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Photoluminescence of gallium antimonide (GaSb) crystals doped with selenium (Se) and tellurium (Te)

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Abstract. This paper provides a brief overview of main works, dedicated to the study of photoluminescence in gallium antimonide crystals, as well as the features of the band structure and classification of impurity states. Photoluminescence of gallium antimonide doped with selenium and tellurium in the concentration range is presented (10^{17}-$9{\cdot}10^{17}$ cm^{-3}. $1{,}8{\cdot}10^{17}$-$2{\cdot}10^{18}$ cm^{-3}) for selenium and tellurium respectively. It is shown that GaSb photoluminescence is determined by changes from the conduction band to the second ionized state of the double natural acceptor. The shape and position of the maximum of the emission lines is determined by the Coulomb potential of the impurity, additional minima of the conduction band and resonant impurity states associated with them.It was revealed that the difference in the photoluminescence spectra of gallium antimonide crystals containing different donor impurities (selenium, tellurium) is due to the influence of resonant impurity levels associated with the L-minima of the conduction band. Theoretical calculations of the shape of GaSb photoluminescence spectra were performed. The position of the peak and the short-wavelength wing of the emission lines are essentially determined by the position of the Fermi level, which is due to the sharp increase in barrier transparency with increasing electron energy. The practical significance of the results in this article is also related to the widespread use of optical and photoluminescence phenomena in various types of semiconductor devices.

Key words: photoluminescence, impurities, crystal, recombination, concentration, level, energy

Introduction

This paper is dedicated to the study of photoluminescence of doped gallium antimonide crystals.Gallium antimonide is one of the most important materials for infrared optoelectronics – LEDs, lasers and photoconverters[1].

The galliumantimon ideis interesting because it has additional minima of the conduction band located close to the main minimum (~80 meV), as well as the magnitude of the spin-orbit

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splitting Δ is approximately equal to the width of the bandgap E_9 and others.These properties of gallium antimonide distinguish it from the number of A^3 B^5 compounds. For example, by creating uniaxial deformation, it is possible to easily invert the extrema of the conduction band and thereby study the influence of the overlying L-length on the optical properties of the crystal.

Increased interest in the study of photoluminescence in gallium antimonide crystals is stimulated not only by purely scientific objectives: obtaining important data on the band structure of semiconductors, the mechanisms of recombination processes in them, etc. The practical significance of the issues discussed in the article is also associated with the widespread use of optical and photoluminescence phenomena in various types of semiconductor devices. In particular, coherent radiation sources and photodetectors in a wide spectral range are manufactured based on these compounds. These same compounds are also of significant practical interest for use in traditional semiconductor electronics. The spectral region of gallium antimonide (1.1-1.6 μm) contains the main transmission maxima of quartz fibers, most of which are used in fiber optic communications.

When growing GaSb single crystals from gallium-enriched melts, the concentration of this acceptor increases toward the end of the ingot due to the accumulation of excess gallium in the melt. The increase in acceptor concentration, however, occurs more slowly than the increase in Te donor concentration [2].

A number of studies have been devoted to the study of recombination radiation of gallium antimonide. The photoluminescence spectrum of this compound is complex. The introduction of certain impurities, as well as various external influences, significantly influence the shape and position of the emission lines. Therefore, studies by various authors lack a unified view on the nature of these or those emission lines.In the photoluminescence spectra of undoped P-GaSb crystals, two emission lines are observed [3] the authors [4] the positions of these lines with maxima $h\upsilon_1$ = 0.797 eV and $h\upsilon_2$ = 0.776 eV weakly depend on temperature. According to these authors, the high-energy line is associated with the recombination of excitons bound to impurities.

