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Optical investigations on oxygen doped GaAs

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By using different optical techniques, the semiconducting oxygen doped GaAs crystals grown in our laboratory were investigated. The absorption coefficients of GaAs determined by laser calorimetry at 10.6 μm were found to be $(0.015 \pm 0.005)\mathrm{cm}^{-1}$ in good accordance with the values obtained from spectrophotometrical measurements obtained by scanning in a Fourier transform infrared (FTIR) Perkin Elmer spectrometer over the spectral range 2.5–20 μm.

1. Introduction

The ever growing interest to obtain optical quality materials available for laser optics has been accompanied by specific investigations for characterization of such materials.

Our primary emphasis here is on some methods of measuring the optical properties of the oxygen doped GaAs grown by LEC method. It is well known that the high resistivity single crystals of GaAs are obtained by compensation of either the more shallow acceptor levels with deep donors – oxygen, or of the more shallow donor levels by acceptors – Cr. The resistivity is by an order of magnitude higher in chromium compensated GaAs than in the case of oxygen doping [1]. In the oxygen doped GaAs, the concentration of free carriers induced by the causal impurities [2] is about $10^{12}-10^{14}$ cm^{-3}. The correlation of the optical properties with the electrical ones provides a useful rule of thumb to decide on the opportunity to use GaAs samples in laser optics. A comparative and critical overview of the investigation methods allows the evaluation of the material characteristics as well as the relative value of the methods and techniques used.

2. Experimental

Semi-insulating oxygen doped GaAs crystals are cut in plano-parallel wafers and then mechanically polished, the latter realized in a two step process: firstly, a rough lapping using a grinding pad, secondly, a fine polishing using 0.3 μm grain alumina.

The samples were optically polished, the parallelism being within 15–20 s of arc, the flatness measured with a Möller–Wedel interferometer was half a fringe at 632.8 nm and the roughness better than 100 Å rms. The samples were carefully cleaned before each measurement: 10 minutes of ultrasonic cleaning using unhydric ethylic alcohol, rinsing in unhydric ethylic alcohol flow and drying in high purity

nitrogen puff. Prolonged exposure to the atmosphere as well as to biological contamination were avoided by correctly handling the samples during the cleaning process and the measurement accomplishement.

The spectral characteristics were scanned in a FTIR Perkin Elmer 1760 X spectrometer over the spectral range of 2.5 – 20 μm. The FTIR Perkin Elmer 1760 X spectrometer is a single beam instrument. Special care was taken to make corrections due to the particular character of the investigated samples: high refractive index, thickness and polished faces which influence the optical path in the interferometer. Any IR disturbing source able to influence the accuracy of the measurements was also rigorously avoided, the sensitivity of the instrument being very high.

To determine the optical constants n, k for GaAs specimen, the spectrometrical data were processed according to a program for the evaluation of spectrophotometrical curves [3]. Commonly accepted theoretical expressions [4] for transmittance and reflectance were employed in the program. The optical constants are the solutions to the system of equations:

$$R(n, k, d, \lambda) - R_{exp}(\lambda) = 0,$$
$$T(n, k, d, \lambda) - T_{exp}(\lambda) = 0$$

where: n – refractive index,
k – extinction coefficient,
d – thickness of the sample,
λ – wavelength.

From the available dispersion formulas the one given by SELMEIER [5] was used to fit the values of the refractive index. The thickness of the samples was determined using a Microdigit electronic micrometer. The absorption losses including both the surface and bulk contributions were determined by laser calorimetry at 10.6 μm using a home made set-up containing a cw CO_2 laser of 40 W stabilized power. The increase of temperature was measured using termistors (IAP fabricated). The electrical resistivity and the Hall effect were evaluated by conventional method. The ohmic electrical contacts have been obtained by heating all faces of rectangular parallelipedic shape with indium spheres 0.8 – 1.0 m diameter in ultrapure graphite crucibles at 500°C in vacuum conditions (10^{-5} Tr). To minimize the photoconductive effect, which could be important, all the measurements were carried out in darkness.

