ELECTROREFLECTANCE OF n-GaP AND n-GaAs EPITAXIAL FILMS

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Electroreflectance (ER) spectra of homoepitaxial n-GaP (111) films with the concentration of electrons $n = (1 \div 5) \times 10^{22}$ m^{-3} and homoepitaxial n-GaAs (100) films with $n = 10^{23} \div 10^{24}$ m^{-3} have been studied. The former films were studied in the spectral range 2.5—3.2 eV using the electrolytic technique, and the latter in the spectral range 1.3—1.65 eV using the Schottky barrier method. Measurements were carried out at room temperature and using unpolarized light. From the quantative analysis of the ER spectra, the following parameters were obtained: the electrooptical energy $\hbar\theta$, the surface electrical field F_s, the collisional broadening parameter Γ, and the relative phase factor ψ. The connection between the periods of Franz—Keldysh oscillations ΔE_m and the electrooptical energy $\hbar\theta$ has been analyzed.

1. Introduction

The ER spectroscopy, owing to its high resolution, is at the head of researches of the band structure of solids. In comparison with classical spectroscopy, the ER one is more sensitive to variations in the energy spectrum of a semiconductor. The practical advantage of this method is that, in order to determine both the optical and electrophysical properties of the near-surface layer of the investigated semiconductor, it will suffice only to create an electric field in it [1–4]. It is common knowledge that near-surface layers in semiconductors are the basic operation area in electronic devices of the new generation. The behavior of the physical processes in such layers may undergo substantial modifications in comparison with that in the bulk owing to changes of the band structure, the mobility of current carriers, and the time of their energy relaxation, as well as to the availability of the surface potential. It is the band bending and the mobility of current carriers that define the functionalities of electronic devices. The application of the ER effect allows the structural perfection and the electronic parameters of the near-surface layers to be monitored and the influence of physical and chemical treatment on the state of the surface to be revealed.

Gallium phosphide and arsenide, owing to their optical and electrophysical properties, are important compounds for practical use in electronic devices. GaP and GaAs are materials for the manufacture of radiation detectors, light-emitting diodes, photodiodes, lasers, Gunn generators, electrooptical light modulators, solar cells, etc. This is why both single crystals and epitaxial films of GaP and GaAs are studied intensively.

This work aims at monitoring the structural perfection of n-GaP and n-GaAs epitaxial films making use of ER modulation spectroscopy.

2. Experimental Method

The ER spectra of homoepitaxial n-Ga (111) films with the concentration of electrons $n = (1 - 5) \times 10^{22}$ m^{-3} were studied making use of the electrolytic technique. A 1 N aqueous solution of KCl served as electrolyte. Measurements were carried out in the spectral range 2.5—3.2 eV which includes the direct transitions E_0 ($\Gamma_{8v} - \Gamma_{6c}$) with the energy of 2.74 eV (the energy of the first extremum in the ER spectrum) and $E_0 + \Delta_0$ ($\Gamma_{7v} - \Gamma_{6c}$) with the energy of 2.84 eV in non-polarized light.

The measurements of homoepitaxial n-GaAs (100) films with the concentration of electrons $n = 10^{23} - 10^{24}$ m^{-3} were also fulfilled in non-polarized light, but making use of the metal—semiconductor Schottky barrier [1, 5, 6], fabricated by sputtering a semitransparent chrome layer onto the surface of the GaAs film. All measurements were carried out in the range of transition E_0 ($\Gamma_{8v} - \Gamma_{6c}$) in the spectral interval 1.3—1.65 eV.

The experimental results for the materials concerned were obtained at room temperature, at the frequency of the first modulation harmonic ($f = 2.2$ kHz), with a threshold sensitivity of 5×10^{-6}, and the spectral resolution of 3×10^{-3} eV.

3. Results and Their Discussion

Figure 1 exhibits the ER spectrum of an n-GaP epitaxial film with the concentration of free electrons $n = 3 \times 10^{22}$ m^{-3}. The polarity of ER extrema and the dependence of their amplitudes on the applied voltage evidence for the existence of a depletion layer near the surface. According to the ER data, the amplitude of the flat band potential is $\phi_{\text{fb}} = -1.8$ V.

