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PHOTOACOUSTICS,
NEW METHODS TO STUDY OPTICAL PROPERTIES OF DISLOCATIONS ?

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Résumé - Les principes des méthodes dites photoacoustiques sont brièvement rappelés et les trois approches expérimentales principales sont décrites : la cellule close et le microphone ; le transducteur piézoélectrique en cellule ouverte ; la détection par effet mirage de la déflexion photothermique.

Ces méthodes étant sensibles aux transitions non-radiatives sont complémentaires de la photoluminescence. L'avantage principal de la photoacoustique par rapport à l'absorption classique est une sensibilité importante due à une détection sur "fond noir" tout comme en photoluminescence. Par exemple des valeurs d'absorbance de 10^{-6} cm^{-1} ont pu être obtenues pour des transitions purement non radiatives.

En plus de la spectroscopie, les méthodes photoacoustiques permettent aussi la topographie avec des possibilités d'analyse en profondeur.

Bien que l'application de ces méthodes aux propriétés optiques des dislocations soit encore dans l'enfance, les quelques premiers résultats prometteurs de la littérature seront présentés.

Abstract - The principles of the so-called photoacoustic methods are briefly described and the three main types of experimental approaches are reviewed : microphone and closed cell ; piezoelectric transducer and open cell ; mirage effect detection of photothermal deflection.

Such methods being sensitive to non-radiative transitions are complementary to photoluminescence. The main advantage of photoacoustics over regular absorption is a good sensitivity due to detection over a "black background" just as in photoluminescence excitation. For instance, absorbance values as small as 10^{-6} cm^{-1} have been detected for non-radiative centers.

Besides spectroscopy, photoacoustic methods can also be used for topography with depth profiling capabilities.

Though the applications of these methods to optical properties of dislocations is still in its infancy, the first few promising results in the literature shall be reviewed.

I. INTRODUCTION -

New interest has been in recent time devoted to optical properties of dislocations in semiconductors and more particularly in III-V compounds [1] [2] [3]. This is because of their believed detrimental influence on light emitting device performances. However there is still a lack of understanding about the important question of the non-radiative and radiative transitions induced by dislocations [1] [2] .

As a complement to the "classical" methods of photo-(cathodo)luminescence and optical absorption, we propose here to apply photoacoustic methods to the study of optical properties of dislocation. We review why these new methods could be use to investigate the deep electronic levels in the band gap which may act as non-radiative recombination centers.

Principles involved in photoacoustic methods are essentially based upon the detection

of non radiative transitions by mean of their thermal effects. As summarized on Fig.1 optical properties of dislocations are classically detected through a photon or electron excitation by either luminescence or absorption. In photoacoustic methods, either the thermal waves or the resulting elastic waves are detected.

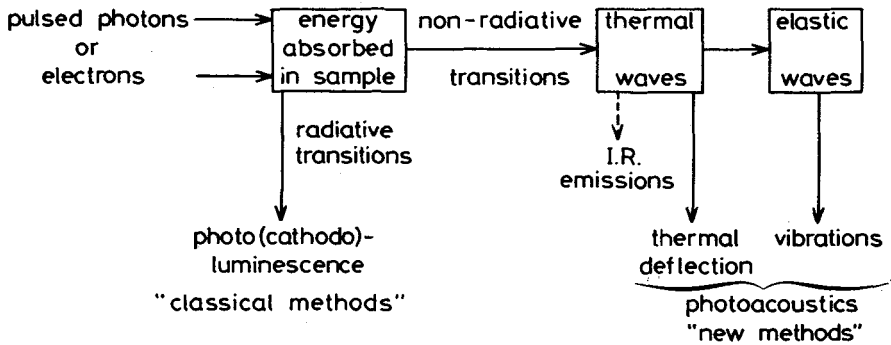


Fig. 1

In the following, we shall review briefly the three more usual photoacoustic methods where are respectively detected: elastic waves by microphone or piezo-detection and thermal waves by thermal deflection (mirage effect).

