Classification

Physics Abstracts

42.65G — 72.20J — 72.80E — 78.50G

Picosecond investigation of photorefractive and free carrier gratings in GaAs : EL2 and CdTe : V

Philippe Delaye ⁽¹⁾, Kestutis Jarasiunas ^(1, *), Jean-Claude Launay ⁽²⁾ and Grald Roosen ⁽¹⁾

⁽¹⁾ Institut d'Optique Thorique et Applique, URA 14 CNRS, Centre Scientifique d'Orsay, B.P. 147, 91403 Orsay Cedex, France

⁽²⁾ Ple de Recherche Aquitain pour les Matriaux dans l'Espace (PRAME), B.P. 11, 33165 Saint Mdard en Jalles Cedex, France

(Received 30 November 1992, revised 18 January 1993, accepted 9 February 1993)

Abstract. — Laser induced picosecond transient gratings are used to study carrier transport *via* free carrier and photorefractive nonlinearities in semi-insulating undoped GaAs and vanadium doped CdTe crystals. Absorption and carrier generation mechanisms are investigated through the variation of grating strengths with input fluence. We show that grating decays are governed by mostly ambipolar diffusion and by its variation with fluence. Ambipolar mobilities are measured : $\mu_a \approx 135 \text{ cm}^2 \cdot \text{V}^{-1} \text{ s}^{-1}$ in CdTe and $\mu_a \approx 760 \text{ cm}^2 \cdot \text{V}^{-1} \text{ s}^{-1}$ in GaAs.

Introduction.

Semi-insulating photorefractive semiconductors reveal optical nonlinearities from different origins under excitation by short laser pulses. A significant effort is currently done towards the understanding of the origin of photorefractive and non-photorefractive nonlinearities in GaAs and both undoped and vanadium doped CdTe [1-10]. A detail analysis of carrier transport in the space charge field usually created in photorefractive media would allow to optimize crystal parameters for different applications in optoelectronics, holography and microelectronics.

Light diffraction on transient gratings has proved to be a simple but powerful tool for carrier transport studies at high excitations [11-14].

1. Experimental arrangement.

We present carrier transport properties of vanadium doped CdTe and semi-insulating GaAs analyzed with one of the most common configurations of transient grating techniques i.e.

(*) On leave from Material Research Laboratory « INTEST », Semiconductor Physics Department, Vilnius University, Sauletekio all.9, Bld.3, 2054 Vilnius Lithuania.

degenerate 4-wave mixing. We use a YAG laser, emitting at wavelength $1.06\text{ }\mu\text{m}$, with pulse duration $\tau_L = 28\text{ ps}$ at FWHM and output energy up to 10 mJ.cm^{-2} (Fig. 1). Two s-polarized beams of equal intensities record a grating with period $\Lambda = 1.8\text{ }\mu\text{m}$. The samples were cut with faces along crystallographic directions (110), (-110) , and (001).

Fig. 1. — Experimental set-up : G. P. : Glan Polarizer, $\lambda/2$: Half wave plate, D : Detector.

The semi-insulating CdTe crystal ($\rho = 5 \times 10^8\text{ }\Omega\text{.cm}$) was grown by using the modified Bridgman technique and vanadium doped to produce deep donor level at $\approx E_c - 0.75\text{ eV}$. The absorption coefficient of the 5 mm thick sample is measured equal to 2 cm^{-1} at $1.06\text{ }\mu\text{m}$. Growth, spectroscopic and CW photorefractive investigation of this crystal are presented in reference [9]; an effective trap density $N_{\text{eff}} = N_0 N_+ / (N_0 + N_+) \approx 4.3 \times 10^{15}\text{ cm}^{-3}$ was measured (N_0 and N_+ are the concentrations of the neutral and ionized deep level respectively).

The GaAs sample was a nominally undoped semi-insulating GaAs crystal, grown by the liquid encapsulated Czochralski (LEC) technique [15]. The absorption coefficient of the 1.5 mm thick sample is measured equal to 1 cm^{-1} at $1.06\text{ }\mu\text{m}$. It is well known that the dominant defect in undoped GaAs materials is an intrinsic deep donor level EL2 that alone fully determines the absorption at $1.06\text{ }\mu\text{m}$ [16, 17]. In this way we estimate the deep donor concentration in our sample as $N_{\text{EL2}} = 1.5 \times 10^{16}\text{ cm}^{-3}$. This is a typical value for LEC-grown GaAs crystals [18]. The concentration of the ionized part of EL2 in this sample was estimated through photorefractive measurements we conducted in parallel to be $N_{\text{EL2}^+} \approx 3 \times 10^{15}\text{ cm}^{-3}$.

