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DIFFERENTIAL SURFACE PHOTOVOLTAGE SPECTROSCOPY OF δ-DOPED GaAs / AlAs QUANTUM WELLS *

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A study of excitonic lines in differential surface photovoltage (DSPV) spectra of p-type (Be) and n-type (Si) δ-doped GaAs / AlAs multiple quantum well (MQW) structures was carried out. The energies and line broadening parameters for a large number of QW related excitonic transitions were determined from the line shape analysis of the DSPV spectra. The transition energies were found to be in a good agreement with theoretical values calculated within the envelope function approximation that took into account the nonparabolicity of energy bands. Analysis of the dependence of the exciton line width on the quantum subband number allowed us to evaluate line broadening mechanisms and interface roughness in the MQW structures. It was found that doping with Si donors bleaches and broadens more effectively the lowest energy excitonic lines as compared to doping with Be acceptors.

Keywords: delta-doped quantum wells, surface photovoltage spectroscopy, excitonic transitions

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1. Introduction

Multiple quantum wells (MQWs) doped with shallow impurities whose intersublevel transition energies lie within the terahertz (THz) spectral range have attracted particular interest as possible active components in sensing THz devices. The advantage of such components is the possibility to tune the transition energies and, hence, the THz detection frequency by the variation of the impurity confining potential. In particular, transition wavelength for Be acceptors or Si donors embedded in GaAs / AlAs QWs by δ-doping method can be tailored over a wide spectral range by changing the thickness of the wells [1, 2]. This enables the development of novel detectors and emitters in THz electronics. Nevertheless, the well width fluctuations (interface roughness) in MQWs will result in inhomogeneous broadening of electronic states [3]. For technological development of electronic devices such inhomogeneities are unacceptable and must be kept as small as possible. Thus, the knowledge about the structural disorder of δ-doped GaAs / AlAs MQWs is particularly important for improving the crystalline quality of heterostructures. A great deal of useful information concerning the structural inhomogeneities of low-dimensional semiconductor structures can be obtained from a study of excitonic line width which is very sensitive to disorder [3, 4]. The differential surface photovoltage (DSPV) spectroscopy, as a non-destructive and contactless technique, is extremely useful in such kind of investigations [5, 6].

In this work, the DSPV spectroscopy was used to study the excitonic transitions in Be and Si δ-doped GaAs / AlAs MQW structures at 90 and 300 K temperatures. The line shape analysis of the DSPV spectra allowed us to extract information on the energies and line broadening parameters of excitonic transitions for a large number of QW subbands. The investigations were mainly concentrated on exciton line broadening which is directly related to the structural quality of MQW systems.

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Table 1. Characteristics of the samples: the repeated period, the quantum well width ($L_{\rm w}$), the δ-doping Be (Si) density $N_{\rm A}$ ($N_{\rm D}$), and the growth temperature of the epitaxial layer (T).

Samples	Periods	$L_{\rm w}$ (nm)	$N_{\rm D}$ ($\rm cm^{-2}$)	$N_{\rm A}$ ($\rm cm^{-2}$)	T (°C)
1303	50	15	–	$2.5{\cdot}10^{12}$	540
1392	40	20	–	$2.5{\cdot}10^{12}$	540
1807	100	20	–	$5{\cdot}10^{10}$	550
L29	40	20	$4{\cdot}10^{9}$	–	670
L30	40	20	$1.4{\cdot}10^{11}$	–	670
L80	40	15	undoped	–	700
L78	40	15	$1.4{\cdot}10^{11}$	–	715
L79	40	15	$4{\cdot}10^{11}$	–	698

2. Samples and experiment

The samples used in the experiment were grown by molecular beam epitaxy technique on a semi-insulating (100) GaAs substrates followed by a 300 nm GaAs buffer layer. Each of the MQW structures contained the 15 or 20 nm wide wells separated by the 5 nm wide AlAs barriers. The QWs were δ-doped at the well centre with Be acceptors or Si donors. The structures were capped with a 100 nm GaAs layer. The main characteristics of each sample are summarized in Table 1.

