ABNORMALLY HIGH MOBILITY OF CHARGE IN Al$_x$Ga$_{1-x}$As: C/GaAs (100) HETEROSTRUCTURES

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Abstract. Using structural and spectroscopic methods, we have studied epitaxial layers of solid solutions Al$_x$Ga$_{1-x}$As with n-conductivity obtained by means of MOS-epitaxy. During carbon doping of solid solutions Al$_x$Ga$_{1-x}$As at $1.2-6.7\times10^{17}$cm$^{-3}$, the mobility for this concentration of impurities and composition was shown to be anomally high and twice as high as the calculation value. It was assumed that the arranged position of carbon in a metal sublattice of the solid solution causes changes in the average distance between the ions of the impurities, i.e. an increase in the length of a free path of a charge carrier and thus of its mobility.

The identified effect has practical implications for different technological methods of increasing the performance of functional elements of the contemporary optoelectronic base. Physical properties of the materials due to the use of the effects of an abnormally high mobility of charge carriers in the epitaxial layer of a heteropair opens up new possibilities in creating new Al$_x$Ga$_{1-x}$As-based structures.

Keywords: epitaxial heterostructures, abnormally high mobility of charge carriers, semiconductors, A$_3$B$_5$.

INTRODUCTION

Heterostructures Al$_x$Ga$_{1-x}$As/GaAs are fundamental elements of most optoelectronic devices these days such as solid semiconductor lasers, photodetectors, solar elements, etc. The major factor in the use of heterostructures is a high quality of crystal lattices of heteropairs.

GaAs and Al$_x$Ga$_{1-x}$As used to be profoundly investigated but these days there are a few research applications of these semiconductors [1—5]. First, the growing interest is caused by the modernization of the growth methods of these structures where the major one is MOCVD. At the level of technology and structure, the contemporary optoelectronic industry seeks a better insight into the optimization of the characteristics of the final device in terms of the growth parameters used and major properties of semiconductor materials. MOCVD provides accurate control of the thickness of a layer and assists the doping of epitaxial layers with high concentrations of different kinds of impurities [6—8].

Excellent electrophysical properties of GaAs-alloys allows the production of high-frequency devices, more optimal than silicon-based ones. Highly alloyed layers of A$_3$B$_5$ semiconductors are commonly used as fundamental or contact layers of bipolar ballistic charge transfer heterotransistors. High conductivity of contacts and low resistance of these layers improves the performance of corresponding optoelectrical devices. Doping using different impurities causes dramatic changes in the energy composition of semi-conductor epitaxial solid A$_3$B$_5$ solutions and thus impacts electrical properties of designed devices [9—12].

It should be noted that one of the most important parameters in the development of semi-conductor devices based on epitaxial solid solutions is mobility of charge carriers [13—14]. The mobility of charge carriers is known to be in direct proportion with dis-
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dispersion depending on a level of an impurity introduced at room temperature and is largely determined by the dispersion of charge carriers on a lattice and ionized impurity (Coulomb scattering).

Hall mobility of electrons for gallium arsenide is given by the expression [15]:

$$
\mu = \frac{\mu_0}{\left(1 + 10^{-17}n\right)^{\frac{1}{2}}},
$$

(1)

where $\mu_0 \approx 9400$ (cm$^2$ V$^{-1}$ s$^{-1}$), $n$ is Hall concentration of impurity.

In turn, the dependence of the mobility of charge carriers (electrons) on their concentration in epitaxial solid $\text{Al}_x\text{Ga}_{1-x}\text{As}$ solution can be specified by the following room temperature ratio (300 K) [16]:

$$
\mu = \frac{\mu_0}{1 + \left(\frac{n}{n_0}\right)^{\frac{1}{m}}},
$$

(2)

where $n$ is Hall concentration of the impurity; $n_0 = 10^{17}$ cm$^{-3}$; $m=0.4$; $\mu_0 = 7600$ cm$^2$ V$^{-1}$ s$^{-1}$ (x=0.10); $\mu_0 = 5800$ cm$^2$ V$^{-1}$ s$^{-1}$ (x=0.20); 4600 cm$^2$ V$^{-1}$ s$^{-1}$ (x=0.25); 3700 cm$^2$ V$^{-1}$ s$^{-1}$ (x=0.30); 3000 cm$^2$ V$^{-1}$ s$^{-1}$ (x=0.40).

The simplest analysis of the ratios (1) and (2) indicate that the mobility of charge carriers is inverse to their concentration, i.e. decreases as the latter increases.

