Photorefractive effect in GaAs at low temperature : influence of the metastable state of the EL2 defect

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PHOTOREFRACTIVE EFFECT IN GaAs AT LOW TEMPERATURE : INFLUENCE OF THE METASTABLE STATE OF THE EL2 DEFECT

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Abstract. We present here a theoretical and experimental analysis of photorefractive two-beam coupling in undoped GaAs as a function of temperature. Three major features are experimentally observed, firstly, a change of sign of the photorefractive beam coupling gain around 150K, secondly, an enhancement of the space charge field by a factor 2 compared to the diffusion field and, finally, a strong peak of the absorption grating amplitude around 150K. A photorefractive model is established that includes the metastable state of the EL2 defect with its optical properties (optical generation and optical recovery). It predicts all observed features correctly and is in good agreement with the experimental data.

Introduction. Important parameters which influence the photorefractive effect (PRE) are the nature and the characteristics of the deep defect in which carriers are redistributed. Its concentration and occupancy ratio as well as its optical cross-sections greatly influence the strength of the PRE. A lot of studies are currently performed in order to identify the deep level involved in the PRE in different materials sensitive to the infrared (GaAs[1], InP[2, 3], CdTe[4]). To that purpose an optical contactless technique was proposed : the Deep Level Photodiffractive Spectroscopy (DLPS) [5]. In this technique the study of the variation of the strength of the PRE with temperature gives information on the deep levels present in the sample. This technique was used for InP:Fe to deduce the presence of a secondary defect that influences the PRE [3]. In undoped GaAs, due to peculiar metastability properties of the EL2 defect, drastic variation of the PRE at low temperature were expected and experimentally observed [6, 7]. The aim of this paper is to present both experimental and theoretical investigations performed in GaAs:EL2 with the DLPS technique.

Set-up and sample. We performed two-beam coupling experiments with a semi-insulating GaAs sample. Two s-polarized beams of a diode pumped Nd:YLF laser (emitting at 1.047μm) interfere on the crystal (propagations along [$\bar{1}$10] direction) and create a PR grating with a grating vector k along [001] (grating spacing $\Lambda=3\mu$m). The typical total illumination used in the experiment was 60 mW.cm^{-2} inside the crystal. The PRE creates a refractive index grating leading to an energy transfer from the pump beam towards the probe beam we measure. In order to separate the photorefractive gain Γ from an eventual absorption grating component we use the symmetry of the PRE. In the configuration we use, turning the crystal by 180° around the [110] axis changes the sign of Γ, whereas the absorption grating $\Delta\alpha$ keeps the same sign. Thus measurements in both orientations allow to separate the

photorefractive gain Γ from the absorption grating $\Delta\alpha$. In all our experimental measurements we make this treatment and present directly Γ and $\Delta\alpha$. The sample was placed in a liquid nitrogen cryostat and cooled under illumination. In order to avoid the vibration induced by boiling nitrogen, the measurement was performed during warming-up of the cryostat, after all the nitrogen was evaporated. The temperature range of the experiment was 90-300K. The warming of the sample occured with a maximum rate of about 2K.min^{-1}. Around room temperature a resistor was used to heat the sample. The temperature was measured with a Nickel-Chromium thermocouple placed on the sample carrier. The temperature variation was slow enough to allow the measurement of temperature at each instant when Γ and $\Delta\alpha$ were measured and to ensure that the sample was in thermal equilibrium. We used an undoped GaAs samples with high excess shallow acceptors and [EL2^0]$\approx$[EL2$^+$]. Absorption, Electron Paramagnetic Resonance (EPR) and room temperature photorefractive measurements performed with this sample [1] gave [EL2]= 1.3×10^{16}cm^{-3} and [EL2$^+$]= 6×10^{15}cm^{-3}.

Experimental results. The behavior of the photorefractive gain Γ is rather peculiar (Fig.1). Firstly, between 300K and 150K a small decrease corresponding to the decrease of the diffusion field with temperature is observed. At 150K a strong decrease of the gain occurs followed by a change of sign. This reverse of the sign is followed by a peak of the gain Γ centered around 140K, the maximum value being around 0.12-0.15 cm^{-1}, a value that corresponds to a space charge field higher than the diffusion field. After this peak the gain decreases with temperature. When decreasing the illumination on the sample we observe a displacement of the peak of the gain of about 10K. The other effect we observe concerns the absorption part $\Delta\alpha$ of the beam coupling (Fig.2). The absorption grating is negligible at room temperature as expected in that type of materials and as observed previously [8]. At low temperature we see the appearance of a strong peak of the absorption part of the beam coupling centered at 140K. The absorption grating vanishes at lower temperature. All these features can not be explained by the electron hole competition model with only one defect EL2 that explains the PRE in GaAs at room temperature[1]. In order to explain these features we introduce a metastable state of the EL2 defect in the band transport model that is known to appear in GaAs at low temperature.