The surface photovoltage (SPV) measurements were performed in a chopped light geometry by using a capacitor-like system with a transparent conducting top electrode under normalized incident light intensity conditions [7]. Sample was mounted on copper plate, which was the back electrode. Top conducting electrode was indium–tin-oxide (ITO) coated glass plate. A light from 100 W tungsten halogen lamp was passed through a grating monochromator SPM-2 and focused onto the sample. In DSPV spectra measurements, we have used a wavelength modulation technique [4, 6]. The wavelength λ of the incident probe light was modulated by vibrating a fused silica plate at 87 Hz located near the exit slit of the monochromator. The alternating current (ac) signal generated by a chopped light beam in SPV measurements or by a wavelength modulated light beam in DSPV experiment were recorded by a conventional lock-in detection system. The measurements were performed at 90 and 300 K.

3. Results and discussion

3.1. DSPV spectra

The influence of the doping type and doping density on the excitonic transitions in GaAs / AlAs MQWs was examined by investigations of both Si- and Be-doped samples (Table 1). The DSPV spectra of undoped and differently either Be or Si δ-doped GaAs / AlAs MQW structures with $L_{\rm w} = 15$ nm at 300 and 90 K temperatures are presented in Figs. 1(a) and 1(b), respectively. At energies higher than $E_g^{\rm GaAs}$ (1.424 eV at 300 K and 1.507 eV at 90 K), the spectra are dominated by the derivative-like features mnH(L) that could be associated with excitonic transitions in the MQW region of the samples. The notation mnH(L) signifies the transitions between the mth electron and nth heavy- (H) or light-hole (L) subbands. The dominant transitions are due to allowed ($n = m$) excitons 11H, 11L, 22H, 33H, and 44H. However, forbidden ($n \neq m$) exciton transition like 13H is also observed due to the mixing of the heavy- and light-hole energy states. As the temperature is decreased, the DSPV features are shifted to higher energies and become narrower owing to the smaller broadening of the energy states. As can be seen, the most prominent excitonic resonances in the DSPV spectra both at low and room temperature are revealed for nominally undoped sample L80, while the excitonic features appearing in DSPV spectrum of Be strongly δ-doped sample 1303 are somewhat reduced in amplitude and broadened. However, the spectral lines for the excitonic transitions of the lowest subbands of Si δ-doped samples exhibit considerably more significant changes even at the smaller doping levels. Thus, for strongly doped sample L79, the 11H and 11L excitons are spectrally overlapped and the resulting line shape in DSPV spectra becomes asymmetric due to presence of a sizeable band-to-band component. On the other hand, excitonic transitions that are related to higher subbands are still clearly observed in the DSPV spectra. Thus, it may be concluded that the ground state exciton bleaching could be mainly related to the effects of the phase space filling and exchange interaction. The effect of the Coulomb screening on the higher order excitonic transitions in Si-doped GaAs / AlAs QWs seems to be insignificant up to doping density $N_{\rm D} \leq 4{\cdot}10^{11}$ $\rm cm^{-2}$. A detailed analysis of the DSPV line shape allowed us to determine quantitatively the abovementioned changes in parameters of optical transitions.

The model used in the line shape analysis is based on the fact that the SPV signal of GaAs / AlAs MQW structures is mainly generated in the GaAs buffer / substrate region. In this case, the SPV signal is expected to be a product of the light absorption as a function of photon energy in the MQW layers and the spectral response of the buffer layer [4, 8]. The latter is slowly varying with wavelength compared to the sharp