Contribution of a photorefractive (PR) nonlinearity was separated from a coexisting free carrier (FC) non linearity by monitoring the polarization states of transmitted probe beam and of diffracted one [5, 10, 19]. The cases when pure PR grating (i.e. p-diffracted component of s-polarized probe beam for grating vector $\mathbf{k} \parallel (110)$) or pure FC grating (i.e. p-diffracted component of p-polarized probe beam at $\mathbf{k} \parallel (110)$ and at $\mathbf{k} \parallel (001)$) were analyzed experimen-

tally. A $\lambda/2$ plate was used for selecting the probe beam polarization (p or s) (Fig. 1). A Glan polarizer, oriented to reflect p-polarization, was used for separation of the Bragg-diffracted beam counterpropagating to one of recording s-polarized beams. Then the diffracted beam was directed to a fast silicon photodiode through a second p-transmitting polarizer. Such a set-up allowed us to realize a noise-free interferometer able to detect diffracted signals as low as 10^{-6} - 10^{-7} . Diffracted beam intensity $\langle I_1 \rangle$ or grating efficiency $\eta = \langle I_1 \rangle / \langle I_T \rangle$ (here $\langle I_1 \rangle$ and $\langle I_T \rangle$ are time-integrated values of diffracted and transmitted probe beams) as a function of excitation level I and of probe beam delay time Δt were studied. Sample transmittivity kinetics $\langle I_T \rangle = f(\Delta t)$ were also measured at different excitation levels or probe beam polarizations in order to analyze light absorption mechanisms and their peculiarities.

2. Exposure characteristics.

The exposure characteristic (EC) of a semiconductor (i.e. dependence of the diffracted beam intensity $\langle I_1 \rangle$ versus excitation energy I) contains the main information about carrier generation processes. If light induced changes of optical properties are not too large (diffraction efficiency $\eta < 0.01$), EC follows a power law dependence, $\langle I_1 \rangle = I^\gamma$. Assuming that carrier concentration increases with excitation as $N = I^\beta$, we have in log-log scale the following relationship between slopes γ and β [20, 11]:

$$\langle I_1 \rangle = \langle I_T \rangle (\pi \Delta n d / \lambda)^2 = I_p T (\pi n_{eh} \Delta N d / \lambda)^2 = A I^{2\beta+1} = A I^\gamma \quad (1)$$

where ΔN is the spatially modulated FC concentration, n_{eh} is the refractive index change due to one electron-hole pair according to Drude model [21], d and T are respectively the thickness and the linear transmission of the sample, and I_p is the probe beam intensity. In this way the carrier generation rate parameter β can be easily obtained.

In our CdTe sample, γ -values reveal three excitation regions corresponding to different light absorption mechanisms. For FC gratings (Fig. 2, curve FC), we observe $\gamma = 4$ at low fluences

Fig. 2. — The dependence of diffracted probe beam intensity $\langle I_1 \rangle$ and diffraction efficiency η vs. recording energy E for free carrier (FC) and photorefractive (PR) gratings (the value is taken at the maximum of the signal at delay time $\Delta t = 26$ ps). CdTe crystal orientation is (110) along grating vector $\mathbf{k}$.

indicating carrier generation from deep levels altered by two-step transitions *via* impurity states (i.e. electron and hole generation *via* deep level). At medium fluences, two-step and two-photon absorption are preminent ($\gamma = 5$) and, finally, the saturation of diffracted beam intensity is observed at high excitations. Photorefractive nonlinearity (Fig. 2, curve PR), due to its lower sensitivity, reveals two-step and two-photon carrier generation ($\gamma = 5$) at lower excitations followed by a decrease of the slope from value $\gamma = 5$ to $\gamma = 3$ at higher excitations. For FC signal the saturation of diffraction intensity or tendency to it was found typical of non homogeneous amplitude-phase gratings with non-photorefractive nonlinearities [3, 20]. Tests of sample transmittivity vs. excitation have shown absorption bleaching at low fluences, then its weakening and a subsequent sublinear region of transmittivity what confirmed the observed processes of carrier generation. At an excitation level of 4 mJ.cm^{-2} , diffraction efficiencies $\eta = 2 \times 10^{-4}$ and 7×10^{-6} were reached for FC and PR gratings respectively. It corresponds to induced refractive index modulation of $\Delta n_{\text{FC}} = 1.5 \times 10^{-6}$ and $\Delta n_{\text{PR}} = 3 \times 10^{-7}$, calculated with an effective thickness $d = (1 - \exp(-\alpha d))/\alpha$. We then deduce FC concentration $\Delta N \approx 6 \times 10^{14} \text{ cm}^{-3}$ and space charge field $E_{\text{sc}} \approx 50 \text{ V.cm}^{-1}$. At this fluence of 4 mJ.cm^{-2} , the PR signal is far from saturation and the corresponding space charge field is about one order of magnitude smaller than the diffusion field $E_d = k k_B T/e \approx 870 \text{ V.cm}^{-1}$.

