Photorefractive wave mixing in undoped LEC GaAs at 1.5μm : validation of photorefractive modeling

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We present photorefractive measurements in undoped GaAs performed at 1.06 µm, 1.32 µm, and at 1.55 µm. Using concentrations of $EL2^{0/+}$ that we determined through optical absorption and electron paramagnetic resonance measurements in the same sample, we show that a single defect model with an electron-hole competition quantitatively explains our results of photorefractive wave mixing.

The photorefractive effect (PRE) is based on the redistribution of charges in a deep defect under a nonuniform illumination. The effect is thus closely linked to the photoionization properties of this deep defect. Discussions currently occur on the exact nature of the deep levels that are source of the photorefractive effect in III–V and II–VI materials.[1–3] GaAs is the most studied among the infrared photorefractive materials, for ten years[4] and correlations were found between the occupancy of the EL2 defect estimated from absorption measurements and the photorefractive performances of samples issued from different parts of the same ingot.[5] We present a comprehensive analysis of semi-insulating GaAs for the sensitivity range of this material, i.e., 1–1.55 µm, with different techniques, such as optical absorption, electron paramagnetic resonance (EPR), and photorefractive effect. On a given sample, we determine the EL2 total concentration as well as the EL2 defect occupancy and photorefractive energy transfer gain. Comparison of theoretical predictions and experimental data obtained for the three wavelengths 1.06, 1.32, and 1.55 µm are presented and discussed, showing that the PRE is perfectly explained by the presence of the EL2 native defect.

The electrical compensation at $T=300$ K of undoped semi-insulating GaAs grown by the liquid encapsulated Czochralski (LEC) technique from pyrolytic BN crucibles has been successfully described in a three defect model based on the interaction of shallow effective mass like donors N_D, shallow effective mass like acceptors N_A, and the deep double donor EL2 with its 0/+ level at $E_c - 0.748$ eV.[6] Typical concentrations in state of the art materials are $N_D \approx 10^{14}$ cm^{-3}, $N_A \approx (2-5) \times 10^{15}$ cm^{-3}, and $N_{EL2} \approx 2 \times 10^{16}$ cm^{-3}.

The EL2 defect is responsible for the extrinsic photoconductivity of this material in the 1–1.5 µ wavelength range. With the Fermi level pinned by the $EL2^{0/+}$ level we have the possibility of the two transitions: $EL2^0 \rightarrow EL2^+ + e_{CB}^-$ and $EL2^+ \rightarrow EL2^0 + h_{VB}^+$ with the corresponding photoionization cross sections S_n and S_p, whose absolute values have been determined in the wavelength range of interest.[7] From the

curve at 300 K in Ref. 7 we deduce: $S_n = 1 \times 10^{-16}$ cm^2 and $S_p = 4 \times 10^{-17}$ cm^2 at 1.06 µm, and $S_n = 1.5 \times 10^{-17}$ cm^2 and $S_p = 5 \times 10^{-17}$ cm^2 at 1.32 µm. For $\lambda = 1.55$ µm, $S_p = 2 \times 10^{-17}$ cm^2 and no value can be extracted on this curve for S_n. In order to determine the value of S_n, we use the result at 78 K which gives a ratio $S_n/(S_n + S_p)$ that is nearly the same at 1.32 and 1.55 µm. Assuming that this property is preserved at 300 K, we deduce $S_n = 5 \times 10^{-18}$ cm^2.

The modeling of the energy transfer gain in photorefractive two-beam coupling experiments requires the knowledge of the $EL2^0$ and $EL2^+$ concentrations. Both of them can be obtained by pure optical absorption measurements at the two different photon energies $h\nu_1$ and $h\nu_2$ according to:

$$\alpha(h\nu_1) = S_n(h\nu_1)[EL2^0] + S_p(h\nu_1)[EL2^+]$$

$$\alpha(h\nu_2) = S_n(h\nu_2)[EL2^0] + S_p(h\nu_2)[EL2^+]$$

where α and S are the absorption coefficient and the photoionization cross section, respectively.[8] However, it has been shown that this all-optical method is not sufficiently reliable for the determination of the $EL2^+$ concentration.[8] Therefore we prefer to determine the $EL2^+$ concentration from the intensity of the $EL2^+$ related EPR spectrum. The optical absorption coefficient α (Fig. 1) is determined from a transmission experiment and deduced with the formula: $T = (1-R)^2 \exp(-\alpha l)/[1 - R^2 \exp(-2\alpha l)]$ for normal incidence, R being the reflectivity of the surface of the sample.[9,10] It has been shown, that the surface state of the sample influences the reflectivity R and that the ideal value deduced from the refractive index must be corrected.[11] Values of $R = 0.2990$ at 1.1 µm and $R = 0.3045$ at 1.0 µm have been proposed. This fact is confirmed by our experiment as we found slightly negative absorption at $\lambda = 2$ µm where almost zero absorption is expected. We keep in our measurement the expression of n with λ from Ref. 9, which presents the advantage of giving a variation of R with photon energy, and we correct our absorption measurement with the
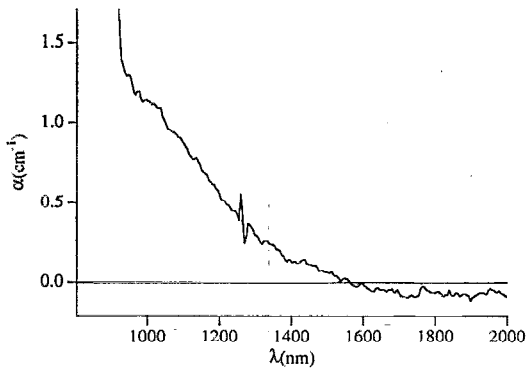
FIG. 1. Absorption spectrum of GaAs sample D2.

offset of absorption at 2 μm. From the absorption measurement, we deduce the EL2 concentration. We do not use the calibration factor of Ref. 12 as it was established for samples where $[EL2^0] \gg [EL2^+]$ (which would lead to an underestimation of the total concentration of EL2 in our sample). We prefer to use the wavelength of 1.19 μm[8] where electron and hole photoionization cross sections are equals: $S_n = S_p$ =4.9×10^{-17} cm^2.[7] At λ=1.19 μm, we have α=0.65 cm^{-1} which gives for the EL2 total concentration, $[EL2]$=1.3×10^{16} cm^{-3}.

The $EL2^+$ concentration of the sample is determined by electron paramagnetic resonance (EPR) spectroscopy from the arsenic antisite related quadruple EPR spectrum.[13] The measurement is performed at a temperature of T=16 K under thermal equilibrium conditions. The sample shows only one EPR spectrum corresponding to the EL2 related arsenic antisite defect (Fig. 2). This spectrum is fully quenched by absorption in the intracenter transition at 1 μm. The spin Hamiltonian of the As_{Ga}^+ defect, which is an electronic spin S=1/2, nuclear spin I=3/2(100%) system is:
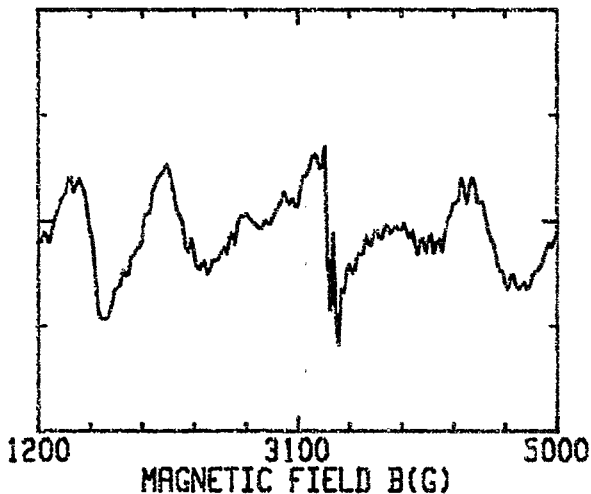

FIG. 2. EPR spectrum of sample D2 showing the arsenic antisite related spectrum.

$$H = g\beta BS + AIS,$$

with the parameters g=2.04±0.01 and A=(890±10)×10^{-4} cm^{-1}, both isotropic. β is the Bohr magnetron and B is the magnetic field. The spin concentration is obtained by double integration of the experimental first derivative spectrum of the magnetic susceptibility χ ($\partial\chi/\partial B$) and by comparison with a spin standard sample Al$_2$O$_3$:Cr (National Bureau of Standards). We find $[EL2^+]$=6×10^{15} cm^{-3}. The relative error is estimated to be ±10%. We then deduce $[EL2^0]$=7×10^{15} cm^{-3}.