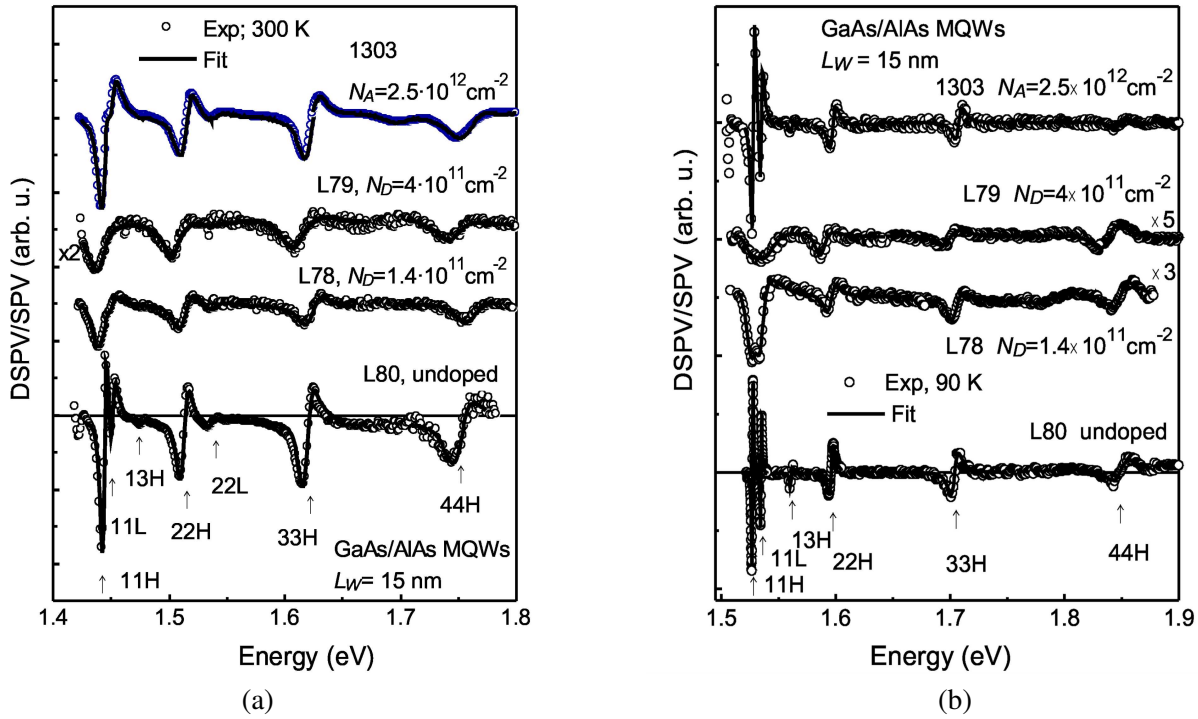

Fig. 1. The DSPV spectra of GaAs / AlAs MQW structures with $L_{\rm w} = 15$ nm for undoped and Be or Si δ-doped samples at (a) 300 and (b) 90 K temperatures. Open circles represent experimental data, while the continuos lines are the result of the fit with derivative functional form (2). The arrows indicate the excitonic transition energies determined from the least-squares fitting.

spectral lines of the optical transitions in the MQWs. Therefore, for the regime of weak excitation, the SPV spectra can be associated with the absorption spectra of the MQW layer. As a result, the normalized DSPV / SPV or $(\mathrm{d}V/\mathrm{d}E)/V$ spectra should be related to the first derivative of the absorption coefficient α of MQW layer versus photon energy E [9]

$$\frac{1}{V}\frac{\mathrm{d}V}{\mathrm{d}E} \sim \frac{1}{d_{\rm w}}\frac{1}{I}\frac{\mathrm{d}I}{\mathrm{d}E} = -\frac{\mathrm{d}\alpha}{\mathrm{d}E}\,, \qquad (1)$$

where V is the SPV signal, $d_{\rm w}$ denotes the thickness of the MQW absorbing layer, and I labels the light intensity absorbed in the GaAs buffer layer.