Given the modern tendencies of the development of material technology for opto- and nanoelectronics, studies of possible changes towards increasing mobility of carriers is of great interest. The development of methods and technologies for improving the performance of electronic devices is in high demand these days. It is known to be addressed in different ways among which are decreasing basic microchip elements, development of multi-processor and multi-nuclear technology, development of electronic components based on new materials with improved characteristics and enhancing the mobility of charge carriers [17, 18]. It should be noted that the best way to address the above is to use the fourth solution.

Therefore, the objective of the paper is to carry out a study of ways of increasing the mobility of charge carriers in heterostructures based on solid $\text{Al}_x\text{Ga}_{1-x}\text{As}$ solutions as well as of structural, optical and energy properties of the materials.

1. OBJECTS AND METHODS OF THE STUDY

The samples were heterostructures $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}(100)$ grown to control the quality of epitaxial $\text{AlGaAs}$ layers not purposefully alloyed. The heterostructures were grown in MOCVD of hydride epitaxy EMCORE GS3/100 in a vertical reactor with a high rotation velocity of susceptor. The temperature of susceptor was maintained at 700 °C, the pressure in the reactor was 77 torr, the rotation velocity of susceptor was 1000 turns/min. The initial reagents were trimethyl gallium $\text{Ga(CH}_3)_3$, trimethylaluminum $\text{Al(CH}_3)_3$ and arsine $\text{AsH}_3$. The thickness of an epitaxial layer of both samples was ~ 2 mkm. The concentration of the carriers was determined by the six-probe method by Hall at room temperature (see Table 1).

<table>
<thead>
<tr>
<th>Table 1. Composition and growth conditions of heterostructures $\text{Al}<em>x\text{Ga}</em>{1-x}\text{As}:\text{C} / \text{GaAs}(100)$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Sample</strong></td>
</tr>
<tr>
<td>EM2889</td>
</tr>
<tr>
<td>EM2887</td>
</tr>
</tbody>
</table>

The structural quality of the heterostructures and parameters of the lattices of the solid solutions were identified using X-ray diffusion on a diffractometer DRON 4—07.

Infrared reflection spectra of the heterostructures were obtained using infrared spectrometer Vertex-70 Bruker. The concentrations of the elements in the solid solution were made clear using the method of X-ray spectra analysis by means of Oxford Instruments device for the electron microscope JEOL.

Photoluminiscence spectra of the heterostructures in the range of 750—1000 nm were excited by a semi-conductor laser modulus KLM-660/80 with the wavelength 660 nm and power 80 μWatt and in the range of 600—800 nm by a semiconductor laser with the wavelength 532 nm and power 100 μWatt were recorded using a diffraction monochromator MDR-4 and photo receiver with a Toshiba CCD camera TCD-1304DG. The measurements were taken at the temperature of liquid nitrogen.
2. RESULTS OF THE RESEARCH AND DISCUSSION

Structural and microstructural research

Using the method of X-ray spectral microanalysis, the concentrations of the elements making up the investigated epitaxial films. In order to analyze the accelerating stress of electrons of 20 kilovolts and areas of the range 750×750 mkm were examined. The effective depth of the microanalysis was about ~0.5 mkm. The X-ray spectral microanalysis data are in Table 1. It should be noted that due to special features of the method of X-ray spectral microanalysis, there was no exact information about the concentration of carbon, however the results showed fractions of atomic percent of it to be present in epitaxial films.

The measurement of the concentrations as well as mobility of free charge carriers in the obtained epitaxial films at room temperature showed the Hall concentration of impurities being 1.2—6.7·10¹⁷сm⁻³, which is typical for regular doping. However, mobility of charge carriers was anomalously high for the concentration, which was proved by the calculation based on the ratios (1) and (2). As Table 1 suggests, the obtained high level of mobility of charge carriers is almost twice as high as the calculation value for the specified concentration.

In order to identify changes occurring in a crystal grid of epitaxial solid solutions AlₓGa₁₋ₓAs with anomalously high mobility and at different levels of concentration of aluminium in the metal subgrid, the parameters of crystal grids of solid solutions must be very precise.

For our research we have chosen a line (600) in the range from 140° to 144°. Despite its low intensity, due to high dispersion of reflection at large Bragg angles, there is a significant division of diffracted Co Kα₁ radiation by the film and sublayer and the contribution of the sublayer into the distribution of the intensity of the line (600) is much less. The profiles of X-ray (600) diffraction of the samples of the heterostructures EM2889 and EM2887 are illustrated in Fig. 1.

