STUDY OF EXCITONIC TRANSITIONS IN δ-DOPED GaAs/AlAs QUANTUM WELLS

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Received 11 June 2009; revised 9 September 2009; accepted 15 September 2009

Investigation of excitonic lines in differential surface photovoltage (DSPV) spectra of p-type (Be) δ-doped GaAs/AlAs multiple quantum well (MQW) structures is reported. From the lineshape analysis of the DSPV spectra, the energies and line broadening parameters for a large number of QW related excitonic transitions were determined. It is found that transition energies are in a good agreement with calculations carried out within the envelope function approximation taking into account the nonparabolicity of energy bands. Examining the dependence of the exciton linewidth on the QW thickness, the line-broadening mechanisms were revealed and interface roughness in the MQW structures was evaluated. An asymmetrical lineshape of certain excitonic transitions in SPV spectra of MQW structures was shown to be related to the Fano resonance.

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

PACS: 73.21.Fg, 78.55.Cr, 78.67.De

1. Introduction

The strong interest in δ-doped quantum wells (QWs) is motivated by the potential application of the nanostructures as active media in compact terahertz (THz) devices ([1] and references herein). The tunability of the energy levels of shallow impurities embedded in QWs opens up the possibility to vary the THz detection frequency by changing the profile of QW confining potential [2, 3]. It is expected that this particular material system with Be dopants in GaAs/AlAs heterostructures will provide detectors covering the wavelength range from 40 to 60 µm (7.5–5 THz). In GaAs, beryllium is an acceptor impurity-doped species commonly used in devices, and it is relatively stable with respect to diffusion. To find an optimal device design, it is of particular importance to know an electronic structure and physical properties of the QW structures, such as presence of imperfections, interface quality, and internal electric fields. The information needed can be obtained by modulation spectroscopy methods, which allow one to study QW structures with high spectral sensitivity. During the last years, we have employed the contactless methods of photoreflectance and differential surface photovoltage (DSPV) for the characterization of Be δ-doped GaAs/AlAs MQW structures designed for THz sensing applications [4–6]. So far the investigations have been mainly performed at room temperature. In this work, to gain further insight into the excitonic properties with increasing doping level, we extended our previous DSPV spectroscopy studies [4, 6] of Be δ-doped GaAs/AlAs multiple QW (MQW) structures to low temperature region of 90 K. The lineshape analysis of the DSPV spectra allowed us to extract information on the excitonic parameters for a large number of QW subbands. Special attention was paid to an exciton line broadening mechanism in MQWs. A high quality of the studied samples allows us to observe the Fano resonance effect [7] for definite excitonic lines in the SPV spectrum.

2. Samples and experiment

A series of Be δ-doped GaAs/AlAs MQWs were grown by MBE on a semi-insulating (100) GaAs substrate. Prior to the growth of the MQWs, a 300 nm thick GaAs buffer layer was grown. Each of the MQW structures investigated contained the same, 5 nm wide
Table 1. Characteristics of the samples: the repeated period, the quantum well width $L_w$, the $\delta$-doping Be concentration $N_A$, and the growth temperature of the epilayer $T$. $N_D$ signifies the $\delta$-doping Si concentration.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Periods</th>
<th>$L_w$ (nm)</th>
<th>$N_A$ (cm$^{-2}$)</th>
<th>$T$ (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1795</td>
<td>400</td>
<td>3</td>
<td>$2 \cdot 10^{10}$</td>
<td>550</td>
</tr>
<tr>
<td>2068</td>
<td>300</td>
<td>5</td>
<td>$5 \cdot 10^{10}$</td>
<td>550</td>
</tr>
<tr>
<td>1794</td>
<td>200</td>
<td>10</td>
<td>$5 \cdot 10^{10}$</td>
<td>550</td>
</tr>
<tr>
<td>1303</td>
<td>50</td>
<td>15</td>
<td>$2.5 \cdot 10^{12}$</td>
<td>540</td>
</tr>
<tr>
<td>1392</td>
<td>40</td>
<td>20</td>
<td>$2.5 \cdot 10^{12}$</td>
<td>540</td>
</tr>
<tr>
<td>1807</td>
<td>100</td>
<td>20</td>
<td>$5 \cdot 10^{10}$</td>
<td>550</td>
</tr>
<tr>
<td>L80</td>
<td>40</td>
<td>15</td>
<td>undoped</td>
<td>700</td>
</tr>
<tr>
<td>L29</td>
<td>40</td>
<td>20</td>
<td>Si: $N_D = 4 \cdot 10^9$</td>
<td>670</td>
</tr>
</tbody>
</table>