Attention is drawn to the fact that the ER spectrum in Fig. 1 contains additional Franz—Keldysh oscillations which are imposed onto a rather weak ER signal stimulated by electron transitions from the valence band, which is split off owing to the spin-orbit interaction, into the conduction band. This testifies that the weak-field approximation [7] is not satisfied under the conditions of measurements, and theoretical calculations of the ER curve can be made only numerically making use of the generalized Airy functions [1, 5, 8].

In the framework of the one-electron theory, the variation of the real part of the dielectric permittivity

$$\Delta\varepsilon_1(\omega, F) = \varepsilon_1(\omega, F) - \varepsilon_1(\omega, 0),$$

which is caused by the electric field F, is equal to

$$\Delta\varepsilon_1(\omega, F) = \frac{B_j \theta^{1/2}}{\omega^2} G(\eta)$$

for a three-dimensional critical point of the type 3DM_0. Here, B_j is a constant; $G(\eta)$ the electrooptical function of the second kind which is expressed in terms of Airy functions that describe the one-dimensional motion of free current carriers in a uniform external electric field [1, 5, 8—11] ;

$$\eta = \frac{E_0 - \hbar\omega + i\Gamma}{\hbar\theta};$$

E_0 is the electron transition energy; $\hbar\omega = E$ the energy of a photon; Γ the collisional broadening parameter; $\hbar\theta = (e^2F^2\hbar^2/2\mu)^{1/3}$ the characteristic parameter of the theory of the Franz-Keldysh effect (the electrooptical energy); $\mu^{-1} = (m_e^*)^{-1} + (m_p^*)^{-1}$ the inverse reduced effective mass; and m_e^* and m_p^* are the effective masses of electrons and holes, respectively, which participate in the optical transition under consideration.

According to work [12], for the experimental ER spectrum obtained in the strong-field mode of measurements, the relation

$$E^2(E - E_0)\left(\frac{\Delta R}{R}\right) \sim \exp\left(\frac{2\Gamma(E - E_0)^{1/2}}{(\hbar\theta)^{3/2}}\right) \times$$

$$\times \cos\left[\psi + \frac{4}{3}\left(\frac{E - E_0}{\hbar\theta}\right)^{3/2}\right] \qquad (1)$$

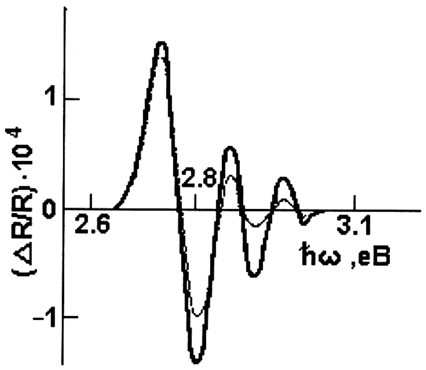

Fig. 1. ER spectrum of an n-Ga (111) epitaxial film with the concentration of electrons $n = 3 \times 10^{22}$ m^{-3}. The solid curve corresponds to the experimental data. The dashed curve is the result of theoretical calculations for $E_0 = 2.74$ eV, $\hbar\theta = 0.055$ eV, and $\Gamma = 0.034$ eV

is satisfied, where $\Delta R/R$ is the relative variation of the specimen reflectance in the electric field F, and ψ is the relative phase factor.

It stems from relationship (1) that, in the case of strong-field ER spectra, the energies E_m which correspond to the extrema of $|\Delta R/R|_m$ satisfy the equality

$$m\pi = \psi + \frac{4}{3}\left(\frac{E_m - E_0}{\hbar\theta}\right)^{3/2}, \qquad (2)$$

where m is the oscillation number, and E_m the energy coordinate of the oscillation extremum.

