Single Si δ -Doped GaAs Investigations by New Photothermal Wavelength Modulated Photocurrent Technique

Fikret HAJIEV, Yener ÖZKAN

TUBITAK-UEKAE, The National Institute of Electronics, P.O Box 21, 41470, Gebze, Kocaeli-TURKEY **M. Çetin ARIKAN** Faculty of Sciences, University of Istanbul, Vezneciler, 34459, İstanbul-TURKEY

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Abstract

New Photothermal Wavelength Modulated Photocurrent (PWMPC) technique is reported. This technique is used for investigation of the MBE grown p-GaAs sample in which a single Si-layer was embedded with a δ -type profile. Two spectral features were observed at 1.5137 eV and 1.5115 eV at 20 K. These peaks were attributed to the (D°,X) and (A°,X)-excitons bound to neutral donors and acceptors, respectively. We studied the temperature dependence of these excitonic peaks positions at temperatures between 20-90 K. Additionally, we demonstrate a blue shift of photocurrent spectra under a low level illumination intensity. PWMPC technique and the nature of the registered signals were explored in detail. The dependence of the excitonic peaks on the chopping frequency and the intensity are discussed.

1. Introduction

In recent years, new crystal growth techniques, such as molecular beam epitaxy (MBE), have made it possible to form different semiconductor microstructures. The optical and electronical properties of these low-dimensional semiconductors are remarkably different from those of bulk semiconductors. Delta (or atomic plane) doping of compound semiconductors has attracted considerable interest because of the possibility of fundamental studies of the two-dimensional system as well as appealing applications to optoelectronical devices [1]. Such systems exhibit strong excitonic effects as a result of the confinement and correlation properties of electronic particles.

The present paper deals with an investigation of band structures of the MBE-grown p-GaAs layers containing a single delta layer of Si dopants. These structures have been the subject of various magneto-transport [2-3], magneto-photoluminescence[4] and strain-modulated photoluminescence [5] studies. Photoluminescence has been widely used to investigate single [5-6] and periodically Si d-doped GaAs [7]. The oscillations above the GaAs fundamental band gap have been investigated by using photoreflectance spectroscopy [8]. To the best of our knowledge, the first observation of the PWMPC phenomena in a single Si d-doped p-GaAs structure was reported in [9].

2. Experimental Details

Sample W53 was grown by MBE on semi-insulating [001]-oriented GaAs substrates. The delta-layer of Si (20 Å thickness and $N_d = 8^* 10^{12} \text{ cm}^{-2}$ nominal donor doping concentration) lies between 2.5 mkm and 1 mkm thickness layers of GaAs [6]. The sample employed Ohmic contacts which were formed by diffusing indium through all the layers. When the two-dimensional electron density exceeds a critical value, the δ -doping layer shows metallic conductivity at low temperatures. Since the GaAs wells and δ -layer are connected in parallel (longitudinal geometry), this modification resulted in significant increase in the sensitivity of the PWMPC. This allows us to study both the electronic structure of the GaAs buffer and the confinement donor dopants in a quasi monatomic layer, simultaneously.

The experimental setup for observation of PWMPC phenomena is similar to the lateral photoconductivity method. The sample was placed in a He closed-cycle cryostat enabling temperature stabilization between 20 and 300K. The sample was illuminated with a 50W quartz halogen source, whose light was chopped at frequency f = 5...10 Hz and dispersed through a 0.5m grating monochromator. The modulated photocurrent (PC) due to incident chopped light was measured by a conventional lock-in technique. Wavelength of the incident light with intensity I scanned near the band gap region, The Ohmicity of the contacts was verified from the linearity of the current J and voltage characteristics of the sample, both in dark and under illumination.

In comparison to a conventional AC PC spectroscopy, in addition to the fundamental (f), we separately measured values of the PC vector and its phaseshift on the 2f, 3f... etc. frequencies, i.e. higher harmonics of PC. The measurements were carried out with an electric field of about 150 V/cm along the layer, which is orders of magnitude smaller than the one employed during the measurement of transverse PC. The spectral resolution of the spectrums was ± 0.3 meV.