The energies and broadening parameters of optical transitions responsible for observed DSPV features (Fig. 1) were determined by fitting the experimental DSPV / SPV spectra with derivative functional form proposed by Aspnes [10]:

$$\frac{1}{V}\frac{\mathrm{d}V}{\mathrm{d}E} = \mathrm{Re}\left[C\mathrm{e}^{\mathrm{i}\Theta}(E - E_{\rm ex} + \mathrm{i}\Gamma)^{-m}\right], \qquad (2)$$

where C, Θ, $E_{\rm ex}$, and Γ are amplitude, phase, energy, and broadening parameter of the spectral line, respectively. The value of parameter $m = 2$ was usually used in the calculations. In this case, the expression (2) represents quite well the first derivative of an excitonic dielectric function with a Lorentzian absorption profile [10], which is appropriate for excitonic transitions in QW structures. As already mentioned, for the strongly Si doped sample L79 the lowest 11H(L) excitonic levels are strongly damped, and band-to-band transitions become important at the fundamental band edge of MQWs. In this case the value of parameter $m = 1$ was also used in order to reproduce the experimental line shape. For higher energy transitions of this sample, the excitonic line shape profile was employed. As seen from Fig. 1, the DSPV spectra both at room and low temperatures were sufficiently well fitted to the first derivative of a Lorentzian-type function (solid lines). The arrows in Fig. 1 indicate the transition energies determined from the least-squares fitting.

To identify the spectroscopic data, we have carried out the calculations of the interband transition energies for symmetrical square GaAs / AlAs QWs both for parabolic and nonparabolic energy bands in the envelope function approximation [11]. As follows from the results of the calculation, parabolic band model predicts correctly the energies of the lowest transitions but overestimates the energies of the higher ones. However, the experimental data are in reasonable agreement with calculations performed taking into consideration the energy bands' nonparabolicity effects. It was found that for the most transitions studied the calculated interband energies exceeded the experimental

data by ∼5–10 meV. This difference should be attributed to exciton binding energy. However, it is somewhat smaller than the exciton binding energies in the undoped GaAs / AlAs QWs obtained from photoluminescence excitation studies [12]. Such discrepancy could be related to many-body effects [13].

3.2. Exciton line broadening

The DSPV spectra of GaAs / AlAs MQWs (Fig. 1) include a large number of well-resolved features associated with higher energy QW transitions. These spectroscopic data make it possible to extract information about the excitonic line broadening mechanisms and the interfacial quality of the structures. For this purpose we examined the dependences of the broadening parameter Γ which represents the full width at half maximum (FWHM) of exciton lines on quantum number n for allowed heavy hole related excitonic transitions. Figure 2 shows the experimental data at (a) 300 and (b) 90 K for undoped and differently Be- or Si-doped MQWs with well width $L_{\rm w} = 15$ nm.

The experimental data (Fig. 2, symbols) can be modelled in terms of several broadening mechanisms: temperature-independent inhomogeneous broadening of the exciton energy levels ($\Gamma_{\rm inh}$) caused by structural imperfections in the MQWs, such as interface roughness, as well as temperature-dependent homogeneous broadening ($\Gamma_{\rm hom}$) due to exciton–phonon interaction and exciton scattering by ionized impurities and free carriers. The inhomogeneous broadening component $\Gamma_{\rm inh}$ due to well width fluctuations usually is estimated from the experimental dependences $\Gamma(n)$ because this component predominantly causes the increase of parameter Γ with n. Such behaviour of Γ can be understood considering $\Gamma_{\rm inh}$ as the change of the exciton energy E_n with the variation of the well width $L_{\rm w}$ [3],

$$\Gamma_{\rm inh}(n) = 2.36\,\frac{{\rm d}E_n}{{\rm d}L_{\rm w}}\,\delta L_{\rm w}\,, \tag{3}$$

where $\delta L_{\rm w}$ is the standard deviation of the well width fluctuations obeying a Gaussian distribution. In the case of QW systems with infinite potential barriers, the derivative ${\rm d}E_n/{\rm d}L_{\rm w}$ is proportional to the square of quantum index n and $L_{\rm w}^{-3}$ and can be expressed as

$$\frac{{\rm d}E_n}{{\rm d}L_{\rm w}} = \frac{\hbar^2\pi^2}{\mu L_{\rm w}^3}\,n^2\,, \tag{4}$$