AIAs barrier, while each GaAs well layer was $\delta$-doped at the well centre with Be acceptors. The structures were capped with a 100 nm GaAs layer. For comparison, undoped and weakly Si $\delta$-doped MQWs were also studied. The doping level and the main characteristics of samples are summarized in Table 1. The SPV measurements were done in a chopped light geometry using a capacitor-like system with a transparent conducting top electrode under normalized incident light intensity conditions [8]. The illumination intensity was selected at sufficiently low levels in order not to affect the line shape of the spectra. In DSPV spectra measurements, a wavelength-modulation technique was used. The wavelength 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 SPV and wavelength-modulated DSPV signals were recorded by a conventional lock-in detection system. The measurements were performed at 90 K temperature.

3. Experimental results and discussion

3.1. DSPV spectra

Figure 1 shows the typical DSPV spectra for the Be $\delta$-doped GaAs/AIAs MQW structures (Table 1). At energies higher than band gap $E_g$ of GaAs (1.506 eV at 90 K), the spectra are dominated by the optical features $mnnH(L)$ which correspond to excitonic transitions in the MQW region of the samples. The notation $mnnH(L)$ signifies the transitions between the $m$th electron and $n$th heavy-hole (H) or light-hole (L) subbands. By considering excitonic features we assumed that the spectra of the normalized DSPV/SPV signal are qualitatively equivalent to the first derivative of the absorption spectra. The energies and broadening parameters of the optical transitions responsible for the observed DSPV features were determined by best fits of the DSPV spectra to the first derivative of a Lorentzian-type function [9]. As can be seen from Fig. 1, the experimental DSPV data (open circles) are reasonably well described by the adopted lineshape model (full lines). The optical transition energies (arrows in Fig. 1) were found to vary with the QW thickness $L_w$ as expected from the quantum confinement effect. The heavy- and light-hole related excitonic transitions are well resolved due to comparatively small broadening of the energy states at 90 K. Also, it was observed that the shift of excitonic features of $\sim 82$ meV with respect to their positions at room temperature [4] is in good agreement with the temperature shift of the GaAs bandgap energy.

3.2. Optical transition energies

To identify the optical features observed, we have carried out calculations of energy levels and interband transition energies for GaAs/AIAs QWs studied. The transition energies were primarily calculated for undoped rectangular QW both for parabolic and nonparabolic energy bands in the envelope function approximation [10]. We used the parameters of calculations employed in our previous work [4]. The calculations of energy levels by solving Poisson and Schrödinger equations have also been performed in order to study possible effects arising from the $\delta$-doping. It was found that the change in the ground state optical transition energy due to a distortion of the rectangular QW potential profile by $\delta$-doping is negligible and reaches only $\leq 3$ meV for the acceptor density of $2.5 \cdot 10^{12}$ cm$^{-2}$ in the 20 nm QW. For the narrower QWs and higher order optical transitions the deviations were found to be even smaller. It is worth noting that a solution of the coupled Poisson-Schrödinger equations has shown that the QWs with the acceptor density of $2.5 \cdot 10^{12}$ cm$^{-2}$ should not be degenerate.

The results of the calculation and experimental data are presented in Fig. 2. Symbols represent the experimental excitonic energies determined from the fitting procedure, while the lines correspond to theoretical results, obtained within the nonparabolic energy bands’ approximation. It should be noted that the calculated transition energies slightly differ from experimental ones for GaAs/AIAs MQW sample with $L_w = 3$ nm (Fig. 2). The well width $L_w$ of the narrow QWs can be slightly different from a nominal value due to a possible technological inaccuracy. However, generally, the experimental data are in quite good agreement with calculations. It was found that for most optical transitions studied the calculated interband energies ex-
Fig. 1. DSPV spectra of Be $\delta$-doped GaAs/AlAs MQWs for different well width $L_w$ measured at 90 K. The arrows show the excitonic transitions energies obtained from a best fit of experimental data (open circles) to the first derivative of a Lorentzian-type function (solid curves).