In our semi-insulating GaAs undoped sample, we observed an EC slope with value $\gamma_1 = 3.9$ at excitation levels below 4 mJ.cm^{-2} for both FC and PR gratings (Curves 1, Figs. 3 and 4). From the experimental value of γ_1 we estimated the parameter of carrier generation rate $\beta_1 \approx 1.5$ according to relationship given by equation (1). This value of β_1 is determined by linear electron generation from deep EL2 traps which is altered due to nonlinear mechanisms of bipolar carrier plasma creation (i.e. by two-step transitions *via* ionized EL2 traps and by two-photon absorption). Diffraction efficiencies $\eta = \langle I_1 \rangle / \langle I_T \rangle = 4 \times 10^{-3}$ and 4×10^{-5} for FC and PR gratings respectively were reached at excitation level 4 mJ.cm^{-2} . The correspon-

Fig. 3. — Exposure characteristics of light diffraction on free carrier gratings (1) and of sample transmittivity (2). The p-polarized probe beam delay time is $\Delta t = 26 \text{ ps}$; GaAs crystal orientation is (110) along $\mathbf{k}$.

Fig. 4. — Exposure characteristics of light diffraction on photorefractive gratings (1) and of sample transmittivity (2). The s-polarized probe beam delay time is $\Delta t = 26$ ps and the GaAs sample orientation corresponds to (110) along $\mathbf{k}$.

ding values of light induced refractive index modulations were thus estimated: $\Delta n_{FC} = 2 \times 10^{-5}$ and $\Delta n_{PR} = 2 \times 10^{-6}$ (at the end of excitation pulse, when $\Delta t = \tau_L$). This is about one order of magnitude larger than in CdTe. Assuming that the refractive index changes are due to FC concentration ΔN and space charge field E_{sc} , the following absolute values of $\Delta N \approx 6 \times 10^{15} \text{ cm}^{-3}$ and $E_{sc} \approx 650 \text{ V.cm}^{-1}$ were calculated and used in the further analysis of results in GaAs.

At excitation levels above 4 mJ.cm^{-2} , for FC gratings a strong decrease of EC slope was observed (Fig. 3 curve 1, $\gamma_2 = 1.7$), and corresponding value of sublinear carrier generation rate $\beta_2 = 0.5$ was estimated. At these excitation levels, PR grating also exhibits a slowing in space charge field creation with excitation (Fig. 4 curve 1). In addition, exposure characteristics of sample transmittivity (curves 2 in Figs. 3, 4) revealed an increasing light induced absorption with excitation. Many reasons can explain this saturation [22]. The most probable is that induced absorption decreases the energy I of the pump beam leading to a less efficient carrier generation that reduces β . The diffracted signal is consequently reduced in a complex way compared to equation (1) which was established assuming a linear transmission of the different beams [13].

Time resolved measurements of induced absorption have been carried out to investigate its origin. We observed a time-resolved structure of sample induced absorption. The main characteristic is a strong darkening with at least two components (Fig. 5). The first component of darkening evolves as the autocorrelation of the excitation pulse [23] and rises to the value $\Delta\alpha = 2.5 \text{ cm}^{-1}$, i.e. giving more than a 3 times increase of the absorption. Then the next competing component is arising, which reaches $\Delta\alpha \approx 1 \text{ cm}^{-1}$ and relaxes with long time constant. Numerical simulations show that the strong peak is caused by two-photon absorption and the long tail corresponds to free carrier absorption. These features of induced absorption correspond to what has been observed in previous works [24, 25].