According to measurements, the position of the radiation maxima depends on the temperature. It is also noted that the hυ_1line shifts from 0.800 eV to 0.780 eV when the temperature changes from 10° K to 90° K. They, like other authors [4,5,6] associate this line with interband recombination, and explain the shift of the radiation maximum to the long-wavelength side with increasing temperature by the narrowing of the band gap. The origin of the $h\upsilon_2$ line is associated by many authors [7,8] with the recombination of the conduction band – a shallow natural acceptor. Photoluminescence of gallium antimonide, doped with various impurities, was studied in papers [10]. In the spectra of lightly doped and compensated crystals, which are obtained by introducing tellurium and selenium donors, a new $h\upsilon_3$ long-wavelength line appears. At the same time, the main emission lines are extinguished. According to the authors [7,8], the $h\upsilon_3$ line is associated with the emergence of the possibility of radiative transitions from the conduction band to deep natural acceptors.The position of the maximum of this line depends on the concentration of impurities and on the excitation level. By studying the photoluminescence spectra of n-GaSb samples doped with tellurium, the authors came to the conclusion that the long-wavelength emission line A ($h\upsilon_3$ = 0.720±0.730 eV) corresponds to transitions to the levels of the natural double acceptor. Using higher excitation levels, two more high-energy lines A and B can be detected with energies at maximum of 0.780 eV and 0.796 eV.

They suggest that line A is the result of electron recombination through the same double acceptor, rendered neutral by the capture of two holes, while line B is determined by the emission of an exciton bound to the same neutral center. The authors further point out that line B is significantly broader than the typically observed bound exciton lines and suggest that this broad band contains a significant fraction of band-gap electron emission. A shift of the longest wavelength, line A, toward shorter wavelengths is observed with increasing pumping level, which is explained by an increase in the Fermi level with increasing nonequilibrium carrier concentration.In the works [9]the photoluminescence of gallium antimonide heavily doped with tellurium was studied. At a low excitation level the maximum of the photoluminescence spectrum, which consists of one broad emission line, shifts to the short-wavelength side of the spectrum from 0.710 eV to 0.800 eV with an increase in the electron concentration from $3{\cdot}10^{17}$ cm^3 to $4.7{\cdot}10^{18}$ cm^3. Attributing this phenomenon to the Burstein shift, the authors came to the conclusion that the initial states for radiative transitions lie in the conduction band, and the final states are the same deep acceptor levels with an ionization energy of 0.1 eV that participated in the transitions of lightly doped crystals.

The methodology

A standard setup was used to measure the photoluminescence spectra of gallium antimonide. Nonequilibrium carriers were excited by a He-Ne laser (LG-126) with λ= 1.15 μm, and a P вS receiver was used to record the radiation.A special cryostat was used, eliminating the need for optical windows. The sample is placed in a brass bath, which is housed in a Dewar flask with an open top. The radiation emitted by the crystal is collected by a spherical mirror positioned directly above the open-top Dewar flask. The converging rays from the spherical mirror are directed toward the monochromator by a flat mirror and focused on its slit. The optical surface of the samples was prepared using conventional methods: mechanical grinding on powders with minimal grain sizes (10-14 μm) and chemical etching to a mirror finish in a SR-4A solution. The photoluminescence spectra of n-type gallium antimonide crystals doped with selenium and tellurium were studied (the Hall carrier concentrations were: $1{\cdot}10^{17}$-$9{\cdot}10^{17}$ cm^{-3}. $1,8{\cdot}10^{17}$-$2{\cdot}10^{18}$ cm^{-3} for Se and Te, respectively, at a temperature of 77◦K. The studied concentration range corresponds to the electron degeneracy region.

Discussion

The measurements were carried out at low pump levels; the concentration of nonequilibrium carriers was ≈10^{15} cm^{-3}. The photoluminescence spectra under these conditions are a broad, structureless line (Fig. 1,2).