Fig. 2. The experimental (symbols) and calculated (lines) dependencies of interband transition energies on QW width $L_w$ for Be $\delta$-doped GaAs/AlAs MQWs. The results of calculations are presented for nonparabolic energy bands’ approximation.

Fig. 3. Experimental (open circles) and calculated (thick solid curve) linewidth $\Gamma$ of 11H excitonic transitions in Be $\delta$-doped GaAs/AlAs MQWs as a function of the well width $L_w$. The calculated contributions to the line broadening due to well width fluctuations $\Gamma_{inh}$ (thin solid curve), exciton interaction with phonons $\Gamma_\mathrm{th}$ (dotted line), and broadening induced by random electric fields of ionized impurities $\Gamma_\mathrm{St}$ (dashed curve) are also presented. In addition, broadening parameters of undoped and weakly Si-doped MQWs are shown for comparison (triangles).

3.3. Broadening mechanisms of the spectral lines

The DSPV spectra of Be $\delta$-doped GaAs/AlAs MQWs (Fig. 1) exhibit a well-resolved 11H feature associated with ground state QW excitonic transitions. The well-resolved optical feature allows to extract in-
formation about excitonic line broadening mechanisms and interfacial properties, which define the quality of the structures studied. The well width dependence of the broadening parameter $\Gamma$, which represents the full width at half maximum (FWHM) of the lowest energy exciton line, is presented by open circles in Fig. 3. In addition, experimental values of the broadening parameter for undoped and weakly Si-doped MQW samples are presented for comparison (open triangles). As can be seen, the linewidth for Be-doped samples decreases with the QW width from 31 to about 3 meV. However, the lowest $\Gamma$ value in Be-doped samples remains to be somewhat larger than $\Gamma$ in undoped and weakly Si-doped samples (~2 meV).

By analysing the experimental $\Gamma(L_w)$ dependences, several line broadening mechanisms were considered [13]: (i) thermal broadening ($\Gamma_{\text{th}}$) due to phonon scattering, (ii) inhomogeneous broadening of the exciton energy levels ($\Gamma_{\text{inh}}$) caused by structural imperfections, first at all by interface roughness in the QWs, and (iii) broadening due to the Stark effect in a random Coulomb field of ionized impurities ($\Gamma_{\text{St}}$).

Considering the thermal broadening contribution, we used the value of $\Gamma_{\text{th}} = 1$ meV (Fig. 3, dotted line) estimated from the temperature dependence of exciton scattering by phonons in GaAs/AlGaAs QWs [14].

The inhomogeneous line broadening $\Gamma_{\text{inh}}$ due to the well width fluctuations was evaluated as the change of the exciton energy $E_n$ with the variation of the well width [15], i.e.

$$\Gamma_{\text{inh}} = 2.36 \frac{dE_n}{dL_w} \delta L_w,$$

where $\delta L_w$ is the standard deviation of the well width fluctuations obeying a Gaussian distribution and $n$ is the quantum number of the QW subbands. The $\delta L_w$ in thick QWs, where 11H optical transitions are insensitive to a variation in $L_w$ (Fig. 2), were evaluated from the broadening of the higher order quantum confined transitions by examining experimental $\Gamma(n)$ dependences (see [4] for details). The well width fluctuations $\delta L_w$ were found to be equal to ~0.6, ~0.8, and ~1.0 monolayer (1 ML = 2.83 Å) in samples L29, 1392, and 1807, respectively. It should be noted that for $\delta L_w = 1$ ML, inhomogeneous broadening parameter $\Gamma_{\text{inh}}$ is 0.8 meV and 50 meV for the 20 and 3 nm QW widths, respectively. Thus, whereas $\Gamma_{\text{inh}}$ is smaller than the observed linewidth for the widest, 20 nm, QW, it is much larger than the linewidth for the narrowest, 3 nm, QW. The latter result indicates that actually $\delta L_w$ in narrower QWs must be smaller than 1 ML.