The EL2 defect and its metastable state. The electrical and optical properties and parameters of the EL2 defect are well known (Table 1)[9, 10]. An unusual property of this defect is the existence of a metastable state at low temperature called EL2* which is optically and electrically inactive [15]. The transfer of the defect from the neutral state EL2^0 to the metastable state is optically induced by photons with an energy around 1.18eV [15]. With this transfer, known as photoquenching of EL2, a lot of the characteristics of EL2 disappear such as below band-gap absorption [19], electron paramagnetic resonance [20] or diffraction efficiency in PRE measurements [6]. The transfer from EL2^0 to EL2* is optically induced with a cross-section S_n^* [15], whose absolute value at the maximum (experiments were performed at this maximum) is about the tenth of the electron photoionization cross-section

S_n. As there is no evidence of any temperature variation, we assume S_n^* to be constant. The recovery is induced both thermally with a recovery rate r^* [15] and optically with an optical recovery cross-section S_r^* [16]. The spectrum of S_r^* is characterized by two bands centered at 0.85eV and 1.45eV. Between these two maxima the cross-section seems to go to a minimum for our energy of interest (1.18eV) [16]. No absolute value of this cross-section is accessible, nevertheless we can estimate the ratio S_n^*/S_r^* from the experimental curves of ref.[16]. The recovery of the quenched absorption measured at 1.18eV is about 1%. This absorption recovery can be written as a function of the different cross-sections as $\dfrac{S_r^*}{S_r^* + S_n^*}$ [21] which gives an estimation for the ratio $S_n^*/S_r^* \approx 100$. The optical recovery cross-section was also found to be thermally activated [22]. In our study the temperature range, where the metastable state is studied, is between 90 and 150K, a range where S_r^* varies only slightly with temperature [22] (a factor 2 to 3), so we neglect this temperature effect to simplify the present study. So we see that all the parameters that characterize the EL2 defect and its metastable state are well known from literature, including their temperature dependence. We will use these parameters to predict the behavior of the photorefractive effect in undoped GaAs at low temperature.

Theoretical background. To establish a theoretical expression for the photorefractive gain, we extend the band transport model [23, 24], including the metastable state of EL2. We solve it in a classical manner in steady-state without applied field [25]. The zeroth order terms give the concentrations of the different states of the EL2 defect and of free carriers and the first order terms give the modulated space charge field and the eventual absorption grating components.

The concentrations in the different species of EL2 are $\left[EL2^0\right] = A'^*\left(\left[EL2\right] - \left[EL2^+\right]\right)$ and $\left[EL2^*\right] = \left(1 - A'^*\right)\left(\left[EL2\right] - \left[EL2^+\right]\right)$ with $A'^* = \dfrac{r^* + S_r^* I_0}{r^* + S_r^* I_0 + S_n^* I_0}$, the EL2+ concentration will be given by a second order equation (in the case where the free electron concentration is negligible) : $\left[EL2^+\right]^2 - \left[EL2^+\right]\left[\left(\dfrac{\beta_p + S_p I_0}{\gamma_p \, A'^*}\right) + \left(N_A - N_D + \left[EL2\right]\right)\right] + \left(N_A - N_D\right)\left[EL2\right] = 0$. The temperature dependency of this equation occurs through the first term of the second member of the equation and particularly A'*. Indeed A'* depends on the recovery rate r* (Table 1). At high temperature r* goes to a value much greater than $\left(S_r^* I_0 + S_n^* I_0\right)$ and A'* equals approximately 1. When temperature decreases r* goes to zero and A'* decreases to a limit $A'^* = S_r^*/\left(S_r^* + S_n^*\right)$ which goes to zero below 77K as S_r^* decreases with temperature [22]. With these equations we can calculate the temperature variation of the populations in the temperature range 77-300K (Fig.3). From these curves we see an important feature of the EL2 quenching in GaAs, although A'* has decreased by a factor 100, when passing from 300K to 77K (which means that the EL2^0 concentration has decreased by the same factor),

the EL2+ concentration has not changed ($\left[EL2^+ \right] = \left(N_A - N_D \right)$). We have quenched only the neutral part of EL2 and not the ionized part. This is easy to understand physically as the transfer from EL2+ to the metastable state is a transfer in two steps : firstly, the generation of a hole is accompanied by the transformation of EL2+ to EL2^0, then EL2^0 is transferred to EL2*. Then there is competition between the lifetime of holes and the lifetime of the metastable state, resulting in a reduced efficiency of the transfer from EL2+ to EL2*. We can also see on Fig.3 that the concentration of free electrons stays negligible and that free holes are generated at low temperature by the quenching of EL2+.