3. Results

3.1. Spectrophotometrical measurements

All the specimens of GaAs were measured at normal incidence in the FTIR Perkin Elmer 1760 X spectrometer to register the transmittance and reflectance in the IR spectral range. Five GaAs samples provided by different crystal growing processes were investigated.

From the examination of Figure 1 (**a**, **b**), it is easy to notice:

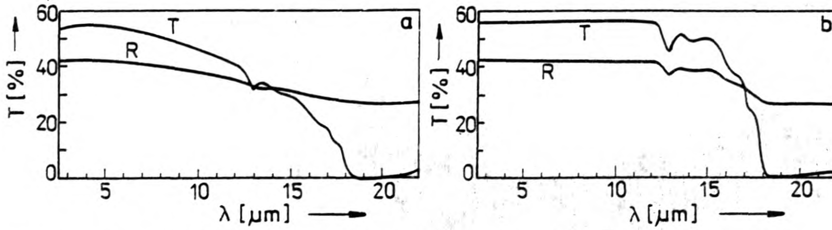

Fig. 1. Transmittance and reflectance curves (**a** — for the GaAs sample A, diameter 30 mm, thickness 4.27 mm, **b** — for the GaAs sample B, diameter 50 mm, thickness 5.21 mm). The transmission spectrum for sample GaAs (**a**) is specific for a low absorbing material, while for sample GaAs (**b**) the presence of a higher impurities level could be important

i) An undoubted difference between the bahaviour of sample A and B as far as the transmission is concerned.

ii) While in Fig. 1**b** the transmission curve has a plateau at 55% up to about 12 μm, in Fig. 1**a** there is no plateau but instead a decrease in the transmission curve is observed.

iii) An absorption band can be noticed at 12 μm.

iv) For sample B, a straight line limit should be observed at the transmittance window for long wavelengths, while there is no such case for sample A, the corresponding curve being different and not specific for a weakly absorbing GaAs material.

The accuracy of the transmittance measurements was $\Delta T = 0.3\%$.

To measure the reflectance characteristics of GaAs samples, a special reflectance attachment was used. A special, very stable and certified, mirror was used as a reference standard for the reflectance measurements. The accuracy of the reflectance measurements was less than for the transmittance $\Delta R = 0.6\%$.

The spectral characteristics and the thickness of the samples were the input data for the computer program, which allows computing the optical constants — refractive index and extinction coefficients — according to the procedure described in detail under the experimental section. The optical constants were computed at equidistant wavelength steps of the scanned IR spectral range (Fig. 2). Both the dispersion of the refractive index and the extinction coefficients are presented in the investigated IR spactral range. It is observed that:

i) For all the samples, the curves for the refractive index dispersion have an exponential behaviour.

ii) The effect of the absorption band at 12 μm determines the anomalous behaviour at this wavelength.

Reminding that the Sellmeier formula is valid outside the absorption band we consider the dispersion of the index within in the 2.5–11 μm range. As can be observed in Figure 2, the index for the sample B varies from 3.32 at 2.5 μm to 3.27 at 11 μm. The precision of the determination is about 0.02 for the refractive index and 5×10^{-6} for the extinction coefficient.

In Figure 3, the extinction coefficients for the samples A and B have the same shape but the extinction coefficients for the sample B are much lower than for

