})
\]

The simple form of \( A \) that correspond to the sample used in this work:

\[
A = 7.6910^{-7} \sqrt{D} \exp(-20.68z_c \sqrt{\pi f})
\]
As the relationship between the PDS amplitude and the modulated frequency we fit the experimental data. The best fit curve, is shown in Fig. 11. The good agreement between the experimental and theoretical results demonstrates the reliability of the adopted theoretical approach. From the experimental results we can deduce by comparison with the corresponding theoretical variation in Fig. 11. The value of $D_s$ ($D_s = 0.27 \text{ cm}^2/\text{s}$). This $D_s$ value is in good agreement with previous work \cite{17, 18}.

CONCLUSION

In this work, we have presented a method to determine quantitatively the optical absorption spectra of thin films using the high sensitive technique of photo thermal deflection spectroscopy. In addition the PDS measurement enables us to evaluate the thermal diffusivity of the sample.

REFERENCES