The well width fluctuations in narrow QWs were estimated by analysing the total $\Gamma(L_w)$ dependence as the sum $\Gamma = \Gamma_{\text{th}} + \Gamma_{\text{inh}} + \Gamma_{\text{St}}$ of partial contributions. To perform such analysis, the Stark broadening $\Gamma_{\text{St}}$ in a random Coulomb field of ionized impurities was estimated by the relation $\Gamma_{\text{St}} = 2 \times 10^{-30} n_i^{1/3} (m_0/\mu)^2 E_b^{-1}$ derived for the bulk, 3D, case in [16], where $n_i$ is the concentration of ionized impurities, $E_b$ is the exciton binding energy, $m_0$ is the free electron mass, and $\mu$ is the reduced electron-heavy hole mass. The experimental data for the Be-doped QWs (Fig. 3, open circles) were fitted reasonably by choosing $\delta L_w = 0.5$ ML (Fig. 3, thin solid curve representing $\Gamma_{\text{inh}}$) and $N_i = 8 \times 10^{16}$ cm$^{-3}$ (Fig. 3, dashed curve representing $\Gamma_{\text{St}}$). One may suppose that the smaller $\delta L_w$ values estimated in narrow QWs as compared to the thick ones could be due to the correlation effects of well width fluctuations in narrow QWs [17]. Note also, that concentration of ionized impurities $N_i = 8 \times 10^{16}$ cm$^{-3}$, deduced from the $\Gamma$-analysis, corresponds to a density $N_i \sim 1.6 \times 10^{10}$ cm$^{-2}$ in the weakly doped samples at 90 K. It should be noted as well that, contrary to the room temperature results [4,6], the broadening parameter $\Gamma$ only slightly depends on doping level at 90 K (see data in Fig. 3 of $L_w = 15$ and 20 nm, which correspond to 1807 and 1392 samples). To explain this effect, one can propose that Coulomb potential of the charged acceptors, located within the plane of $\delta$-layer, is screened at low temperature by free holes [18], and the exciton broadening by ionized impurities is suppressed.

3.4. Influence of Fano coupling on excitonic transitions

The remainder part is devoted to the study of the lineshape features of higher order excitonic transitions that are degenerate with the continuum of the excitonic states related with lower subband pairs. In this case the spectral changes of both the discrete lines and continuum can be expected due to Fano resonance effect [7]. For example, Fano resonance was predicted [19] for the weakly allowed 13H transition, but in comparison with experiment it was hampered by a large inhomogeneous broadening $\Gamma_{\text{inh}}$ introduced by fabrication process of the samples. We believe that a small $\Gamma_{\text{inh}}$ value revealed for the our widest 20 nm QW should allow us to observe the Fano resonance spectroscopically.

Figure 4 shows the absorbance spectrum for the Be-doped GaAs/AlAs MQW structure with 20 nm QW (Table 1, 1807sample), calculated from normalized SPV measurements. From an analysis of the spectrum it follows that the two lowest 11H and 11L peaks, corresponding to heavy- and light-hole ground state excitonic transitions, manifest a Lorentzian-like lineshapes. In contrast, the higher order excitonic transitions 13H,
Fig. 4. Absorption spectrum of Be $\delta$-doped GaAs/AlAs MQWs with well width $L_w = 20$ nm (sample 1807, Table 1) at 90 K. The arrows show the transition energies obtained from a best fit of experimental data (open squares) to Eq. (1) (solid curves). The inset shows an expanded view of the Fano resonance associated with the $13\,^H$ transition.

22H, and 33H exhibit asymmetric lineshapes and can be interpreted as Fano resonances arising from an interaction of these discrete excitonic states with the continuum of lower lying energy levels. The Fano line shape, with a dip below the continuum absorption on the low-energy side, is most distinctly pronounced for the $13\,^H$ exciton.