The spatially modulated space charge field E_1 is calculated from the first order equations [25] :

$$ E_1 = \frac{-i \; m \left(\dfrac{k_B T}{e} \right) k}{1 + A'^* \dfrac{k^2}{k_0^2}} \left[\xi(k) - \left(1 - A'^* - B'^* \right) \right] $$

with $\xi(k) = \dfrac{\alpha_n \left(k^2 + \kappa_p^2 \right) - \alpha_p \left(k^2 + \kappa_n^2 \right)}{A_n \alpha_n \left(k^2 + \kappa_p^2 \right) + A_p \alpha_p \left(k^2 + \kappa_n^2 \right)}$ the electron-hole competition coefficient [24]

and $B'^* = \dfrac{S_n^* S_r^* I_0^2 A'^*}{\left(r^* + S_r^* I_0 \right)^2}$. $A_n = \dfrac{\beta_n + S_n I_0}{S_n I_0}$, $A_p = \dfrac{\beta_p + S_p I_0}{S_p I_0}$ give the influence of the thermal

emission of carriers from EL2 at high temperature. $\alpha_n = S_n \left[EL2^0 \right]$, $\alpha_p = S_p \left[EL2^+ \right]$ are the parts of the absorption that create electrons and holes, respectively. $\kappa_n^2 = \dfrac{e}{k_B T} \dfrac{\gamma_n \left[EL2^+ \right]}{\mu_n}$, $\kappa_p^2 = \dfrac{e}{k_B T} \dfrac{\gamma_p \left[EL2^0 \right]}{\mu_p}$ are the inverse squared of the diffusion lengths of

electrons and holes. Finally, $k_0^2 = \dfrac{e^2}{\varepsilon k_B T} \dfrac{\left[EL2^+ \right] \left[EL2^0 \right]}{\left[EL2 \right]}$ is the inverse squared of the Debye

screening length. The different concentrations [EL2+] and [EL2^0] are given by the zeroth order solution of the equations, as seen previously.
The influence of the metastable state of EL2 appears in the k dependency of E_1 with the term k_0^2 / A'^*. Taking into account zeroth order equations, we obtain :
$\dfrac{k_0^2}{A'^*} = \dfrac{e^2}{\varepsilon k_B T} \dfrac{\left[EL2^+ \right] \left(\left[EL2 \right] - \left[EL2^+ \right] \right)}{\left[EL2 \right]}$. As [EL2+] stays constant between 77K and 300K, we

see that the term k_0^2 / A'^* keeps the value it has at 300K. At lower temperature [EL2+] decreases (as S_r^* decreases) and k_0^2 / A'^* decreases and goes to zero, decreasing E_1. Then the only remaining influence of the metastable state between 77K and 300K is through the term of "hole-electron" competition, i.e. through the value of $R = \xi(k) - \left(1 - A'^* - B'^* \right)$. We will now discuss the variation of this parameter with temperature, by considering its two terms independently to clarify the situation. Considering the values of the different parameters we can make the usual approximation at room temperature : κ_n^2, $\kappa_p^2 << k^2$. Moreover, as thermal emission is negligible for temperatures below 300K we have $A_n = A_p = 1$ and $\xi(k)$ reduces to