The obtained experimental data indicate that diffraction of the samples due to the identical parameters of the grid of the film and sublayer of the heterocouple is overlay of Ka₁ lines of the sublayer and solid solution. In particular, the parameter of the crystal grid of the epitaxial solid solution of the sample EM2889 is in good agreement with the parameter of the sublayer GaAs(100) a=5.6532 Å, and diffraction of this heterostructure is a reflex positioned at the same angle as diffraction of GaAs(100).

Based on the data of X-ray spectral analysis, half-widths of diffraction lines were determined, interplanar spacings and the parameters of the crystal grids in the direction of the growth were calculated. The results of X-ray diffraction are indicated in Table 1.

The constant of the lattice of solid solutions a with a cubic symmetry considering elastic stresses in the heteroepitaxial layer according to the linear theory of plasticity can be found as [19—20]:

\[
a'' = a + \frac{1 - v}{1 + v} a^2 \frac{2v}{1 + v},
\]

Fig. 1. XRD of heterostructures AlₓGa₁₋ₓAs /GaAs (100) with anomalously high mobility of charge carriers.
where \( n \) are Poisson’s coefficients for epitaxial layers.

Due to coherent growth Al\(_x\)Ga\(_{1-x}\)As on the sublayer GaAs(100), the parallel component of the epitaxial layer \( a^\| \) can be replaced by the parameter of the crystal grid GaAs: \( a^\|=a_{\text{GaAs}}=5.6532\text{Å} \).

Therefore, the ratio (3) for the parameter of the lattice of the epitaxial layers Al Ga\(_x\)As given the linear dependence for Poisson’s coefficient:

\[
\nu_{Al, Ga_x As} = x\nu_{AlAs} + (1-x)\nu_{GaAs}
\]

can be written as

\[
\frac{\frac{a^\|_{Al, Ga_x As}}{a^\|_{GaAs}}}{1} = \frac{\nu_{Al, Ga_x As} + (1-x)\nu_{GaAs}}{\nu_{AlAs} + (1-x)\nu_{GaAs}}
\]

(4)

Based on the data of X-ray spectral diffraction and microanalysis as well as Vegard’s linear law for the system Al\(_x\)Ga\(_{1-x}\)As [20], we have calculated the parameters of the crystal lattice of non-stressed solid solutions obtained using the method of MOS-hydride epitaxy. As shown in Table 2, these parameters are not in agreement with the experimental ones, which seems to be due to the concentration of the carbon impurity in the solid solution.

In our previous papers [5, 12] we made a point that carbon can be in the position of regular nodes in the arsenic sublattice in the crystal grid of GaAs-type semiconductors and in high concentrations of the carbon acceptor introduced into the solid solution using the method of MOS-hydride autoepitaxy at low temperatures fourfold solid solutions are created (Al\(_x\)Ga\(_{1-x}\)As\(_{1-y}\)C\(_y\)).

### Table 2. Results of PL spectroscopy. Energy emission peaks in the photoluminescence spectra of heterostructures

<table>
<thead>
<tr>
<th>Sample/composition</th>
<th>Additional PL band</th>
<th>(Al(<em>x)Ga(</em>{1-x})As(_{1-y})C(_y)) Solid solution band</th>
</tr>
</thead>
<tbody>
<tr>
<td>EM2889 (Al(<em>{0.10})Ga(</em>{0.90})As)(<em>{0.9996})C(</em>{0.0004})</td>
<td>1.77</td>
<td>1.65</td>
</tr>
<tr>
<td>EM2887 (Al(<em>{0.26})Ga(</em>{0.74})As)(<em>{0.9997})C(</em>{0.0003})</td>
<td>1.89</td>
<td>1.73</td>
</tr>
</tbody>
</table>

Therefore the calculation of the parameters of the lattice considering the interior stresses for low-temperature epitaxial heterostructures Al\(_x\)Ga\(_{1-x}\)As: C/GaAs(100) can be performed using the methods described in [12] and the general Vegard’s law for a fourfold system of solid solutions (Al\(_x\)Ga\(_{1-x}\)As)\(_{1-y}\)C\(_y\) which will take the following form considering linear approximation:

\[
a(x, y) = a_{Al, Ga_x As}(1-y) + a_{diamond}y = (5.65325 + 0.0078x)(1-y) + 3.5670y \]

(5)

where \( x \) is the concentration of arsenic atoms, \( y \) is the concentration of carbon atoms in a fourfold solid solution and \( a_{diamond} \) is the parameter of the diamond lattice.

During the calculations it was assumed that Poisson’s coefficients for fourfold solid solutions can be found in the same way as Vegard’s law is specified:

\[
\nu(x, y) = \nu_{Al, Ga_x As}(1-y) + \nu_{diamond}y = (0.255x + 0.312(1-x)(1-y) + 0.103y
\]

(6)

Poisson’s coefficient for diamond \( \nu_{diamond} \) is taken from [22].