Summarizing this experimental part, we conclude that the simultaneous study of light diffraction exposure characteristics for FC and PR gratings, is a good tool in order to have a

Fig. 5. — Dynamics of light induced change in absorption of the GaAs sample for a p-polarized probe beam and for a s-polarized pump beam at excitation fluence of 5 mJ.cm^{-2}

better understanding of the carrier emission processes in semiconductors. From diffraction efficiency measurements, we deduce the free carrier population modulation which is quite strong even at moderate excitation levels like 4 mJ.cm^{-2} . This high free carrier population will determine the kinetics of the grating relaxation as we will discuss in the following part.

3. Time-resolved grating decay.

3.1 THEORETICAL APPROACH. — In order to find the time constants which govern decay times of FC and PR gratings, we solve the well known set of basic equations. To get insights, simple analytic equations have to be derived. Consequently some approximations will be made. We first consider that a delta function illumination generates the spatially modulated populations of carriers by absorption from a deep level or two-photon absorption. We then look at the time evolution of these populations and of the related space charge field, neglecting the recombination of carriers that takes place at longer delays. The system of equation to solve is thus :

$$\begin{cases} \frac{\partial n}{\partial t} = \mu_n \frac{\partial}{\partial x} (nE) + \mu_n \frac{k_B T}{e} \frac{\partial^2 n}{\partial x^2} \\ \frac{\partial p}{\partial t} = -\mu_p \frac{\partial}{\partial x} (pE) + \mu_p \frac{k_B T}{e} \frac{\partial^2 p}{\partial x^2} \\ \frac{\partial E}{\partial x} = -\frac{e}{\epsilon} (n - p - N^+) \end{cases}$$

The different quantities are written : $X = X_0 + \text{Re} (X_1 e^{ikx})$.

We solve these equations for spatially modulated components. We obtain the differential equation for E_1 (n_1 and p_1 are governed by the same homogeneous differential equations) :

$$\begin{aligned} & \frac{\partial^2 E_1}{\partial t^2} + \frac{\partial E_1}{\partial t} \left[\frac{e}{\epsilon} \mu_n n_0 + k^2 \mu_n \left(\frac{k_B T}{e} \right) + \frac{e}{\epsilon} \mu_p p_0 + k^2 \mu_p \left(\frac{k_B T}{e} \right) \right] + \\ & + E_1 \left[\left(\mu_p k^2 \left(\frac{k_B T}{e} \right) + \mu_p p_0 \frac{e}{\epsilon} \right) \left(\mu_n k^2 \left(\frac{k_B T}{e} \right) + \mu_n n_0 \frac{e}{\epsilon} \right) - \mu_n \mu_p n_0 p_0 \left(\frac{e}{\epsilon} \right)^2 \right] \\ & = \left[\mu_n k^2 \left(\frac{k_B T}{e} \right) \right] \left[\mu_p k^2 \left(\frac{k_B T}{e} \right) \right] \left(\frac{e N_1}{ik} \right) \end{aligned}$$

Two different cases can occur, depending on the value of the non spatially modulated population of free carriers (n_0 and p_0).

* At low excitation (i.e. small concentration of photoexcited carriers), we have :

$$(\tau_{\text{DIn}})^{-1} = \frac{e}{\varepsilon} \mu_n n_0 \ll k^2 \mu_n \frac{k_B T}{e} = (\tau_{\text{Dn}})^{-1}$$

$$(\tau_{\text{Dlp}})^{-1} = \frac{e}{\varepsilon} \mu_p p_0 \ll k^2 \mu_p \frac{k_B T}{e} = (\tau_{\text{Dp}})^{-1}$$

where $\tau_{\text{DIn}, p}$ are the dielectric relaxation times $((\tau_{\text{DIn}, p})^{-1} = \frac{e(n_0, p_0) \mu_{n, p}}{\varepsilon})$ for electrons and holes), and $\tau_{\text{Dn}, p}$ are the diffusion time constants $((\tau_{\text{Dn}, p})^{-1} = \frac{e}{k^2 \mu_{n, p} k_B T})$ for electrons and holes).

In this case the homogeneous equation becomes :

$$\frac{\partial^2 E_1}{\partial t^2} + \frac{\partial E_1}{\partial t} \left(\frac{1}{\tau_{\text{Dn}}} + \frac{1}{\tau_{\text{Dp}}} \right) + \frac{E_1}{\tau_{\text{Dn}} \tau_{\text{Dp}}} = 0.$$

With the two diffusion time constants τ_{Dn} and τ_{Dp} . In this regime, the space charge field build-up is governed by diffusion as discussed in reference [10].