$\xi_0 = \dfrac{\alpha_n - \alpha_p}{\alpha_n + \alpha_p}$. In our material, we have [EL2⁰]≈[EL2⁺] which means that we have an electron-hole competition 0<ξ_0<1 [1]. With decreasing temperature (around 150K) we are quenching EL2⁰ (Fig.3) and the ratio becomes [EL2⁰]/[EL2⁺]≈1/100 which means that ξ_0 changes its sign and becomes close to -1 (holes becomes the majority carriers). The second term of R can be rewritten $\left(1 - A'^* - B'^*\right) = \dfrac{S_n^* I_0 r^*}{\left(r^* + S_r^* I_0 + S_n^* I_0\right)\left(r^* + S_r^* I_0\right)}$. At room temperature $r^* \gg S_n^* I_0,\ S_r^* I_0$ then $\left(1 - A'^* - B'^*\right) \approx 0$, at low temperature r^* goes to zero then $\left(1 - A'^* - B'^*\right)$ goes to zero, which means that $\left(1 - A'^* - B'^*\right)$ goes through a maximum when we decrease temperature. If we suppose that the only temperature varying parameter is r^* (we suppose that T>77K and then S_r^* is constant), the maximum is obtained for $r_M^* = I_0\sqrt{S_r^*\left(S_r^* + S_n^*\right)}$, the maximum value being $\left(1 - A'^* - B'^*\right)_M = \dfrac{S_n^*}{\left(\sqrt{S_r^*} + \sqrt{S_r^* + S_n^*}\right)^2}$. From the value of r_M^* (Table 1) we deduce the temperature at which the maximum occurs (around 140K in our case). Firstly, we remark that r_M^* depends on the incident illumination. When we decrease illumination, r_M^* decreases and the temperature where the peak is located decreases. Secondly, we want to point out that the maximum of the expression $\left(1 - A'^* - B'^*\right)_M$ is close to 1 considering the value of the ratio S_n^*/S_r^* to be 100, but it is the closer to one, the higher this ratio.

If we now consider both terms of R together, we obtain for R a value of about -2 at the peak, which means that the gain is twice as large than the maximum gain obtainable in a photorefractive material without applied field considering Kukhtarev's model with only one type of carrier. At high grating spacing when $k^2 \ll k_0^2/A'^*$ we obtain for the space charge field $E_1 = 2\ i m k\left(k_B T/e\right) = 2\ i m E_d$ where E_d is the diffusion field. The enhancement of the gain due to the metastable state can be of a factor 2 in ideal conditions. This enhancement is characteristic for the model we develop here. All band transport models derived from Kukhtarev's model including both carriers [24], or secondary traps [2, 3, 26] predict a reduction of the photorefractive effect. In the real case the effect of temperature is more complicated, as both quantities ξ_0 and $\left(1 - A'^* - B'^*\right)$ vary together with temperature. Nevertheless all the features predicted are preserved, as shown on the calculated theoretical curve (Fig.1), particularly the possibility to have $|R|$ higher than one, even if the maximum value of -2 is hardly obtainable.

In the same way that first order equations give the space charge field they also give the modulated charge redistribution that causes the absorption grating. We can calculate [25]:

$$\alpha_1 = \left[S_p - \left(S_n + S_n^*\right)A'^* + S_r^*\left(A'^* - 1\right)\right]\left(i\frac{k\varepsilon}{e} E_1\right) + m\left(1 - A'^* - B'^*\right)\left(S_r^* - S_n - S_n^*\right)\left[EL2^0\right]$$

The first term corresponds to the classical absorption grating term that exists at room temperature and is very small. The second term is more interesting as it does not exist at room temperature. As it is proportional to [EL2⁰] and to $\left(1 - A'^* - B'^*\right)$, we see it to a peak around

140K. So the absorption grating goes through a maximum (up to 0.2cm^{-1}) as shown on the calculated curves of the absorption grating strength (Fig.2).

Conclusion. Photorefractive two beam coupling experiments performed at low temperature in undoped GaAs reveal peculiar characteristics of this material such as a reversal of the sign of the photorefractive gain, strong enhancement of the value of this photorefractive gain and a peak of the absorption grating strength at low temperature. These results are explained theoretically by introducing the well known metastable state of the EL2 defect in the photorefractive model that appears at low temperature. All experimental characteristics of the photorefractive effect are perfectly described by this model, in particular the fact that space charge fields higher than the diffusion field are observed.