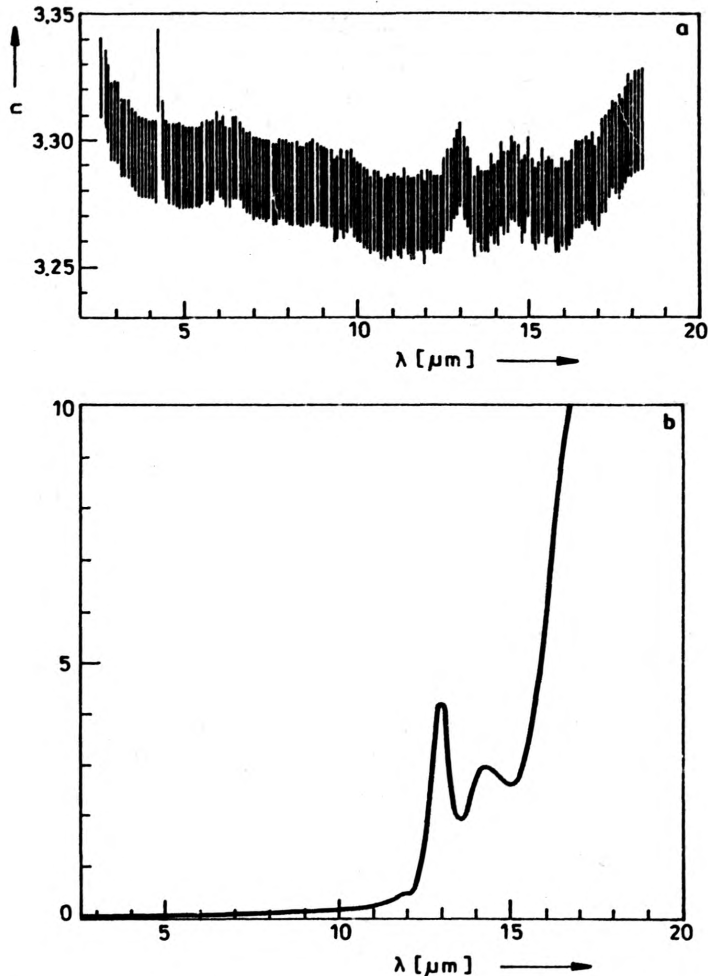

Fig. 2. Dispersion of the optical constants for the GaAs sample B, determined by the computer processing of the spectrophotometrical curves scanned in a FTIR Perkin Elmer 1760X spectrometer. a – refractive index, b – extinction coefficient

sample A. The extinction coefficients for all the samples were evaluated according to the formula:

$$\alpha = \frac{4\pi k}{\lambda}$$

where: α – absorption coefficient,
k – extinction coefficient.

In the Table, the geometrical and the spectrophotometrical characteristics of the investigated samples as well as the optical constants computed at the wavelength 10.6 μm are presented for comparison with the laser calorimetry investigations.

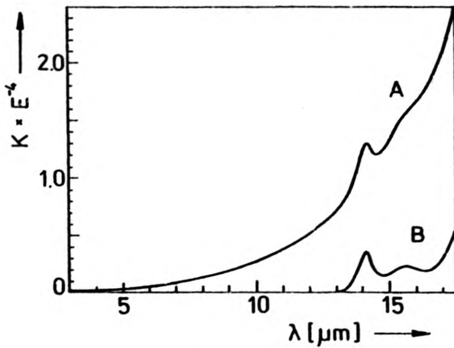

Fig. 3. Dispersion of the extinction coefficients for the GaAs sample A (curve A), and the GaAs sample B (curve B)

Table

Sample	*d* [mm]	*r* [Ω cm]	*T* [%]	*R* [%]	*n*	*k* [$\times 10^{-6}$]	α [cm^{-1}]	α_c [cm]
GaAs A ø 30	4.27	3.3×10^{-1}	43.8	38.4	3.27	42	0.498	0.52
GaAs B ø 50	5.21	3.6×10^{5}	55.7	43.9	3.27	0.7	0.008	0.012
GaAs M ø 40	5.36	3.7×10^{5}	55.8	43.7	3.26	1.0	0.012	0.014
GaAs N ø 50	4.86	2.9×10^{5}	56	43.9	3.26	0.8	0.009	0.010
GaAs P ø 50	4.225	3.2×10^{5}	55.9	44	3.27	0.6	0.007	0.012

d — thickness, *r* — resistivity.