This approach allows not only the calculation of the parameters of the lattice for the samples of Al\(_x\)Ga\(_{1-x}\)As: C, but also to determine the coefficients of doping acceptor impurity, i.e. carbon (Table 2). The calculation showed that the carbon content in the epitaxial film should be 0.05 atom%, which is enough to create a solid solution (Al\(_x\)Ga\(_{1-x}\)As\(_{1-y}\)C\(_y\)) according to a complex mechanism. The achieved level of the impurity concentration was a lot lower (see Table 2) than that that was previously observed in [12] where the samples were obtained by means of MOS-hydride autoepitaxy at lower temperatures and the concentration was within ~0.5 atom % with the atoms of the acceptor impurity concentrating on the defects of the crystal grid of the epitaxial solid solution resulting in nanoclusters.

**Photoluminescence spectroscopy**

The formation of carbon solid solutions inevitably affects energy characteristics of the material, thus the use of photoluminescence spectroscopy for analyzing the energetic composition of the samples is understandable.

Photoluminescence spectra of the samples in Fig. 2 were obtained at the liquid nitrogen temperature in the
energy area corresponding with the area-area transition for threefold solid solutions Al$_{x}$Ga$_{1-x}$As.

As seen from the results, in the photoluminescence spectrum of the solid solution (Al$_{0.10}$Ga$_{0.90}$As)$_{0.9996}$C$_{0.0004}$ of the heterostructure EM2889 there is intensive emission line localized in the energy area 1.77 eV and displaced related to it, a low intensive wide strap with the centre near 1.65 eV. As for the photoluminescence spectrum of the epitaxial film (Al$_{0.26}$Ga$_{0.74}$As)$_{0.9997}$C$_{0.0003}$ (heterostructure EM2887), there is high intensive emission in the area 1.73 eV and two low intensive maximum localized near 1.89 eV and 1.62 eV. Second of them is due to PL of Al$_{0.26}$Ga$_{0.74}$As solid solution.

Based on the interpoly scheme [23] suggested for Vegard’s law of the fourfold solid solution (Al$_{x}$Ga$_{1-x}$As)$_{1-y}$C$_{y}$ we can similarly write the dependence of the width of the forbidden band on the atom concentration — Kuffala equation:

$$E_{g(Al_{x}Ga_{1-x}As)} = E_{g(Al_{x}Ga_{1-x}As)}(1-y) + E_{g(diamond)}y = (1.437 + 1.247x)(1-y) + 15.3y.$$  

(7)

The width of the forbidden band of the diamond $E_{g(diamond)}=15.3$ eV was taken from [15].

Using the equation (7) for (Al$_{x}$Ga$_{1-x}$As)$_{1-y}$C$_{y}$ emission energy $E_{g}$ can be determined in the photoluminescence spectra corresponding with the width of forbidden bands of solid solutions with the specified concentration $(x, y)$. The calculation showed that transition energy for solid solutions (Al$_{x}$Ga$_{1-x}$As)$_{1-y}$C$_{y}$ of the samples EM2889 and EM2887 are identical with those determined experimentally.

Based on the suggested dependence of the width of the forbidden band on the atom concentration (7) as well as the fact that as a result of X-ray diffraction we observe no extra reflexes of the solid solution, we can assume that high energy peaks in photoluminescence spectra of both samples are due to local clusterization of carbon in the matrix of solid solution. Here according to the suggested dependence of energy on the concentration, we can identify the concentration of carbon atoms in the cluster. The calculation shows (see the compositions in Fig. 2) that the concentration of the impurity is within $y$~0.01 which is several times as high as the one we determined using the data of X-ray structural analysis. Therefore, we can say that local concentration of carbon can be higher than the mean value calculated for the solid solution (Al$_{x}$Ga$_{1-x}$As)$_{1-y}$C$_{y}$ according to the data of X-ray structural analysis and photoluminescence.

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**Fig. 2.** Results of PL spectroscopy of heterostructures Al$_{x}$Ga$_{1-x}$As/GaAs (100) with abnormally high mobility of charge.
3. DISCUSSION OF THE RESULTS

To sum up the results of structural, spectroscopy and electrophysical research of epitaxial films Al$_x$Ga$_{1-x}$As, we can conclude the following. Firstly, using the method of MOS-hydride epitaxy heterostructures based on carbon autodoped solid solutions Al$_x$Ga$_{1-x}$As can be obtained with anomalously high mobility of charge carriers for a specified concentration of carriers and composition of the epitaxial film. Secondly, epitaxial films grow on GaAs(100) sublayers coherently and have a high structural quality as indicated by the results of X-ray diffraction. Thirdly, the results of the optical study of the obtained samples as well as the calculation based on the experimental data showed that the concentration of the carbon impurity in epitaxial solid solutions is enough for solid solutions (Al$_x$Ga$_{1-x}$As)$_{1-y}$C$_y$ to form.