* At high excitation when :

$$(\tau_{\text{DIn}})^{-1} = \frac{e}{\varepsilon} \mu_n n_0 \gg k^2 \mu_n \frac{k_B T}{e} = (\tau_{\text{Dn}})^{-1}$$

$$(\tau_{\text{Dlp}})^{-1} = \frac{e}{\varepsilon} \mu_p p_0 \gg k^2 \mu_p \frac{k_B T}{e} = (\tau_{\text{Dp}})^{-1}$$

the homogeneous equation becomes :

$$\frac{\partial^2 E_1}{\partial t^2} + \frac{\partial E_1}{\partial t} \left[\frac{e}{\varepsilon} \mu_n n_0 + \frac{e}{\varepsilon} \mu_p p_0 \right] + E_1 \left[\frac{e}{\varepsilon} \mu_n n_0 \mu_p k^2 \left(\frac{k_B T}{e} \right) + \frac{e}{\varepsilon} \mu_p p_0 \mu_n k^2 \left(\frac{k_B T}{e} \right) \right] = 0.$$

With again two time constant τ_+ (fast) and τ_- (slow) which expressions are :

$$(\tau_+)^{-1} = \frac{e}{\varepsilon} (\mu_n n_0 + \mu_p p_0)$$

$$(\tau_-)^{-1} = k^2 \left(\frac{k_B T}{e} \right) \mu_a \quad \text{and} \quad \mu_a = \frac{n_0 + p_0}{\frac{n_0}{\mu_p} + \frac{p_0}{\mu_n}}$$

Under these conditions of high excitation, the kinetics of the spatially modulated electron and hole populations and space charge field are governed by the above same two time constants. A more detailed analysis including a carrier photoemission by a Gaussian beam [26] leads to similar expressions.

The fast time constant τ_+ , given by $\tau_+ = \frac{\varepsilon}{e(n_0 \mu_n + p_0 \mu_p)}$, corresponds to the first decay component of the free carrier grating modulation. The same time constant governs the build-up

of the space charge field. The slow time constant τ_- is given by $\tau_- = \frac{e}{k^2 k_B T \mu_a}$, with

$$\mu_a = \frac{n_0 + p_0}{\frac{n_0}{\mu_p} + \frac{p_0}{\mu_n}}$$
 the ambipolar mobility. This time constant corresponds to the second decay

component of free carrier grating and to the decrease of space charge field. In fact, in our crystals, due to the mobility values and with our excitation conditions, the fast time constant τ_+ is always smaller than the laser pulse duration and cannot be observed. It also means that we can not resolve the build-up of the space charge field with these picosecond experiments. Only the slow time constant τ_- is observed in our experiments. Its variation with fluence indicates how the ratio n_0/p_0 varies with energy.

This regime of high excitation corresponds to non spatially modulated free carrier concentrations : $n_0, p_0 \gg k^2 \frac{k_B T}{e} \frac{\varepsilon}{e}$ where $k = \frac{2\pi}{\Lambda}$. In our materials and for our experimental parameters ($\Lambda = 1.8 \mu\text{m}$) this leads to $n_0, p_0 \gg 2 \times 10^{14} \text{ cm}^{-3}$. Note that in this regime the non modulated part of free carrier concentration is $n_0 \approx n_1/m$ [6] (with $m \leq 1$ the fringe modulation ratio). For all the range of energy, we have a slope $\gamma > 4$ which indicates a strong influence of two photon mechanisms in absorption (direct band-to-band absorption or *via* deep levels). This means that we have generation of nearly equal electron and hole densities, i.e. $n_0 \approx p_0$. From the above analysis of our results (see Sect. 2) we derive that at fluences as low as 4 mJ.cm^{-2} , the carrier concentration is $n_0 \approx p_0 \geq 6 \times 10^{14} \text{ cm}^{-3}$ in CdTe and $n_0 \approx p_0 \geq 6 \times 10^{15} \text{ cm}^{-3}$ in GaAs. The ambipolar model is thus applicable for describing our experiments. Experimental results will show the variation of τ_- with the ratio n_0/p_0 with a minimum value for $n_0 = p_0$. As photogeneration of electrons is preminent in our crystals at low excitation (as it was in C.W. regime) in both CdTe and GaAs, τ_- is reduced (faster decay) when the fluence is increased as p_0 tends to equal n_0 (Fig. 6).