Relation (2) means that, knowing the slope of the dependence of the quantity $\frac{4}{3\pi}(E_m - E_0)^{3/2}$ on the oscillation number m, one can determine the value of $(\hbar\theta)^{3/2}$ and, hence, the electrooptical energy $\hbar\theta$ [13, 14]. In Fig. 2, the dependence of $\frac{4}{3\pi}(E_m - E_0)^{3/2}$ on the oscillation number m is plotted for the ER spectrum shown in Fig. 1. The slope of this dependence amounts to $(\hbar\theta)^{3/2} = 1.29 \times 10^{-2}$ eV$^{3/2}$. This corresponds to an electrooptical energy of 0.055 eV.

The surface electric field is

$$F_S = \left[\frac{2\mu(\hbar\theta)^3}{e^2\hbar^2}\right]^{1/2} \approx$$

$$\approx 5.125\left(\frac{\mu}{m_0}\right)^{1/2}(\hbar\theta)^{3/2} \times 10^9\ V/m = 2.1 \times 10^7\ V/m. \qquad (3)$$

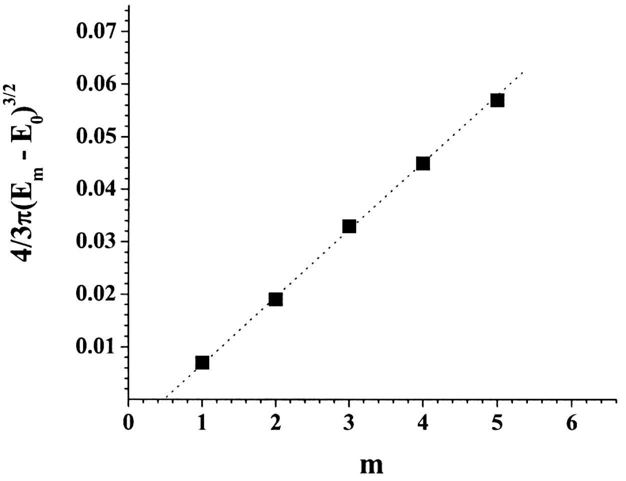

Fig. 2. Dependence of the quantity $\frac{4}{3\pi}(E_m - E_0)^{3/2}$ on the oscillation number m for an epitaxial n-Ga (111) film with the concentration of electrons $n = 3 \times 10^{22}$ m^{-3}

While calculating F_S, we used the following values of the effective masses of electrons and holes in GaP: $m_e^* = 0.126 m_0$ and $m_p^* = 0.5 m_0$ [15].

The collisional broadening parameter Γ was determined from the damping of Franz-Keldysh oscillations [1, 12, 16]. From relationship (1), it follows that the slope of the linear dependence of $\ln\left[E_m^2(E_m - E_0)\left|\frac{\Delta R}{R}\right|_m\right]$ on $(E_m - E_0)^{1/2}$ makes it possible to find the value of $2\Gamma/(\hbar\theta)^{3/2}$ and hence Γ. Really,

$$\ln\left[E_m^2(E_m - E_0)\left|\frac{\Delta R}{R}\right|_m\right] \sim \frac{2\Gamma\,(E_m - E_0)^{1/2}}{(\hbar\theta)^{3/2}} +$$

$$+\ln\left[\cos\left[\psi + \frac{4}{3}\left(\frac{E_m - E_0}{\hbar\theta}\right)^{3/2}\right]\right] =$$

$$= \frac{2\Gamma\,(E_m - E_0)^{1/2}}{(\hbar\theta)^{3/2}} + \ln(\cos m\pi) =$$

$$= \frac{2\Gamma\,(E_m - E_0)^{1/2}}{(\hbar\theta)^{3/2}} + \ln|\pm 1| = \frac{2\Gamma\,(E_m - E_0)^{1/2}}{(\hbar\theta)^{3/2}}.$$

Figure 3 depicts the dependence of $\ln\Big[E_m^2\,(E_m - E_0) \times \times \left|\frac{\Delta R}{R}\right|_m\Big]$ on $(E_m - E_0)^{1/2}$ for an epitaxial n-GaP (111) film with the concentration of free electrons