The literature review shows that epitaxial growth of films Al$_x$Ga$_{1-x}$As as purposefully as well as carbon autodoped has been studied a lot and the results are summed up in [24—26] and in our paper [5] as well. It is well known that carbon doping of solid solutions of Al$_x$Ga$_{1-x}$As, which is the acceptor, causes p-type conductivity in the epitaxial layer. It has been shown on multiple occasions that carbon concentrations may go up as high as $\sim 10^{20}$ cm$^{-3}$ in particularly when the method of autoepitaxy is used [27].

However, there are studies reporting another possible behaviour mechanism of a carbon impurity during epitaxial growth of GaAs and Al$_x$Ga$_{1-x}$As. Under the selected growth conditions, carbon was shown to be capable of replacing atoms in the metal lattice of a solid solution causing p-type conductivity of the epitaxial layer [28—30]. It was concluded that the inclusion of carbon into the position of the metal C$_{Ga}$ occurs in the concentrations of the impurity in the epitaxial film as high as $\sim 10^{15}$ cm$^{-3}$—$10^{20}$ cm$^{-3}$ (which was achieved in the previous paper) and this behaviour of carbon is caused by a low partial pressure of arsine vapors during epitaxy [31], which is consistent with the literature data.

Therefore, it becomes clear that the doping impurity (carbon) under the selected technological conditions of epitaxy does not replace arsenic in the solid solutions (Al$_x$Ga$_{1-x}$As)$_{1-y}$C$_y$ but takes the regular atom positions of the metal lattice, which accounts for electronic conductivity of the epitaxial layer.

It is well known that mobility of charge carriers is directly connected with an average relaxation time it takes them to diffuse in the semiconductor. However, relaxation time is significantly influenced by the diffusion mechanism, which for the impurity semiconductor is largely contributed by the diffusion by ionized impurities. In this case, the dependence of relaxation time on the concentration of impurities is as follows [32]:

$$\tau \sim \frac{m^* \gamma}{N_i \ln \left[1 + \frac{a}{N_i^{1/3}}\right]^2},$$

where $m^*$ is an effective mass, $a$ is a coefficient, $N_i$ is a concentration of impurity atoms.

(8) means that relaxation time and thus mobility is inversely proportional to the concentration of the impurity. It should be noted that the ratio always holds true when impurity ions with a concentration $N_i$ are distributed in the crystal statically evenly and can be $N_i^{-1/3}$ away from each other.

If we therefore assume that carbon atoms in the metal lattice of the solid solution are arranged in an orderly manner, firstly, it causes effects associated with an arranged impurity ion field and hence extra emission transitions and thus more complex photoluminescence spectra. Secondly, arrangement influences the average distance between ions, i.e. a longer free path of a charge carrier and thus mobility.

To sum up, it should be emphasized that quicker performance of functional elements of the modern optoelectronic base can be achieved using different technological methods considering the physical properties of materials due to employing the effects of anomalously high mobility in the epitaxial layer of the heteropair.

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REFERENCES


Высокая подвижность заряда в гетероструктурах
$\text{Al}_x\text{Ga}_{1-x}\text{As}: \text{C}/\text{GaAs}$ (100)

© 2015 П. В. Середин1, А. С. Леньшин1, В. А. Терновая1, И. Н. Арсентьев2, Tatiana Prutskij3

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Аннотация. Структурными и спектроскопическими методами нами были изучены МОП-гидридные эпитаксиальные гетероструктуры на основе твердых растворов Al$_x$Ga$_{1-x}$As с $n$-типом проводимости. Показано, что при легировании твердых растворов Al$_x$Ga$_{1-x}$As углеродом на уровне 1.2—6.7·10$^{17}$ см$^{-3}$ величина подвижности электронов оказалась аномально высокой для данной концентрации примеси и превосходила расчетную величину в два раза. Высказано предположение, что упорядоченное расположение углерода в металлической подрешетке твердого раствора приводит к изменению среднего расстояния между ионами примеси, т. е. к увеличению длины свободного пробега носителя заряда, а, следовательно, и величины подвижности носителей.

Ключевые слова: эпитаксиальные гетероструктуры, аномально-высокая подвижность носителей заряда, полупроводники, А$_3$В$_5$.