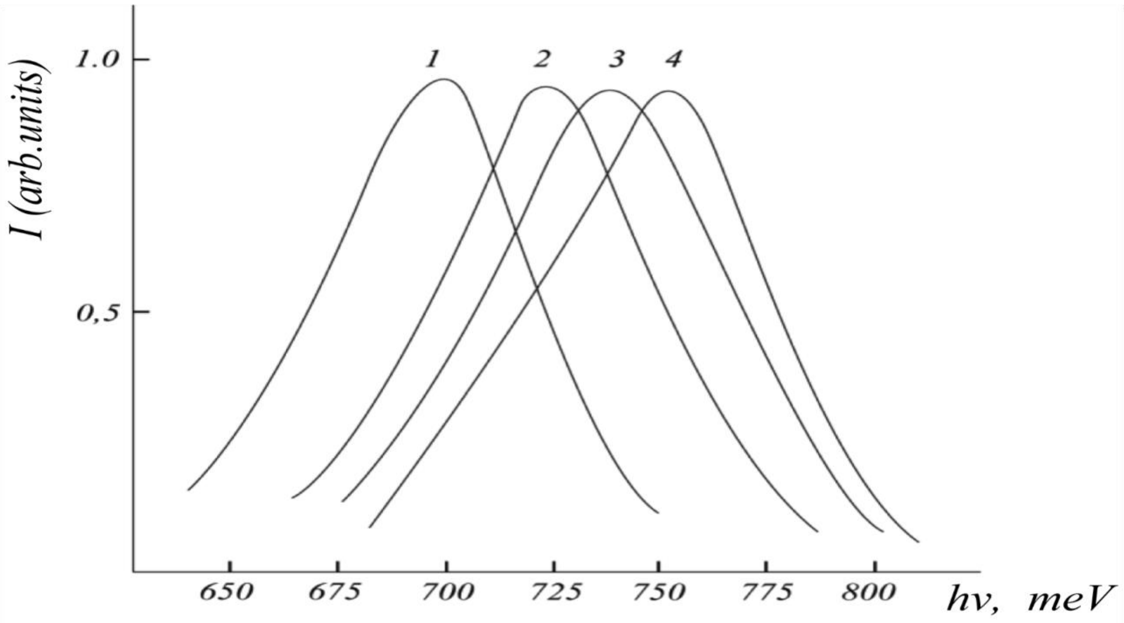

Figure1. Spectra of n-type GaSb (Se) crystals luminescence, T=77°K
1-0,8; 2-3,9; 3-5,7;4-7,5 10^{17} cm^{-3}