3.2 CdTe. — In CdTe, for FC gratings, we first see a fast decay followed by a long tail (Fig. 7). The first time constant decreases with fluence and is attributed to the ambipolar decay time τ_- . The tail corresponds to an absorption grating due to the spatial modulation of the deep level. The decay time constant of this absorption grating is $\tau \approx 1.5 \text{ ns}$, this time constant can be identified as the recombination time constant on deep levels.

Fig. 6. — Theoretical variation of the ambipolar diffusion time constant with the n_0/p_0 ratio. The calculation was done with the parameters of GaAs ($\mu_n = 5200 \text{ cm}^2.\text{V}^{-1}.\text{s}^{-1}$, $\mu_p = 400 \text{ cm}^2.\text{V}^{-1}.\text{s}^{-1}$) and a grating period $\Lambda = 1.8 \mu\text{m}$.

In order to determine the first time constant values, we make an adjustment of experimental data with a double exponential curve. Good agreement is obtained. The long decay time is fixed for all curves at $\tau \approx 1.5$ ns. We determine that the first decay time decreases with fluence and saturates to the value $\tau \approx 240$ ps (grating decay time corresponds to time interval $\Delta t = \tau$ when diffracted signal decreases to level e^{-2}) (Fig. 7). The values of τ_1 determined for each experimental curve are not sensitive to variations around 1.5 ns for the long decay time constant.

Fig. 7. — Free carrier grating dynamics at high excitation levels : $E = 8.5 \pm 0.5 \text{ mJ.cm}^{-2}$ (1), $6.5 \pm 0.3 \text{ mJ.cm}^{-2}$ (2), $4.5 \pm 0.25 \text{ mJ.cm}^{-2}$ (3); the decay time constant saturates at the value $\tau \approx 240$ ps. The thin line curves present the best fits with double exponential curves. The CdTe sample orientation is (110) along $\mathbf{k}$.

As previously explained, this limiting values is reached when $n_0 = p_0$ when increasing energy. This is what is expected in this regime where two-photon absorption predominates (direct band-to-band absorption or *via* deep levels). We thus determine the ambipolar mobility

$$\text{value : } \mu_a = \frac{2 \mu_p \mu_n}{\mu_p + \mu_n} = 135 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$$

The same procedure is applied to study the photorefractive grating decay. We see an acceleration of the decay, the time constant decreasing to a saturation value of about 260 ps (Fig. 8). This value corresponds to the one found for FC grating. We again see the appearance of a signal at longer delays which is due to the space charge field created in the deep level. Its time constant is of the order of 1 to 2 ns, which corresponds quite well to the decay time constant found for the absorption grating.

3.3 GaAs. — In GaAs, the same features are observed, with a decrease of the decay time constant of FC grating until $\tau_1 \approx 45\text{-}50$ ps (Fig. 9a) which corresponds to the limit $n_0 = p_0$. So we deduce the ambipolar mobility value $\mu_a \approx 760 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$

As for CdTe, we observed a long tail (Fig. 9b) which, here again, reveals an absorption grating induced by deep level photoionization. The decay time of this tail is about 1 to 2 ns, which is of the order of the known value for electron recombination time constant in GaAs [1].

Fig. 8. — Photorefractive grating build-up and decay at different excitation levels in CdTe : $E = 6.5 \pm 0.5 \text{ mJ.cm}^{-2}$ (1), $4.75 \pm 0.25 \text{ mJ.cm}^{-2}$ (2) showing the decrease of the time constant to $\tau \approx 260 \text{ ps}$. The thin line curves present the best fits with double exponential curves. The sample orientation is (110) along k .

Fig. 9. — Free carrier grating dynamics at high excitation levels in GaAs : (a) $E = 3.25 \pm 0.25 \text{ mJ.cm}^{-2}$ (1), $5.5 \pm 0.5 \text{ mJ.cm}^{-2}$ (2) (b) $8.5 \pm 0.5 \text{ mJ.cm}^{-2}$. The decay time constant are $\tau_1 \approx \tau_2 \approx 43\text{-}45 \text{ ps}$. The long tail is due to absorption grating.