where $\hbar$ is the Planck constant and μ marks the reduced mass of the electron and hole. For finite barrier QW systems the relation (4) does not hold exactly because the energies of excited states are usually lowered due to penetration of wave functions into barriers and also by the nonparabolicity effects. However, it clearly demonstrates that higher order optical transitions are more sensitive to the interface roughness in heterostructures than the ground state ones. We used this peculiarity of $\Gamma(n)$ to estimate the interface roughness in QWs for samples studied except for the Si doped sample L79 with undefined $\Gamma(n)$ dependence (Fig. 2(a), dashed line).

The values of $\delta L_{\rm w}$ characterizing the interface roughness in MQWs were evaluated from the broadening of the higher order quantum confined transitions by comparing experimental $\Gamma(n)$ dependences (Fig. 2(a, b)) with the relation

$$\Gamma(n) = a + \Gamma_{\rm inh}(n)\,, \tag{5}$$

where the parameter a contains all homogeneous broadening ($\Gamma_{\rm hom}$) contributions. In this analysis, the $\delta L_{\rm w}$ and a were treated as adjustable parameters while the derivatives ${\rm d}E_n/{\rm d}L_{\rm w}$ were calculated numerically using a finite square well potential approximation and taking into account the nonparabolicity of the energy bands.

From the fitting of experimental data to Eq. (5) (Fig. 2, solid lines), the well width fluctuations $\delta L_{\rm w}$ were found to vary in the range of 0.6–1.0 monolayer (ML) for different samples. The obtained $\delta L_{\rm w}$ values are typical of GaAs / AlAs QW systems and show good interface quality of the studied samples. Note that the highest $\delta L_{\rm w}$ value corresponds to the MQW structure 1807 with larger number of periods ($N = 100$) as compared to the structures consisting of 40 periods. This observation implies that, in addition to fluctuations within the same well, the variations in width from well to well can occur in the structures studied.

The broadening parameter Γ of the excitonic 11H transitions as a function of the doping density for different MQW samples at 300 and 90 K temperatures is shown in Figs. 3(a) and 3(b) respectively. As can be seen from the figure, for the doping density below $\sim 10^{10}$ cm^{-2}, the total line width of the 11H excitons is about 6.5 meV and 2 meV at 300 and 90 K, respectively. It should be noted that the evaluated inhomogeneous broadening, $\Gamma_{\rm inh} \leq 1$ meV, for 11H excitons is a nondominant spectral line broadening mechanism, especially at room temperature. Therefore, an additional homogeneous broadening mechanism must be invoked. The thermal exciton line broadening $\Gamma_{\rm th}$ estimated from the temperature dependence of exciton scattering by acoustic and optical phonons [14] is about 6 meV at