3. Results

Two spectral features were observed at 1.5137 eV and 1.5115 eV at 20 K (see Figure 1). These peaks were attributed to the (D°,X) and (A°,X)-excitons bound to neutral donors and acceptors, respectively. The shape of spectral features was similar to those obtained by wavelength modulation spectroscopy methods (strain-modulated photoluminescence, for example). Full width at half maximum (FWHM) of the sharper peak decreased from

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0.6nm to 0.1nm when the incident light intensity increased from 0.2 to 1.0 μ W/cm² at 20K. FWHM depends on temperature, intensity of the incident beam and the applied electric field value. Maximal phaseshift of PC occurred at about 20° for the fundamental frequency and at about 180° for the **4f** component.



Figure 1. The PWMPC spectra of single Si δ -doped p-GaAs. (RMS amplitude and phaseshift component of PC, modulated at second harmonic of fundamental modulation frequency **f**).

Figure 2 illustrates the near band gap spectrums of normalized DC, \mathbf{f} and $2\mathbf{f}$ components of AC PC at 20K. The best signal-to-noise ratio was obtained using of the PC component at the second harmonic of chopping frequency. Since the exciton is electrically neutral, it carries no current, as may be verified by computing the expectation value of the current operator in an excitonic state [10]. Additionally, metallic type conductivity in the Si layer causes high background signal in the DC PC spectrum. Due to these

obstacles, DC PC near band gap changes by 5% only. This means that a mixture of layer (surface) and bulk current was present, and the surface contribution dominated the bulk effect.

Figure 3 shows the temperature dependence of excitonic peaks positions between 20-90K. The dashed curve represent the temperature dependence of the GaAs band gap, calculated from Eg=1.522-5.8*10⁻⁴T² /(T+300) [11]. The solid lines of the temperature dependence of (D°,X) and (A°,X) excitons have parallel shift to lower energies with respect to the GaAs band gap. The best fits of experimental results calculated for (D°,X) peaks are: $E_{D^\circ}=1.5147-5.6*10^{-4}T^2$ /(T+350) and for (A°,X) peaks are: $E_{A^\circ}=1.5120 5.6*10^{-4}T^2$ /(T+350). These excitons are stable at temperatures range up to 65K and 83K respectively.



Figure 2. The PWMPC and DC spectra of single Si δ -doped p-GaAs. Left scale: first and second harmonics of PC. Right scale: DC PC normalize on the dark current (dc) value.

Figure 3. Temperature dependence PWMPC peaks associated with (D°, X) and (A°, X) excitons bound to neutral donors and acceptors (solid lines). Dashed line-temperature dependence of band edge according M.D.Sturge [11](calculated curve).

Figure 4 demonstrates the blue shift of (A°,X)-spectra under low level illumination intensities. This peak shift is reversible. The absolute value of the shift being about 30 times larger than the inhomogeneous exciton bandwidth under the 1.0 μ W/cm² light intensity.

The AC PC signal at the **2f** versus fundamental frequency of modulation is shown in Figure 5. The upper two curves in this figure were obtained under illumination of the sample by light with photon energy close to resonant conditions of spectroscopic features and the lower one-by He-Ne laser light with photon energy higher than band gap. Increasing the intensity of illumination enhanced both the background signal levels associated with second harmonic part of the chopped incident beam and the derivative

component associated with wavelength modulation (WM) due to heating of the sample. The slow temporal dependence of this signal proves the thermal mechanism of PC. PC at 633nm light was about 20 times lower than under resonant illumination at the same intensity in spite of the photon energy being higher than the band gap.

Figure 4. Position of PWMPC peak, associated with (A°, X)-bound to acceptors exciton, versus intensity of excitation light with wavelength $\lambda_1 = 819$ nm.

4. Discussion

PWMPC is a technique which probes the different harmonics of a samples PC when the sample is illuminated with an intensity modulated quasi-monochromatic beam ΔI . When the wavelength of the excitation light is near any critical point in the density of

states, it is assumed that the AC current is modulated through a combined Drude (freecarrier) and thermal mechanism. The theoretical PWMPC signal can be expressed as the modulated PC, ΔJ :

$$\Delta J = \frac{\partial J}{\partial I} \Delta I + \frac{\partial J}{\partial T} \Delta T, \tag{1}$$

where $\partial J/\partial I$ is the photocurrent generating rate; ΔI is the RMS value of modulated light intensity; and ΔT the RMS value of sample temperature. Near the band edge, PC versus temperature dependence is taken to be the sum of a contribution due to the temperature dependence of the edge itself plus a background thermocurrent coefficient:

Figure 5. Dependence of the PWMPC signal at the 2f frequency versus fundamental modulation frequency f. Wavelength of excitation illumination: $\lambda_1 = 819$ nm, $\lambda_2 = 633$ nm,

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In our experiments, the excitation beam is intensity- modulated at \mathbf{f} and its wavelength slow scanned through the band gap energy region. Strong absorption of incident beam at the excitonic peaks, increases the temperature of the sample at the fundamental frequency \mathbf{f} . At the same time the sample has a red shift of energies of the gap and the features of band structure with increasing temperature, i.e. wavelength (energy) modulation of the spectra. Due to this obstacle, the first term of (2) describes the derivative signal. If phase sensitive detection (PSD) at a frequency \mathbf{nf} is performed for the spectroscopic purposes under the wavelength modulation conditions, the PSD signal is predominantly the \mathbf{nth} derivative of the measuring curve. This is discussed in the appendix. According to (1) in this case, PSD at \mathbf{f} is the superposition of ordinary AC PC and the first derivative component of the PC spectrum. Phases of these two components of PC are different and a phase relation up to 180° is possible (Figure 1).

We used of an approximately square-wave intensity modulation source. RMS harmonic distribution of the incident beam are listed in Table 1.

Number of	1	2	3	4	5	6	7	8	9
harmonic									
Partial contribution (percent)	100	0.57	20	0.40	8.57	0.37	4.57	0.15	2.40

From the table, we see that the best results may be obtained when spectral features are WM at a frequency **f** and PSD is performed on the resulting second harmonic component **2f** in the measured PC. This harmonic signal is generated by the nonlinear shape of the exciton absorption curve. It is important to minimize spurious signals (first term in (1)) which arise from the presence of **2f** in the excitation beam. Theoretical analysis shows that the **n**th harmonic component in the PC is proportional to the nth power of amplitude or scan width (Λ) of WM, Λ^n , indicating that the higher harmonic components increase in size if Λ is increased. WM scan width in the region of an absorption feature have no constant value and thus the shape of spectrum is not the same as that obtained using absorption spectroscopy. Our experimental results are in good agreement with this explanation.

In Si δ -doped GaAs, the effective Bohr radius is 14.2 nm [6]. The thickness of the δ -layer is less than or equal to 2 nm. In this strong confinement limit case, even at a moderate excitation intensity one may expect the creation of exciton-like excitations within the quantum well. The concentration of particles per unit volume may be much larger than that created in a bulk crystal even at extreme levels of excitation. On the other side, the large but finite number of excitons created inside the quantum well, does not permit the introduction of a gas relevant to the bulk crystal. In the bulk crystal, a decrease in the exciton binding energy due to exciton-exciton interactions, is normally compensated by a band-gap shrinkage because of plasma screening. Thus the exciton

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band does not show any spectral shift with increasing excitation level. In a quantum wells, all effects related to collective processes in electron-hole system are canceled and exciton-exciton interactions are pronounced. Thus, under resonant excitation of these species, a pronounced intensity-dependent blue shift of the exciton band is observed due to exciton-exciton interactions.

Photo-induced blue shift of the Z_3 -exciton band in CuCl crystallites (quantum dots) embedded in glass matrix has been observed previously [12] under the pulse laser excitation conditions.

5. Conclusions

A new WM spectroscopic technique, PWMPC, has been developed and studied. It has been shown that PWMPC is relatively simple and suitable for the study of the band structures of semiconductors with narrow and strong absorption coefficients. We have studied PC properties of Si δ -doped GaAs under photoexcitation near the band gap. Blue shift of spectra under low level excitation has been obtained. The advantages of PWMPC makes it possible to increase sensitivity and spectral resolution by using a narrow band tunable laser. With this new technique opened the way for simple characterization of new semiconductor devices with linear and nonlinear optoelectronical properties.