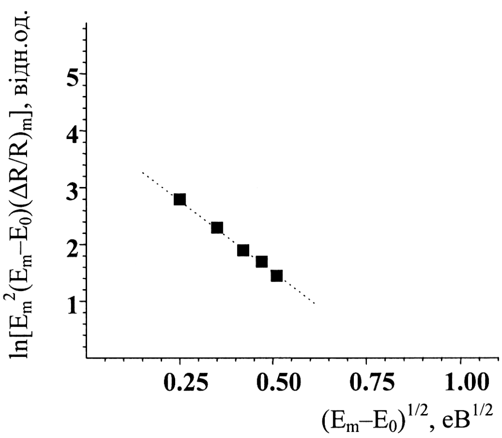

Fig. 3. Dependence of $\ln\left[E_m^2\,(E_m - E_0)\ \frac{\Delta R}{R}\ _m\right]$ on $(E_m - E_0)^{1/2}$ for an epitaxial n-GaP (111) film with the concentration of free electrons $n = 3 \times 10^{22}$ m^{-3}

$n = 3 \times 10^{22}$ m^{-3}. The slope of this dependence is $\frac{2\Gamma}{(\hbar\theta)^{3/2}} = 5.25$ eV$^{-1/2}$. This corresponds to the value of the collisional broadening parameter $\Gamma = 0.034$ eV.

Figure 1 shows the experimental data (the solid curve) together with the results of theoretical calculations (the dashed curve). A satisfactory agreement with experiment is reached for the following values of parameters: $E_0 = 2.74$ eV, $\hbar\theta = 0.055$ eV, and $\Gamma = 0.034$ eV.

Figure 4 demonstrates the ER spectrum of an epitaxial n-GaAs film with the concentration of electrons of 5×10^{23} m^{-3}, measured by the Schottky barrier method. The polarity of ER extrema, similarly to the case of the epitaxial n-GaP film, also points out that there exists a depletion layer near the surface. For the quantitative interpretation of the obtained data, it is necessary to take into account damped oscillations in the high-energy range of the spectrum, the period of which decreases as the energy grows and strongly depends on the applied electric field. Such features are characteristic of the strong-field mode of measurements. Therefore, in Fig. 4, the results of calculations carried out making use of the generalized electrooptical Airy functions [1, 5, 8] are presented (the dashed curve) together with the experimental data. A satisfactory agreement with the experiment is reached for the following values of parameters: $E_0 = 1.427$ eV, $\hbar\theta = 0.04$ eV, and $\Gamma = 0.028$ eV.

The parameter $\hbar\theta$ and the surface built-in electric field F_S were determined similarly to the case of epitaxial n-GaP films. The slope of the dependence of $\frac{4}{3\pi}(E_m - E_0)^{3/2}$ on the oscillation number m (Fig. 5) gives the

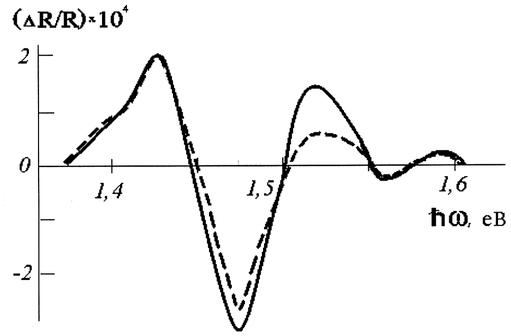

Fig. 4. ER spectra for an epitaxial n-GaAs film with the concentration of electrons of $n = 5 \times 10^{23}$ m^{-3}. The solid curve corresponds to the experiment; the dashed curve to theoretical calculations with the parameters $E_0 = 1.427$ eV, $\hbar\theta = 0.04$ eV, and $\Gamma = 0.028$ eV

evaluation $(\hbar\theta)^{3/2} = 0.8 \times 10^{-2}$ eV$^{3/2}$; whence, $\hbar\theta = 0.04$ eV. The surface electric field $F_S = 9.8 \times 10^6$ V/m. While calculating F_S by formula (3), the following values of the effective masses of electrons and holes in GaAs were used: $m_e^* = 0.065m_0$ and $m_p^* = 0.475m_0$ [17].