The diffraction efficiency of this absorption grating is measured $\eta = \left(\frac{\Delta\alpha d}{2 \cos \theta} \right)^2 = 10^{-6}$ which gives $\Delta\alpha \approx 10^{-2} \text{ cm}^{-1}$ at a delay of 300 ps after the pulse. This value does not correspond to the magnitude of induced absorption $\Delta\alpha \approx 1 \text{ cm}^{-1}$ we observed in our crystal at $\Delta t = 300 \text{ ps}$ in the transmission experiments (Fig. 5). The fact that we do not observe an efficiency corresponding to such an absorption grating ($\eta = 10^{-2}-10^{-3}$) confirms the attribution of this induced absorption to free carrier absorption (see Sect. 2). Indeed, at this time (300 ps) free carriers have diffused and there is no spatial modulation left in their population and then no absorption grating results from the variation of absorption observed in transmission experiments.

For PR grating, we determine the same decay time constant as for FC grating, $\tau_- \approx 45 \text{ ps}$ (Fig. 10). This confirms that the PR grating is caused by Dember field as for CdTe. The lower diffraction efficiency prevents here to observe the space charge field in deep levels.

Fig. 10. — Photorefractive grating dynamics at different excitation levels in GaAs : $E = 2.75 \pm 0.25 \text{ mJ.cm}^{-2}$ (1), $6.75 \pm 0.25 \text{ mJ.cm}^{-2}$ (2) and $8.5 \pm 0.5 \text{ mJ.cm}^{-2}$ (3). The decay time constant decreases and saturates with energy $\tau_1 > \tau_2 \geq \tau_3 \approx 43-45 \text{ ps}$.

4. Conclusion.

We have shown in this study that the investigation of diffraction efficiency of FC and PR gratings with incident energy and time delay, gives a lot of information about carrier generation and transport as well as carrier recombination. Opposite to nanosecond excitation [6], we see that for picosecond illumination, even for moderate fluences, the carrier generation is mainly due to two-photon absorption *via* deep level or direct band-to-band generation. In this regime, the grating decay times are controlled by ambipolar diffusion. We deduce from this study an ambipolar mobility (when $n_0 = p_0$) of $135 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in CdTe and of $760 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in GaAs. Moreover we detect absorption gratings at long delay times. These gratings relax with the recombination time of carriers. We measure $\tau_R \approx 1.5 \text{ ns}$ in CdTe

and $\tau_R \approx 1-2$ ns in GaAs. We also show that, for semiconductors in the picosecond regime, we easily have fluences for which the photorefractive effect is due to Dember field. This field is established during the excitation pulse.