To characterize the spectral asymmetry of the $13\,^H$, $22\,^H$, and $33\,^H$ lines (Fig. 4), we have fitted the observed optical features to the phenomenological Fano line-shape function [7]

$$\alpha = \alpha_{\text{cont}} \frac{(q + \varepsilon)^2}{1 + \varepsilon^2},$$  

(1)

where $q$ is the ratio between the optical matrix elements of transitions to the discrete state and to the continuum, $\varepsilon$ is normalized energy defined by $\varepsilon = 2(E - \Omega)/\Gamma$, $\Omega$ is the energy of the corresponding Fano resonance, $\Gamma$ is the broadening (linewidth) of resonant state due to the Fano coupling, and $\alpha_{\text{cont}}$ describes the absorption of the continuum without coupling. Depending on the sign of $q$, which itself depends on the relative signs of the transition and coupling matrix elements, Eq. (1) describes a line shape with the dip on the low ($q > 0$) or high ($q < 0$) energy side of the peak. A fitting of the experimental data (squares) to the Fano function (1) is presented by curves in Fig. 4. The fitting shows that the $22\,^H$ and $33\,^H$ lines are almost symmetric ($q = -12$) and practically do not present a Fano resonance, while the $13\,^H$ line (see inset of Fig. 4) presents a very distinct Fano resonance ($q = 1.3$) indicating a strong coupling. This could be associated with a relatively weak oscillator strength of the forbidden $13\,^H$ transitions. Note also that $q$ is positive, i.e., the dip occurs below the exciton peak. The lineshape is in agreement with theoretical calculation [19] and experimental observations in other QWs [20].

4. Conclusions

In summary, DSPV spectra of Be $\delta$-doped GaAs/AlAs MQW structures have been measured at 90 K temperature. From the lineshape analysis of DSPV spectra the transition energies and line-broadening parameters for a large number of QW-related excitonic transitions were determined. The transition energies were found to be in a good agreement with the calculated ones for non-parabolic energy bands. It was revealed that in QWs thinner than 10 nm the dominant line broadening mechanism is due to the half-ML well-width fluctuations. In QWs thicker than 10 nm, the average well width fluctuations are found to be 0.6–1 ML. In thick QWs, the thermal broadening, the inhomogeneous line broadening due to well width fluctuations, and Stark broadening due to the random electric fields of the ionized impurities contribute to the observed line broadening $\Gamma$. In addition, the distinct behaviour of the Fano resonance of forbidden $13\,^H$ excitonic transitions in SPV spectra for 20 nm wide QWs was disclosed.

Acknowledgements

The work was supported, in part, by the Lithuanian State Science and Studies Foundation under contract C-07004/C-17/2009 project.

The research in the Semiconductor Physics Institute is conducted under topics Optical spectroscopy of composite nanostructures and quantum structures (158J) and Terahertz optoelectronics: Devices and applications (179J).

References


Santrauka
Plėtojant THz prietaisų technologiją, svarbu detaliai žinoti kvantinių duobių su priemaišomis energinę sandarą ir optines savybes. Šiame darbe diferencialinio paviršinio fotovoltinio atsakų metodą ištirti GaAs/AlAs kvantinių duobių su berilio priemaišomis eksitoninių optinių šuolių ypatumai. Bandinius sudarė skirtingo pločio $L_w$ GaAs duobių su 5 nm storio AlAs barjerais dariniai. Duobių centrineje srityje buvo įterptas Be akceptorių sluoksnis. Priemaišų tankis kito nuo $5 \times 10^{10}$ iki $2,5 \times 10^{12}$ cm$^{-2}$. Tyrimų rezultatai rodo, jog optinių bandinių atsaką lemia eksitoninių šuolių Fano efektas. Optinių šuolių prigimtis buvo atskleistaapskaičiavus energijos lygmenis perašos matricų metodu. Eksperimentinės optinių šuolių energijos vertęs dera su teorinėmis, apskaičiuotomisatsižvelgiant į energijos juostų neparabolikumą. Išanalizavus eksitoninių linijų išplitimo parametrų priklausomybes nuo duobės pločio bei priemaišų tankio, buvo nustatyti eksitoninių linijų išplitimo mechanizmai. Gauti rezultatai rodo, kad tirtose kvantinėse duobėse pasireiškia keletas linijų išplitimo veiksnių. Siaurose duobėse ($\leq 5$ nm) linijos išplitimą lemia kvantinių duobių pločio fluktuacijos ($\delta L_w \leq 0,5$ sluoksnio); platesnėse duobėse eksitonų linija išplinta visų pirma dėl sąveikos su fononais ir jonizuotomis priemaišomis.