We analyze the photorefractive effect in a two-beam coupling experiment performed at the three wavelengths λ=1.06 μm and λ=1.32 μm given by cw diode-pumped Nd:YAG lasers and λ=1.55 μm given by a laser diode. We study the photorefractive energy transfer gain variation versus the grating spacing Λ (or the grating wave number $k=2\pi/\Lambda$). The grating vector is along [001] and the beams are vertically polarized (along [1$\bar{1}$0]). For each studied wavelength and especially at 1.55 μm, we verify that the illumination (of the order of some mW cm^{-2}) is high enough to assure that the PRE is saturated according to the incident irradiance, i.e., photoconductivity exceeds dark conductivity. Moreover no induced absorption and no absorption grating are detected in our measurement. The theoretical variation of the photorefractive gain Γ with k is given by an expression developed with the model taking into account bipolar conduction (hole-electron competition) in the case of grating spacing shorter than the diffusion length of the carriers:[14,15]

$$\Gamma = \frac{2\pi n_0^3 r_{41}}{\lambda \cos \theta} \frac{k_B T}{e} \frac{\xi_0 k}{1 + k^2/k_0^2}$$

with

$$\xi_0 = \frac{S_n[EL2^0] - S_p[EL2^+]}{S_n[EL2^0] + S_p[EL2^+]}$$

and

$$k_0^2 = \frac{e^2}{\epsilon k_B T} \frac{[EL2^0][EL2^+]}{[EL2^0] + [EL2^+]}.$$

In these expressions n_0 is the refractive index of the material (n_0=3.44 at 1.06 μm, n_0=3.38 at 1.32 μm, and n_0=3.36 at 1.55 μm)[9,10]; r_{41} is the electro-optic coefficient (r_{41}=1.72 pm V^{-1} at 1.06 μm, r_{41}=1.54 pm V^{-1} at 1.32 μm and r_{41}=1.50 pm V^{-1} at 1.55 μm),[16] θ is the half-angle between the beams inside the crystal (we have for all grating spacings $\cos \theta \approx 1$), ϵ is the static dielectric constant (ϵ=1.13×10^{-10} F m^{-1}.)[10]

ξ_0 is the hole-electron competition coefficient and k_0^2 is the inverse squared of the Debye screening length. These parameters depend on the photoionization cross sections of carriers (S_n for electrons and S_p for holes) from the deep level EL2 (wavelength dependent parameter) and on the concentration of the two state of charges of the EL2 defect (wavelength independent parameter).

Experimentally we measure the energy transfer from a strong pump beam to a weak probe beam as a function of the grating spacing and deduce the photorefractive gain Γ (Fig. 3).[4,14] We measure a change of the sign of the photorefractive
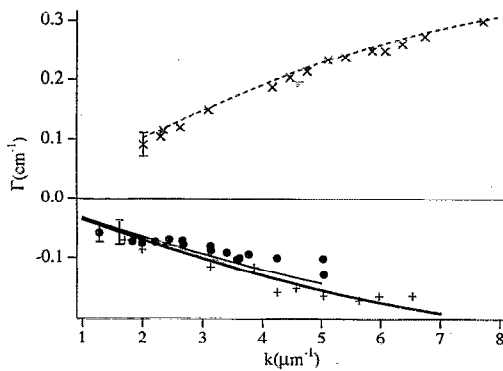
FIG. 3. Experimental (symbols) and theoretical (lines) photorefractive gain Γ as a function of the grating wave number k for different wavelengths: ($\times$) and dashed line for $\lambda=1.06$ μm, (+) and bold straight line for $\lambda=1.32$ μm, (●) and thin straight line for $\lambda=1.55$ μm.

GaAs. We show that the phototrefractive effect in undoped LEC GaAs can be quantitatively explained by the *electron hole competition* model with the single EL2 defect. Comparison of theory and experiment is performed on the sensitivity range (1–1.55 μm) of GaAs. We show that it extends until 1.55 μm, where photorefractive gains close to the absorption loss are obtained ($\Gamma \approx \alpha \approx 0.1$ cm^{-1}). The photorefractive effect in GaAs can thus be used as a contactless optical method of characterization of the compensation level of the EL2 defect, as performances around 1.3 μm are very sensitive to the $[EL2^0]/[EL2^+]$ ratio.

gain between 1.06 μm on the one hand and 1.32 and 1.55 μm on the other hand (Fig. 3). This means that ξ_0 is positive at 1.06 μm (electron dominated photorefractive effect) and ξ_0 is negative at 1.32 and 1.55 μm (hole dominated photorefractive effect). All the parameters that influence the theoretical photorefractive gain are known, so we can compare our experimental results with the theoretical curves (Fig. 3). The accordance between experimental results and theoretical curves is good. It, particularly, well explains the change of sign of the gain between 1.06 μm, 1.32 μm, and 1.55 μm.

In conclusion, using absorption and electron paramagnetic resonance measurements we determine the concentration of the different states of charge of deep level EL2. These results and the known properties of the EL2 defect, allow us to calculate the photorefractive performance of undoped

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2642   Appl. Phys. Lett., Vol. 64, No. 20, 16 May 1994

Delaye *et al.*