II. VARIOUS DETECTION TECHNIQS

II.1 Closed cell and microphone

A modulated monochromatic light beam at wavelength λ excites a sample inside a closed cavity filled with a gas. When excitation gives rise to non-radiative transitions, energy is transformed into modulated heat by the sample which in turn heats the gas. Pressure variations are then detected by a microphone coupled to the cavity by a hole. A synchronous detector analyses the signal.

A one-dimension piston model has been established by Rosencwaig and Gersho (R. G) [4] This theory gives the spatial distribution of the time dependent temperature within the gas layer near the sample surface, as shown on Fig. 2. As clearly seen, thermal

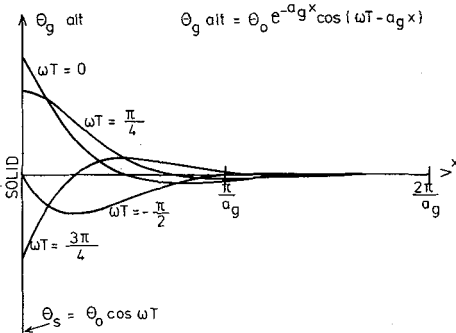


Fig. 2 - Spatial distribution of heat in R.G. model.

variations θ_{alt} . are confined within a distance $\approx 2\pi/a_g$ from the surface sample;

where $a_i = (\omega\rho_i C_i / 2k_i)^{1/2}$, with ρ_i , C_i , k_i , are respectively the density, specific heat and thermal conductivity of medium i . $\mu_i = a_i^{-1}$ is the thermal diffusion length of medium i : ($i=g$ for gas ; $=s$ for sample ; $=b$ for backing of sample). Thermal waves do not propagate at distances, whereas acoustic waves, created near the sample surface by periodic dilatation of gas, do.

The R.G. theory allows one to distinguish 6 different cases for the periodic signal (Q) in the closed cavity. First we consider two categories according to optical absorbance distance β^{-1} with respect to sample size l .

1) Optically transparent samples ($\mu_{\beta} (= \beta^{-1}) > \ell$)a) for thermally thin samples with $\mu_s \gg \ell$ and $\mu_s > \mu_{\beta}$ orb) for $\mu_s > \mu_{\beta}$ and $\mu_s < \mu_{\beta}$, pressure signal is given by :

$$Q \approx \frac{(1-j)}{2a_g} \beta \ell \left(\frac{\mu_b}{k_b} \right) Y$$

with Y proportional to $I_0 \eta / \ell_g$ where I_0 is excitation intensity, η the quantum efficiency for non-radiative transitions and ℓ_g the gas length.

c) for thermally thick samples ($\mu_s > \ell$) and $\mu_s < \mu_{\beta}$

$$Q \approx \frac{-j}{2a_g} \beta \mu_s \left(\frac{\mu_s}{k_s} \right) Y$$

2) Optically opaque samplea) for thermally thin samples ($\mu_s \gg \ell$) and $\mu_s \gg \mu_{\beta}$:

$$Q \approx \frac{(1-j)}{2a_g} \left(\frac{\mu_b}{k_b} \right) Y$$

b) for thermally thick samples ($\mu_s < \ell$) and $\mu_s > \mu_{\beta}$:

$$Q \approx \frac{(1-j)}{2a_g} \left(\frac{\mu_s}{k_s} \right) Y$$

c) for thermally thick samples ($\mu_s \ll \ell$) and $\mu_s < \mu_{\beta}$:

$$Q \approx \frac{-j}{2a_g} \beta \mu_s \left(\frac{\mu_s}{k_s} \right) Y$$

As seen from these relations, information about β is obtained in all cases but 2a and 2b which correspond to signal saturation.

II.2 Open cell and piezotransducer

At variance with the above described technic, vibrations created by thermal waves inside sample are directly detected by a piezotransducer bounded to the sample ; outside gas does not play a role any longer.