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Appendix

The technique of deriving small features of PC spectrum involves modulating the band gap of the sample over a bandwidth Λ and at a frequency f. Output PC J depends on the position of the illuminating light frequency ω beneath the band structure, and the PC may be written as a function Φ of ω and also the applied band gap modulation:

$$J = \Phi(\omega + \Lambda \sin ft)$$

Assuming Λ is small compared with the width of the band gap features (excitonic peaks), i.e. a small signal analysis treatment, this expression may be expanded as a Taylor series:

$$J = \Phi(\omega) + (\Lambda \sin ft)\Phi^{1}(\omega) + \left(\frac{\Lambda^{2}}{2!}\sin^{2}ft\right)\Phi^{2}(\omega) + \left(\frac{\Lambda^{3}}{3!}\sin^{3}ft\right)\Phi^{3}(\omega) + \dots$$
(3)

Here $\Phi^n(\omega)$ is the nth derivative of $\Phi(\omega)$.

After algebraic manipulation, the terms in sin nft may be grouped together. The coefficient of sin nft corresponds to the PC of the *n*th harmonic of the fundamental modulation frequency in the output AC current of the sample. The coefficient of sin nft for n = 1, 2 and 3 are:

 $\sin ft$ coefficient:

$$\Lambda \Phi^1(\omega) + \frac{\Lambda^3}{8} \Phi^3(\omega) + \frac{2}{960} \Phi^5(\omega) + \dots$$
(4)

sin 2ft coefficient:

$$-\frac{\Lambda^2}{4}\Phi^2(\omega) - \frac{1}{96}\Phi^4(\omega) + \dots$$
 (5)

 $\sin 3$ ft coefficient:

$$-\frac{\Lambda^3}{24}\Phi^3(\omega) - \frac{3}{2120}\Phi^5(\omega) + \dots$$
 (6)

If PSD of a PC at a frequency ft is performed, the output signal is predominantly the nth derivative of the band structure curve $\Phi(\omega)$ with smaller contributions from the (n+2)th and (n+4)th higher harmonics.

In the region of a saturated absorption feature of half width at half maximum intensity γ , we may write the PC curve shape as a Lorentzian function of frequency centered at $\omega = 0$, superimposed on a sloping gain curve background. This sloping background (DC PC and wide wings from band gap features) may be represented by a quadratic function of frequency with coefficient α, β, δ

$$\phi(\omega) = \frac{\gamma^2}{\omega^2 + \gamma^2} + \alpha \omega^2 + \beta \omega + \delta \tag{7}$$

Differentiating $\phi(\omega)$ with respect to (ω) we find

$$\Phi^{1}(\omega) = -\frac{2\omega\gamma^{2}}{(\omega^{2} + \gamma^{2})^{2}} + 2\alpha\omega + \beta$$

$$\Phi^{2}(\omega) = -\frac{6\omega^{2}\gamma^{2} - 2\gamma^{4}}{(\omega^{2} + \gamma^{2})^{2}} + 2\alpha$$

$$\Phi^{3}(\omega) = -\frac{24\omega\gamma^{4} - 24\gamma^{3}\gamma^{2}}{(\omega^{2} + \gamma^{2})^{4}}$$
(8)

The first derivative, corresponding to phase sensitive detection at the fundamental frequency gives the well known dispersion shaped curve superimposed on a straight background. This representation may be justified by comparing this expression with the curve of figure 2. The second derivative is a symmetric function of wavelength. The third derivative eliminates the background effects altogether and gives a PSD zero voltage crossing at line center ($\omega = 0$).

Equations (4-6) show that the *n*th harmonic component in the PC output modulation is proportional to Λ^n , indicating that the higher harmonic components increase in size if the band gap modulation is increased. Intuitively, one would expect the magnitude of the third harmonic component of the chopping frequency in the PC output to be smaller than the first harmonic component for the same modulation amplitude. This is indeed so, and the optimum modulation width for maximum third harmonic signal is larger than that for the first harmonic PC for the same intensity of incident beam (heating of the sample and band gap modulation). Experimentally, when we increased incident light intensity, higher harmonics PC increased faster, than PC on a fundamental frequency. Unfortunately, for these third (or higher) harmonic modulation bandwidth, the modulation width plus insufficient monochromatisity of incident light, becomes comparable to the separation of the band gap features. This obstacle reduces the signal-to-noise ratio of PC spectrum and also the resolution of the components which tend to merge into each other.