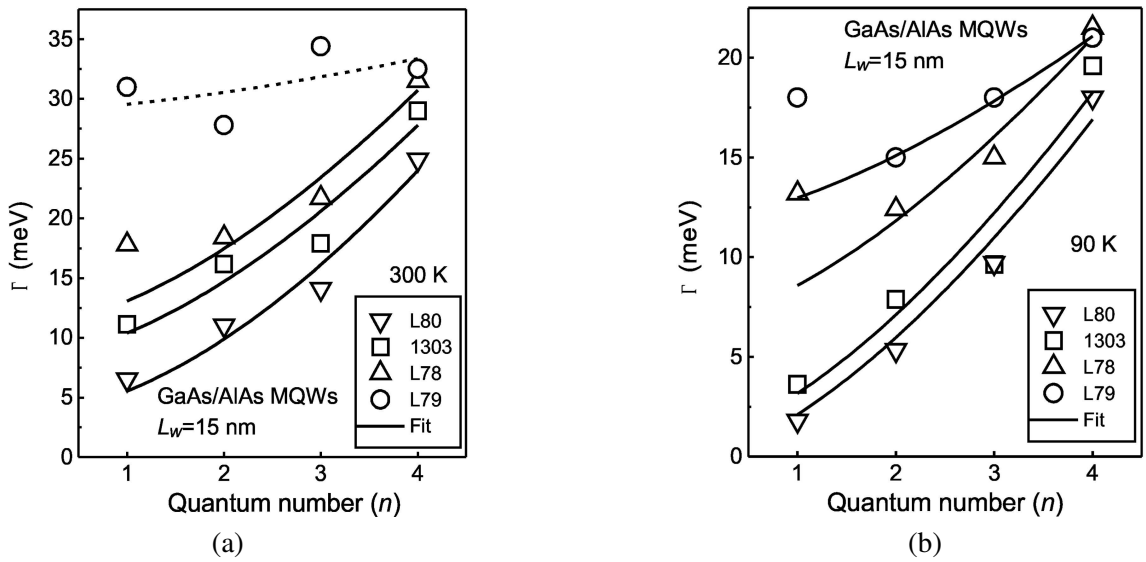

Fig. 2. The dependences on quantum number n of the spectral line width Γ (FWHM) of heavy hole related excitonic transitions determined from the line shape analysis of DSPV spectra at (a) 300 and (b) 90 K. Symbols represent experimental data, while the continuos lines are the result of the fit. The dashed line is a guide to the eye.

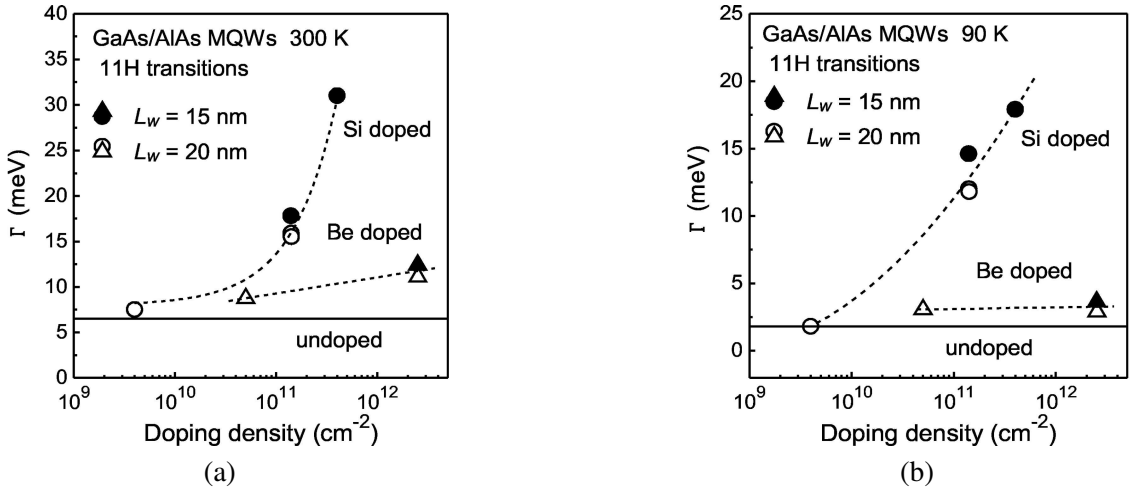

Fig. 3. The dependences on doping density of the spectral line width Γ(FWHM) of 11H excitonic transitions determined from the line shape analysis of DSPV spectra of Si and Be δ-doped GaAs / AlAs MQWs at (a) 300 and (b) 90 K. Symbols represent experimental data, while the dashed lines are guides to the eye.

300 K. Thus, the total line width, $\Gamma = \Gamma_{\rm th} + \Gamma_{\rm inh}$, is comparable to experimental Γ (6.5 meV) for undoped structure at the room temperature. These results imply that in weakly doped structures the exciton–LO phonon scattering should be the dominant mechanism of spectral line broadening at room temperature.