Theory of this technic has been investigated by Jakson and Amer [5] . They have discussed three cases :

1) Optically thick, thermally thick sample

$$(\mu_s \ll \ell ; \mu_{\beta} \ll \ell)$$

voltage signal at transducer output is then :

$$V \approx \frac{2MI_0 a_s}{j\omega \ell \rho_s C_s}$$

M characterizes the transducer ; as is linear expansion of sample.

2) Optically thick, thermally thin sample

$$(\mu_{\beta} \ll \ell \ll \mu_s)$$

$$V \approx \frac{MI_0 a_s}{j\omega \ell \rho_s C_s}$$

3) Optically thin, thermally thick sample

$$(\mu_s \ll \ell \ll \mu_{\beta})$$

$$v \approx \frac{MI_0 a_s}{j\omega \ell \rho_s C_s} \left\{ 1 - \exp(-\beta \ell) - \frac{6}{\ell \beta} \left[\left(1 - \frac{\ell \beta}{2} \right) \left(1 + \frac{\ell \beta}{2} \right) \right] \right\}$$

for small β then $v \propto \beta$

cases 1) and 2) indicate saturation of signal.

II.3 Photothermal deflection ("mirage effect")

This method, proposed by Boccara et al. [6], makes use of a probe beam (Ne-He laser) to detect thermal waves in the gas layer of thickness $2\pi/a_g$ (Fig.2). The modulated thermal gradient deflects the probe beam by the corresponding refraction index gradient ("mirage effect"). A position sensor gives a signal proportional to the deflection angle ϕ :

$$\phi = d \frac{dn}{dn} \left(\frac{dn}{dT} \right) \left(\frac{dT}{dx} \right)$$

where d is the interaction length between excitation beam at wave length λ and probe beam ; x is the coordinate normal to sample surface ; (dT/dx) is given by R.G. theory. So the thermal deflection leads to same indications as the microphone technic with often however a better sensitivity. A more complete theory of this technic has been given in [7] .

III. APPLICATIONS TO OPTICAL PROPERTIES OF DISLOCATIONS STUDIES

III.1. Orders of magnitude and features of photoacoustic spectroscopy (P.A.S.).

The typical features of PAS are the following :

- PAS provides essentially non-radiative transitions spectra and as such gives the negative image of photoluminescence excitation spectra. As such it is a necessary complement to photoluminescence, since in absence of luminescence it can also solve the dilemma : it is due to a lack of electronic levels or to a poor radiative quantum efficiency ? [1]

- Sensitivity is higher than for regular absorption because signal is obtained on "black background" and is directly proportional to excitation. Typical orders of magnitude are the following [7] :

P.A.S. technic	Sensitivity $\beta \lambda \times I_0 (W)$
microphone	$10^{-4} - 10^{-5}$
PZT	10^{-5}
mirage effect (transverse)	10^{-5}
(colinear)	$10^{-7} - 10^{-8}$

practically $\beta \approx 10^{-6} \text{ cm}^{-1}$ can be detected in solids by a photon excitation of 1 W.

- Due to the important role of $\mu_i = (2k_i/\omega \lambda_i C_i)^{1/2}$ in experimental conditions, a depth-profiling capability is obtained simply by varying ω .

Typically : $\mu_i = 0.65 \text{ mm}$ at 20 Hz in GaAs (65μ at 2 KHz) ; $\mu_i = 56 \mu$ at 2 KHz in air.

- As seen later in examples, topography with photoacoustics has been obtained by scanning the excitation source (either photons or electrons) over the sample surface.

III.2. Optical features of dislocations in semi-conductors by "classical" methods.

In order to verify the adequacy of the "new methods" to an eventual study of dislocations, their typical optical features have to be briefly reviewed.