References

- [1] SMIRL A. L., VALLEY G. C., BOHNERT K. M. and BOGGESS T. F., Picosecond photorefractive and free-carriers transient energy transfer in gallium arsenide at $1 \mu\text{m}$, *IEEE J. Quantum Electron.* **QE-24** (1988) 289 ;
VALLEY G. C. and SMIRL A. L., Theory of transient energy transfer in gallium arsenide, *IEEE J. Quantum Electron.* **QE-24** (1988) 304.
- [2] PETROVIC M. S., SUCHOCKI A. and POWELL R. C., Laser-induced grating spectroscopy of CdTe, *J. Appl. Phys.* **66** (1989) 1359.
- [3] RUCKMANN I., KORNACK J., KOLENDA J. and PETRAUSKAS M., Transient energy transfer in ps two-photon absorption experiments in CdTe at room temperature, *Phys. Status Solidi (b)* **158** (1990) 769.
- [4] SHROEDER W. A., STARK T. S., BOGGESS T. F., SMIRL A. L. and VALLEY G. C., Photorefractive nonlinearities caused by the Dember space-charge field in undoped CdTe, *Opt. Lett.* **16** (1991) 799.
- [5] SHROEDER W. A., STARK T. S., DAWSON M. D., BOGGESS T. F., SMIRL A. L. and VALLEY G. C., Picosecond separation and measurement of coexisting photorefractive, bound-electronic, and free-carrier grating dynamics in GaAs, *Opt. Lett.* **16** (1991) 159.
- [6] DISDIER L. and ROOSEN G., Nanosecond four-wave mixing in semi-insulating GaAs, *Opt. Commun.* **88** (1992) 559.
- [7] VAITKUS J., GAUBAS E., JARASIUNAS K. and PETRAUSKAS M., Mapping of GaAs and Si wafers and ion-implanted layers by light induced scattering and absorption of IR light, *Semicon. Sci. Technol.* **7** (1992) A 131.
- [8] ZIARI M., STEIER W. H., RANON P. M., KLEIN M. B. and TRIVEDI S., Enhancement of the photorefractive gain at $1.3-1.5 \mu\text{m}$ in CdTe using alternating electric fields, *J. Opt. Soc. Am. B* **9** (1992) 1461.
- [9] LAUNAY J. C., MAZOYER V., TAPIERO M., ZIELINGER J. P., GUELLIL Z., DELAYE Ph. and ROOSEN G., Growth spectroscopic and photorefractive investigation of Vanadium-doped Cadmium Telluride, *Appl. Phys. A* **55** (1992) 33.
- [10] JARASIUNAS K., DELAYE P., LAUNAY J. C. and ROOSEN G., Space charge field feed-back effect on carrier transport in photorefractive CdTe : V, *Opt. Commun.* **93** (1992) 59.
- [11] JARASIUNAS K. and GERRITSEN H. J., Ambipolar diffusion measurements in semiconductors using nonlinear transient gratings, *Appl. Phys. Lett.* **33** (1978) 190.
- [12] VAITKUS J., SUBACIUS L. and JARASIUNAS K., Transverse ambipolar diffusion of hot charge carriers in GaAs, *Sov. Phys.-JETP Lett.* **45** (1987) 191.
- [13] VAITKUS J., JARASIUNAS K., GAUBAS E., JONIKAS L., PRANAITYS R. and SUBACIUS L., The diffraction of light by transient gratings in crystalline, ion-implanted and amorphous silicon, *IEEE J. Quantum Electron.* **QE-22** (1986) 1298.
- [14] WEINERT H., JARASIUNAS K. and SHUMANN B., Carrier diffusion and recombination in type II GaAs:Ca_xSr_{1-x}F₂ quantum well heterostructures studied by the transient grating technique, *Phys. Status Solidi (a)* **114** (1989) K183.
- [15] JORDAN A. S. and PARSEY J. M., Growth and characterization of GaAs single crystals, *J. MRS Bull.* (1988) 36.
- [16] MARTIN G. M., Optical assessment of the main electron trap in bulk semi-insulating GaAs, *Appl. Phys. Lett.* **39** (1981) 747.
- [17] DOBRILLA P. and BLAKEMORE J. S., Experimental requirements for quantitative mapping of midgap flaw concentration in semi-insulating GaAs wafers by measurement of near-infrared transmittance, *J. Appl. Phys.* **58** (1985) 208.

- [18] THOMAS R. N., MCGUIGAN S., ELDRIDGE G. W. and BARRET D. L., Status of device-qualified GaAs substrate technology for GaAs integrated circuits, *Proc. IEEE* **76** (1988) 778.
- [19] PARTOVI A., GARMIRE E. M. and CHENG L. J., Enhanced beam coupling modulation using the polarisation properties of photorefractive GaAs, *Appl. Phys. Lett.* **51** (1987) 299.
- [20] JARASIUNAS K. and VAITKUS J., Investigation of non-equilibrium processes in semiconductors by the method of transient holograms, *Phys. Status Solidi (a)* **44** (1977) 793.
- [21] JAIN R. K. and KLEIN M. B., Degenerate Four-Wave Mixing in Semiconductors, *Optical Phase Conjugation*, (New York : Acad. Press, 1983) p. 307.
- [22] JARASIUNAS K., DELAYE P. and ROOSEN G., Optical nonlinearities and carrier transport in GaAs:El2 at high excitation levels, *Phys. Status Solidi (a)* **175** (1993) 445.
- [23] TOMITA Y., SHIBATA M. and BERGQUIST J., Pulsewidth dependence of time-resolved two-photon absorption with picosecond pump-probe excitation, *J. Appl. Phys.* **71** (1992) 2102.
- [24] VALLEY G. C., BOGGESS T. F., DUBARD J. and SMIRL A. L., Picosecond pump-probe technique to measure deep-level, free-carrier, and two photon cross-sections in GaAs, *J. Appl. Phys.* **66** (1989) 2407.
- [25] TOMITA Y., SHIBATA M. and BERGQUIST J., Effects of free-carrier absorption on time-resolved pump-probe two-photon absorption in semi-insulating semiconductors, *J. Appl. Phys.* **72** (1992) 1075.
- [26] VALLEY G. C., DUBARD J. and SMIRL A. L., Theory of high gain transient energy transfer in GaAs and Si, *IEEE J. Quantum Electron.* **QE-26** (1990) 1058.