According to Eq. (2), the period of the second oscillation is

$$\Delta E_2 = E_2 - E_1 = \left[\left(\frac{3}{4}(2\pi - \psi)\right)^{2/3} - \right.$$

$$\left. - \left(\frac{3}{4}(\pi - \psi)\right)^{2/3}\right] \times \hbar\theta, \tag{4}$$

where E_1 and E_2 are the energy positions of the first and second oscillations, respectively, and

$$\Delta E_1 = E_1 - E_0 = \left[\left(\frac{3}{4}(\pi - \psi)\right)^{2/3}\right] \times \hbar\theta. \tag{5}$$

The periods of higher-order oscillations are found analogously:

$$\Delta E_3 = E_3 - E_2 = \left[\left(\frac{3}{4}(3\pi - \psi)\right)^{2/3} - \right.$$

$$\left. - \left(\frac{3}{4}(2\pi - \psi)\right)^{2/3}\right] \times \hbar\theta, \tag{6}$$

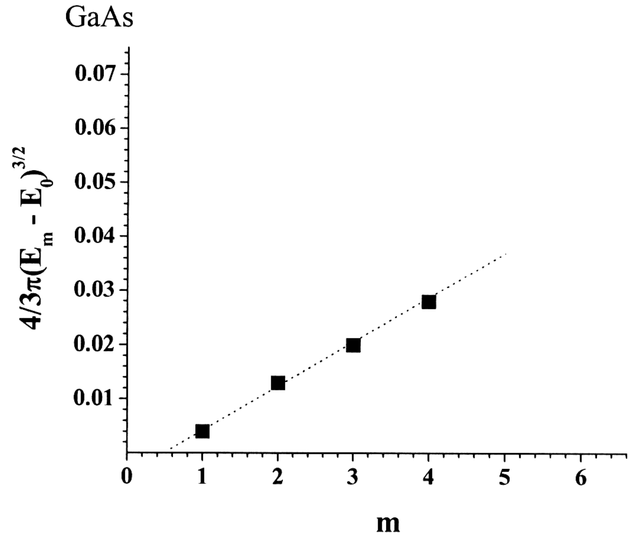

Fig. 5. Dependence of $\frac{4}{3\pi}(E_m - E_0)^{3/2}$ on the oscillation number m for an epitaxial n-GaAs (100) film with the concentration of electrons $n = 5 \times 10^{23}$ m^{-3}

$$\Delta E_4 = E_4 - E_3 = \left[\left(\frac{3}{4}(4\pi - \psi)\right)^{2/3} - \right.$$

$$\left. - \left(\frac{3}{4}(3\pi - \psi)\right)^{2/3}\right] \times \hbar\theta, \tag{7}$$

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$$\Delta E_m = E_m - E_{m-1} = \left[\left(\frac{3}{4}(m\pi - \psi)\right)^{2/3} - \right.$$

$$\left. - \left(\frac{3}{4}([m-1]\pi - \psi)\right)^{2/3}\right] \times \hbar\theta, \tag{8}$$

where $m = 2, 3, 4 \ldots$.

According to our experimental data, the value of ψ calculated from the extrapolation of the straight lines in Figs. 2 and 5 and using formula (2) is equal to $\pi/2$. The modulation spectrum $\Delta R/R$ is known to possess a sharp extremum at the energy of the gap width E_0, quickly decays below E_0 (in the classically forbidden energy region $\hbar\omega < E_0$), and oscillates above E_0 (the classically allowed region for the photon energies $\hbar\omega > E_0$) [1,2,8,10].

In the case $\psi = \pi/2$, formulae (4)—(8) look like

$$\Delta E_1 = E_1 - E_0 = \left(\frac{3\pi}{8}\right)^{2/3} \hbar\theta, \tag{9}$$