In absorption, the dislocation induced variation in Ge has been found to be $\approx 10 \text{ cm}^{-1}$ at 0.1 eV [8] at a dislocation density level of 6.10^9 cm^{-2} . Appearance of induced absorption needs a high dopant concentration ($\geq 10^{15} \text{ cm}^{-3}$).

Another value $\beta \lambda \approx 0.24$ has been obtained for a density of 2.10^8 cm^{-2} at a concentration of $2.10^{16} \text{ cm}^{-3}$ [9] .

In emission, at a dislocation, a contrast of a much as 0.5 due to a proportionally

reduced radiative quantum efficiency has been found on GaAs by luminescence topography (spatial resolution $\approx 3 \mu\text{m}$) [2].

All those features ($\beta\lambda$, $\Delta n/n$, spatial resolution) are well within capabilities of photoacoustic technics when a laser excitation is used and a better sensitivity could be expected.

IV. SOME EXAMPLES IN PHOTOACOUSTICS

1) Spectroscopy by mirage effect

Fig. 3 - P.A.S. spectra by mirage effect for different InP substrates.

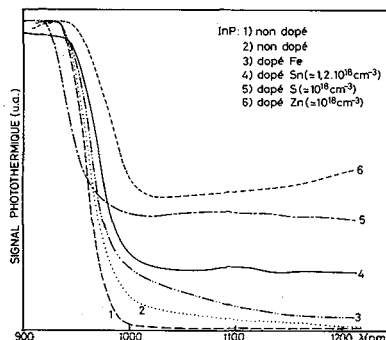


Fig. 3 presents a result we have obtained at CNET by photothermal deflection on various InP substrates. The PAS signal is normalized for excitation above band gap where saturation occurs :

$$\phi = K(1 - e^{-\beta\lambda}) = \phi_{\text{sat}} \cdot (1 - e^{-\beta\lambda})$$

and $\beta\lambda = -\ln [1 - (\phi/\phi_{\text{sat}})]$ which permits to reach $\beta\lambda$.

Though sample 5 has a low dislocation density ($< 10^{-3} \text{cm}^{-2}$) and other samples have higher density, variations in spectra indicate the method to be sensitive to doping concentration. However such result is not yet conclusive because dislocation density here are smaller than those usually considered for absorption studies and such absorption induced by dislocations are known to be concentration dependent [10].

2) Depth profiling and Topography by microphone

Results of Busse [11] show clearly the depth profiling ability of photoacoustics by detecting cavity holes below sample surface at different depth. A spatial resolution of $6 \mu\text{m}$ is typically obtained on topography of an integrated circuit with subsurface defect image detection [12].

3) Depth profiling and topography by piezotransducer and electron excitation (electroacoustic).

Results from Cargill [13] in electron excitation and piezodetection at high frequency ($\omega/2\pi = 6\text{MHz}$) show that thin layer analysis is possible with spatial resolution of $\approx 2 \mu\text{m}$.

4) A first result on dislocations by PAS with piezotransducer

To the best of the author's knowledge, the first proof of the ability of PAS to detect optical spectra of dislocation has been given in a work by Wasa et al. [14] where the piezodetected PAS signal is clearly strengthened when dislocation density of a GaAs sample is increased from 10^4cm^{-2} to 10^5cm^{-2} . On InP a curious result is obtained : contrary to our result of Fig. 3, going from a 10^5cm^{-2} to a 10^3cm^{-2} dislocation density seems to reduce completely non-radiative transitions, i.e. the band gap cannot even be detected.

V. CONCLUSION

Sensitivity, complementarity with luminescence, depth-profiling, spatial resolution in topography make photoacoustics promising methods to solve some of the problems of dislocations in III-V compounds :

- existence of deep levels induced by dislocations.
- mechanism for dislocation induced radiative and non-radiative transitions.

However, in order to have simultaneously all the advantages of photoacoustics a prerequisite is in our view the availability of tunable lasers in the gap range of the considered materials.

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