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name	value	reference
S_n electron photoionization cross-section at 1.06μm (cm^2)	1×10^{-16}	[11]
S_p hole photoionization cross-section at 1.06μm (cm^2)	3×10^{-17}	[11]
σ_n capture cross-section of electrons (cm^2)	$5 \times 10^{-19} + 6 \times 10^{-15} \exp\left(-0.0566 \dfrac{e}{k_B T}\right)$	[9]
σ_p capture cross-section of holes (cm^2)	2×10^{-18}	[12]
v_{nth} thermal velocity of electrons (cm.s^{-1})	$4.4 \times 10^7 \left(\dfrac{T}{300}\right)^{\frac{1}{2}}$	[13]
v_{pth} thermal velocity of holes (cm.s^{-1})	$1.65 \times 10^7 \left(\dfrac{T}{300}\right)^{\frac{1}{2}}$	[13]
$\gamma_{n(p)}$ recombination coefficient (cm^3.s^{-1})	$\sigma_{n(p)} \cdot v_{n(p)th}$	
μ_n electron mobility (cm^2.V^{-1}.s^{-1})	$8000\left(\dfrac{T}{300}\right)^{2.3}$	[13]
μ_p hole mobility (cm^2.V^{-1}.s^{-1})	$320\left(\dfrac{T}{300}\right)^{2.3}$	[13]
β_n electron thermal emission coefficient (s^{-1})	$2.83 \times 10^7 \; T^2 \exp\left(-0.814 \dfrac{e}{k_B T}\right)$	[14]
β_p hole thermal emission coefficient (s^{-1})	$3.3 \times 10^4 \; T^2 \exp\left(-0.813 \dfrac{e}{k_B T}\right)$	[14]
r* recovery rate of the metastabe state(s^{-1})	$2 \times 10^{11} \exp\left(-0.30 \dfrac{e}{k_B T}\right)$	[15]
S_n^* metastable state generation optical cross-section (cm^2)	1×10^{-17}	[15]
S_r^* recovery optical cross-section of the metastable state (cm^2)	$\approx 1 \times 10^{-19}$	[16]
ε_r relative dielectric constant	$12.4 \; (1 + 1.2 \times 10^{-4} \, T)$	[13]
n_0 refractive index	$n_0^2 = A + \left(\dfrac{n_\infty^2 - A}{1 - B(h\nu)^2}\right)$ A=7.10 B = 0.18 (eV)$^{-2}$ $n_\infty = 3.255 \; (1 + 4.5 x 10^{-5} T)$ hν in eV	[13, 17]
r_{41} electrooptic coefficient (pm.V^{-1})	1.72	[18]

TABLE 1

Parameters of the EL2 defect used in the theoretical calculations.

Figure 1 : Experimental photorefractive gain Γ (+) as a function of temperature for sample D2. The straight line represents the calculated photorefractive gain (see text for calculation parameters). The dashed lines represents the theoretical gain for a one carrier model corresponding to a space-charge field of amplitude $\pm E_d$.

Figure 2 : Experimental absorption energy transfer $\Delta\alpha$ (+) as a function of temperature for sample D2. The straight line represents the calculated absorption grating strength (see text for calculation parameters).

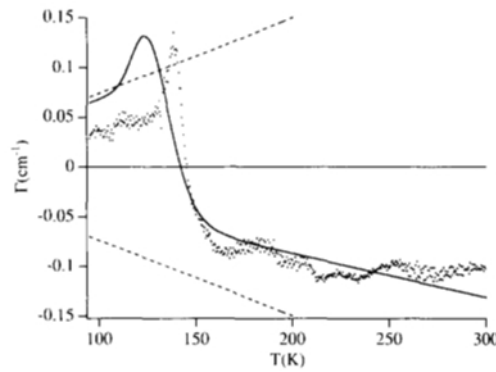Figure 3 : Calculated concentration of the different species present in undoped GaAs as a function of the temperature (illumination I=60mW.cm^{-2}).

Figure 1

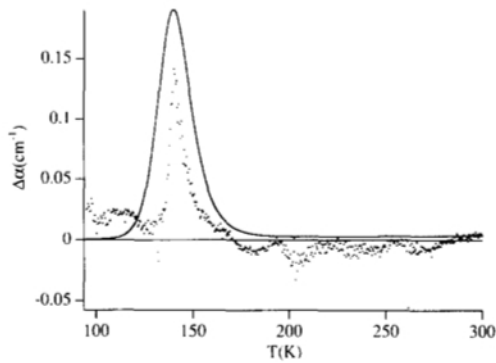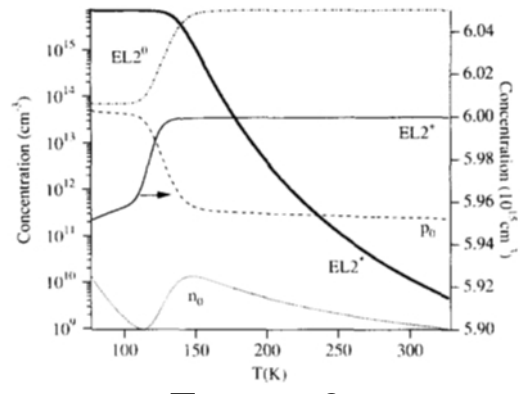Figure 2

Figure 3