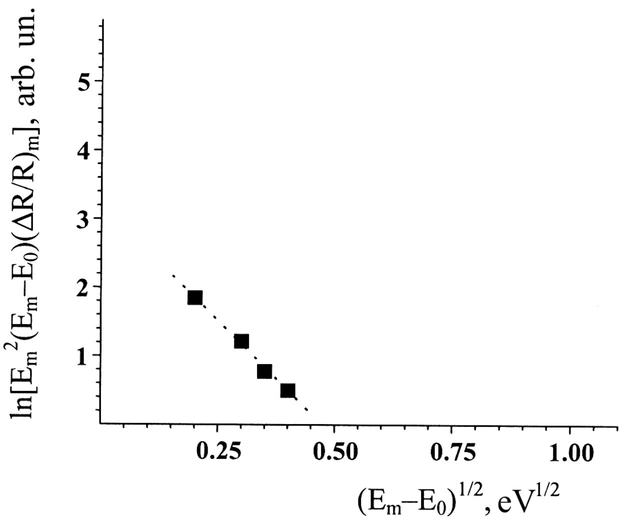

Fig. 6. Dependence of $\ln\left[E_m^2\left(E_m-E_0\right)\frac{\Delta R}{R}_m\right]$ on $(E_m-E_0)^{1/2}$ for the epitaxial n-GaAs (100) film with the concentration of free electrons $n=5\times10^{23}$ m^{-3}

$$\Delta E_2 = E_2 - E_1 = \left[\left(\frac{9\pi}{8}\right)^{2/3} - \left(\frac{3\pi}{8}\right)^{2/3}\right] \times \hbar\theta, \quad (10)$$

$$\Delta E_3 = E_3 - E_2 = \left[\left(\frac{15\pi}{8}\right)^{2/3} - \left(\frac{9\pi}{8}\right)^{2/3}\right] \times \hbar\theta, \quad (11)$$

$$\Delta E_4 = E_4 - E_3 = \left[\left(\frac{21\pi}{8}\right)^{2/3} - \left(\frac{15\pi}{8}\right)^{2/3}\right] \times \hbar\theta, \quad (12)$$

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$$\Delta E_m = E_m - E_{m-1} =$$

$$= \left\{\left[\frac{3(2m-1)\pi}{8}\right]^{2/3} - \left[\frac{3(2m-3)\pi}{8}\right]^{2/3}\right\} \times \hbar\theta. \quad (13)$$

On the basis of formulae (9)—(13), we obtain the approximate relations $\Delta E_1 = 1.115\hbar\theta$, $\Delta E_2 = 1.205\hbar\theta$, $\Delta E_3 = 0.94\hbar\theta$, $\Delta E_4 = 0.82\hbar\theta$, $\Delta E_5 = 0.745\hbar\theta$, and so on.

On the basis of the asymptotic form for the strong-field limit of the electrooptical functions, the authors of work [18] showed that the period of the third oscillation is $\Delta E_3 = 0.94\hbar\theta$ (formula (11)). Our measurements gave $\Delta E_3 = 0.052$ eV for the epitaxial n-GaP film and 0.038 eV for the epitaxial n-GaAs film (Figs. 1 and 4), which is in a good agreement with the results obtained in work [18]. Relationships (9)—(13) are very important for the definition of the electrooptical energy directly from the experimental ER curve, because, in this case, one should not plot dependences (2) which are shown in Figs. 2 and 5. The table quotes the values of ΔE_m, which were determined from Figs. 1 and 4, and $\hbar\theta$, which were calculated by formulae (9)—(13), for epitaxial GaP and GaAs films. The data cited in the table confirm the validity of the use of formulae (9)—(13).

The collisional broadening parameter was found in the same way as while analyzing the ER spectrum of the epitaxial n-GaP film. In Fig. 6, the dependence of $\ln\left[E_m^2\left(E_m-E_0\right)\left|\frac{\Delta R}{R}\right|_m\right]$ on $(E_m-E_0)^{1/2}$ is shown for the epitaxial n-GaAs (100) film with the concentration of free electrons $n=5\times10^{23}$ m^{-3}, which was plotted according to the data of Fig. 4. The slope of this dependence is equal to $\frac{2\Gamma}{(\hbar\theta)^{3/2}} = 7$ eV$^{-1/2}$; whence, $\Gamma = 0.028$ eV.