The line broadening mechanism in highly doped QWs that involve free carriers is more complex. A strong line width enhancement (Fig. 3) is characteristic of these MQWs. Such additional line broadening could occur due to the exciton scattering by ionized impurities and free carriers as well as due to many-body effects: screening of excitonic interaction and phase space filling. A subband filling effect mainly influences on the absorption related only to the lowest subband and, as expected, is more pronounced for n-type doped QWs. Therefore, in comparison with Be-doped MQWs, Si doping more effectively bleaches and broadens the ground state excitonic lines. As a result, distinctly different dependences of broadening parameter Γ on doping density for this type of samples are observed both at 300 and 90 K temperatures (Fig. 3). On the other hand, for Be-doped MQWs at 90 K the comparable broadening Γ values of the lowest energy exciton were found for samples 1807 and 1392 (Fig. 3(b), open triangles), despite of the very different acceptor densities. At the moment this result is not fully understood. Tentatively, it may be explained

by the localization of heavy holes at impurity fluctuation potentials in strongly doped QWs leading to the changes in the Coulomb screening.

4. Conclusions

Using the DSPV spectroscopy we have studied electronic structure and structural quality of Be and Si δ-doped GaAs / AlAs MQW structures. The energies and line broadening parameters for a large number of QW related excitonic transitions were determined from the line shape analysis of DSPV spectra. The transition energies were found to be in a good agreement with calculations within the envelope function approximation taking into account the nonparabolicity of energy bands. Analysis of the dependence of the exciton line width broadening on the quantum subband number allowed us to evaluate the line broadening mechanisms and interface roughness of the MQWs. The well width fluctuations were found to vary in the range of 0.6–1 ML showing therefore high structural quality of the studied samples. It was disclosed that doping with Si donors bleaches and broadens more effectively the lowest energy excitonic lines as compared to doping with Be acceptors. The results of these spectroscopic studies show that DSPV is a powerful technique for examining optical, electronic, and structural properties of δ-doped QW structures.

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GaAs / AlAs KVANTINIŲ DUOBIŲ SU PRIEMAIŠOMIS DIFERENCIALINIO FOTOVOLTINIO ATSAKO SPEKTROSKOPIJA

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Santrauka

Priemaišinių lygmenų kvantinėse duobėse inžinerija atveria naujas galimybes kurti terahercinių bangų emiterius bei detektorius, todėl nanodarinių charakterizavimas nesąlytiniais optiniais metodais yra svarbus, siekiant suprasti tokių prietaisų veikimo ypatumus. Pasitelkus diferencialinio fotovoltinio atsako spektroskopiją, buvo siekiama ištirti Be ir Si priemaišų turinčių ($4{\cdot}10^9$–$2{,}5{\cdot}10^{12}$ cm^{-2}) GaAs / AlAs (15; 20 nm / 5 nm) kvantinių duobių elektroninę sandarą, darinių struktūrinį tobulumą ir priemaišų poveikį eksitoniniams optiniams šuoliams.

Remiantis diferencialinių spektrų forma, buvo nustatyti eksitoninių šuolių, vykstančių tarp pagrindinių bei sužadintų valentinių ir laidumo kvantinių pajuosčių, parametrai bei jų priklausomybė nuo priemaišų tankio. Buvo rasta, jog optinių šuolių energijos gerai derinasi su apskaičiuotomis, atsižvelgiant į energijos juostų neparaboliškumą. Išanalizavus eksitoninių linijų išplitimo parametrų priklausomybes nuo pajuosčių kvantinio skaičiaus, nustatyti eksitoninių linijų išplitimo mechanizmai. Rasta, jog nehomogeninį linijos išplitimą lemia duobės pločio fliuktuacijos, kurių dydis įvairiems bandiniams kinta nuo 0,6 iki 1,0 monosluoksnio. Nustatyta, jog žemesniųjų energijų eksitonines linijas daug efektyviau slopina ir išplečia Si donorai, negu Be akceptoriai. Tyrimų rezultatai rodo, jog diferencialinio fotovoltinio atsako spektroskopija yra informatyvus bekontaktis metodas puslaidininkiniams kvantiniams dariniams su priemaišomis charakterizuoti.