3.2. Laser calorimetry investigations

Thermometrical laser calorimetry was adopted to determine the absorption coefficient. The investigations were carried out using a home-made set-up as is shown in Fig. 4. A cw CO_2 laser delivering 40 W stabilized power was used as a heating source. The temperature on the rim of the sample was appreciated using termistors

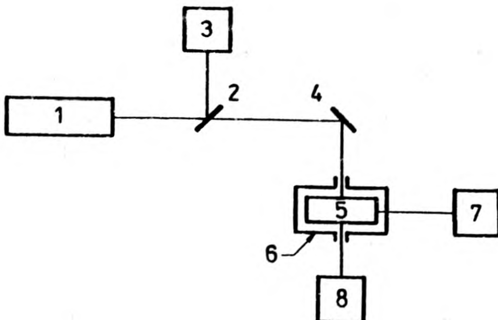

Fig. 4. Experimental set-up for laser calorimetry investigations: 1 — CO_2 cw laser, 2 — ZnSe beam splitter, 3 — power meter, 4 — deflecting mirror, 5 — GaAs sample, 6 — adiabatic enclosure, 7 — thermistor, 8 — power meter for the transmitted power

(4%/°C sensitivity). All the samples were investigated under similar conditions.

The bulk absorption coefficient was computed using the calorimetric expression

$$\alpha d = \frac{mC_p}{P_t}\left(\frac{\Delta T_h}{\Delta t_h} + \frac{\Delta T_c}{\Delta t_c}\right)$$

where: α — absorption coefficient including both bulk and surface losses,
d — thickness of the sample,
P_t — laser power transmitted through the sample,
C_p — specific heat of the material,
m — mass,
$\left(\frac{\Delta T_h}{\Delta t_p} + \frac{\Delta T_c}{\Delta t_c}\right)$ represents the sum of the heating and the cooling rate.

For C_p we used the value 850 J/kgK [6].

In the Table, in the last column, the results for the absorption coefficient calorimetrically measured are presented. The accuracy of the calorimetrical measurements is 16%.

3.3. Hall effect and resistivity measurements

The samples of parallelipedic shape provided by different crystal growth processes were measured to estimate the resistivity.

4. Discussions and conclusions

We investigated five GaAs samples obtained by LEC method in different crystal growing processes. The sample GaAs A was intentionally chosen to be reported for the needs of discussions.

Figure 1b showing the transmittance curve of the GaAs sample B is very close to that predicted for low absorbing GaAs material [7], while the behaviour visible in Fig. 1a could be the result of many factors such as: purity of the raw materials, liquid encapsulant, inert atmosphere, nature of the crucibles, crystal homogeneity.

The optical homogeneity of GaAs samples was reported elsewhere [8]. The results of our measurements are collected in the Table. The optical constants for LEC grown GaAs are in accordance with the reported values [9], and also when considering the lower resistivity of oxygen doped material, we have investigated. Comparing the transmission curves for GaAs samples A and B, the shape difference is obvious. The lower transmission level means a higher absorption, also put into evidence by laser calorimetry.

Since GaAs is a semiconductor-like material, the higher absorption is due to the presence of free carriers — the main responsible for the optical absorption in such materials — producing a poor insulating character and thus a lower value of the resistivity. This is the case for GaAs sample A, chosen for comparison. For all the other investigated B, M, N, P the reported values show a good reproducibility. The absorption coefficients were determined for GaAs samples by two methods: a com-

puter conducted spectrophotometry, and the laser calorimetry, which is more specific for infrared materials. A remarkable accordance was observed between the results obtained for the absorption coefficients, by using both the above mentioned methods.

We believe that a bulk absorption coefficient of 0.015 ± 0.005 cm^{-1} for oxygen doped GaAs investigated samples makes this material suitable for use as substrates in 10.6 μm laser optics for CO_2 industrial lasers [10].

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