On the basis of the Heisenberg uncertainty principle for the energy E and the time t ($\Delta E\Delta t \geq \hbar$), relaxation effects occurring when a crystal absorbs light are described [1] using the phenomenological parameter of broadening Γ which is connected with the energy relaxation time τ of photo-induced current carriers by the relation $\Gamma = \hbar/\tau$. This relationship allows the value of τ for corresponding electron transitions to be estimated using the optical ER method. Therefore, we obtain $\tau = \hbar/\Gamma = 1.94\times10^{-14}$ s for the epitaxial n-GaP (111) film with the concentration of free electrons $n = 3\times10^{22}$ m^{-3}, and $\tau = 2.35\times10^{-14}$ s for the epitaxial n-GaAs (100) film with the concentration of free electrons $n = 5\times10^{23}$ m^{-3}. Using the empirical dependence of the electron mobility $\mu_e(\Gamma)$ [19], we estimate $\mu_e = 0.32$ m^2/(V × s) for the epitaxial n-GaAs film with the concentration of electrons $n = 5\times10^{23}$ m^{-3}.

The values of ΔE_m and $\hbar\theta$ for epitaxial n-GaP (111) films with the concentration of electrons $n = 3\times10^{22}$ m^{-3} and epitaxial n-GaAs (100) films with $n = 5\times10^{23}$ m^{-3}

GaP			GaAs		
m	ΔE_m, eV	$\hbar\theta$, eV	m	ΔE_m, eV	$\hbar\theta$, eV
1	0.061	0.0547	1	0.045	0.04035
2	0.066	0.05477	2	0.048	0.03983
3	0.052	0.05532	3	0.038	0.04042
4	0.045	0.05488	4	0.033	0.04024
5	0.041	0.05503			

4. Conclusions

1. The experimental results of our researches of homoepitaxial n-GaP (111) films with the concentration of electrons of $(1-5)\times 10^{22}$ m^{-3} and homoepitaxial n-GaAs (100) films with the concentration of electrons of $10^{23}-10^{24}$ m^{-3} using the ER method showed that the experimental ER spectra of epitaxial n-GaP and n-GaAs films are well described by the one-electron theory.

2. The following electronic parameters of the films were obtained: the electrooptical energy $\hbar\theta$, the surface electric field F_S, the collisional broadening parameter Γ (analyzing the damping of Franz—Keldysh oscillations), and the relative phase factor ψ (on the basis of the quantitative analysis of the ER spectra). The analysis of coupling between the periods of Franz—Keldysh oscillations ΔE_m and the electrooptical energy $\hbar\theta$ has been carried out, which allowed the value of $\hbar\theta$ to be determined directly from the ER spectrum.

3. The values of the collisional broadening parameter Γ, which were obtained for GaP ($\Gamma = 0.034$ eV) and GaAs ($\Gamma = 0.028$ eV) epitaxial films, evidence for their high perfection. The obtained value of current carrier mobility $\mu_e = 0.32$ m^2/(V $\times$ s) in GaAs films with the concentration of electrons of 5×10^{23} m^{-3} confirms this conclusion.

4. The shape of the ER spectrum of the epitaxial n-GaP (111) film measured using the electrolytic technique is similar to that of the ER spectrum of the epitaxial n-GaAs (100) film measured by the Schottky barrier method.

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ЕЛЕКТРОВІДБИТТЯ ЕПІТАКСІЙНИХ ПЛІВОК n-GaP ТА n-GaAs

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Резюме

Досліджено спектри електровідбивання (ЕВ) гомоепітаксійних плівок n-GaP (111) з концентрацією електронів $n = 10^{22} \div 5\times 10^{22}$ м$^{-3}$ з використанням електролітичної методики в спектральному діапазоні 2,5—3,2 еВ та гомоепітаксійних плівок n-GaAs (100) з концентрацією електронів $n = 10^{23} \div 10^{24}$ м$^{-3}$ з використанням бар'єра Шотткі в спектральному діапазоні 1,3—1,65 еВ. Вимірювання проведено при кімнатній температурі в неполяризованому світлі. Із кількісного аналізу спектрів ЕВ отримано значення електрооптичної енергії $\hbar\theta$, поверхневого електричного поля F_S, зіштовхувального параметра розширення Γ та відносного фазового фактора ψ. Проведено аналіз зв'язку між періодами осциляцій Келдиша—Франца ΔE_m і електрооптичною енергією $\hbar\theta$.