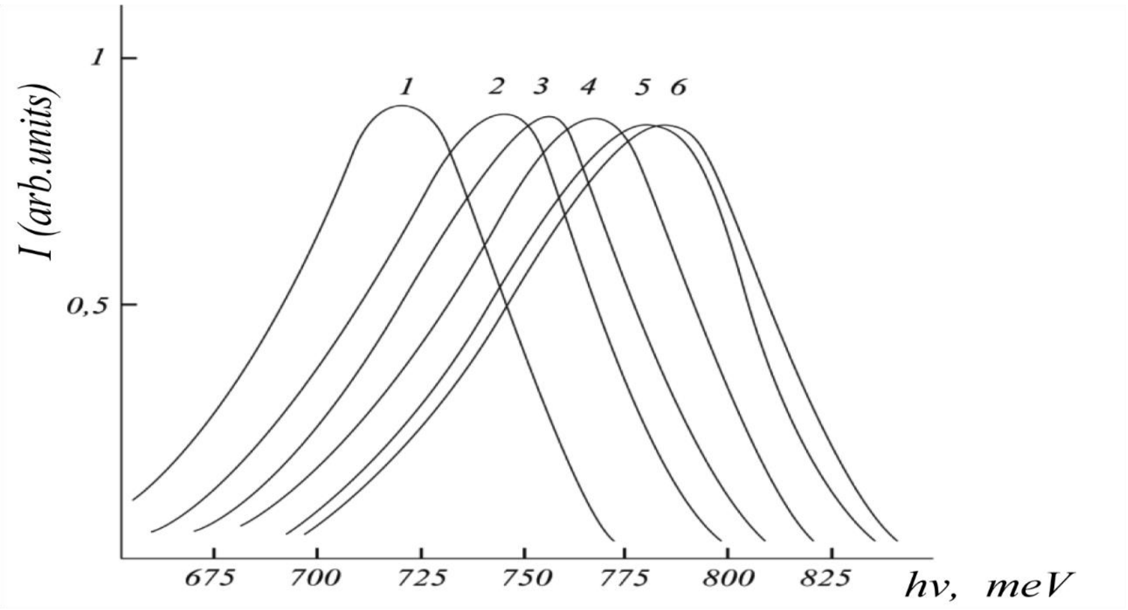

Figure2. Spectra of n-type GaSb (Te) crystal luminescence, T=77°K
N:10^{17} cm^{-3}: 1-2,5; 2-4,2; 3-6,2; 4-8; 5-10.

As the doping level increases, the maximum of the emission lines shifts toward higher energies, and the line broadens. The energy of the maximum of the emission line is always significantly less (80-100 meV) than the optical width of the band gap. Consequently, it can be assumed that the emission line is caused by transitions from the conduction band to a deep acceptor level: for example, to the second ionized state of the double acceptor, which is always present in gallium antimonide. If p-GaSb is doped to degeneracy (N_x=2·10^{17} cm^{-3}, E_F=0.029 eV), then the acceptors are doubly negatively charged. At weak pumping *(($\Delta n, \Delta p \ll n_0$),* nonequilibrium holes are quickly captured by acceptors, which then become singly negatively charged. Consequently, carrier recombination occurs by overcoming the Coulomb barrier, which should be reflected in the line shape and the position of the emission maximum. Figures 3 and 4 show the concentration dependences of the maximum and half-width of the emission line of gallium antimonide doped with Se and Te, respectively.

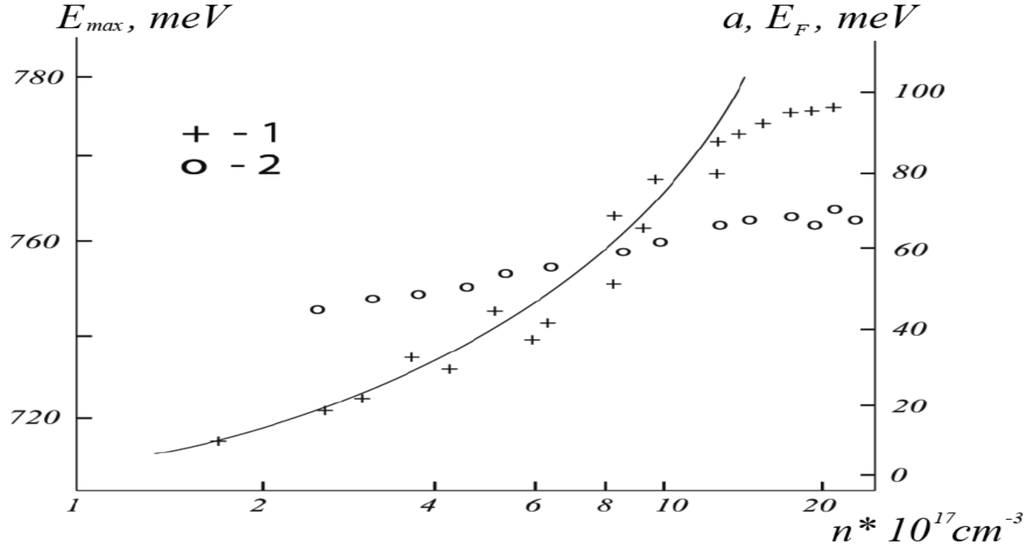

Figure3. Position of the maximum (1), half-width of the emission lines (2) and the Fermi level (3) of GaSb (Te) crystals from the doping level, T=770K

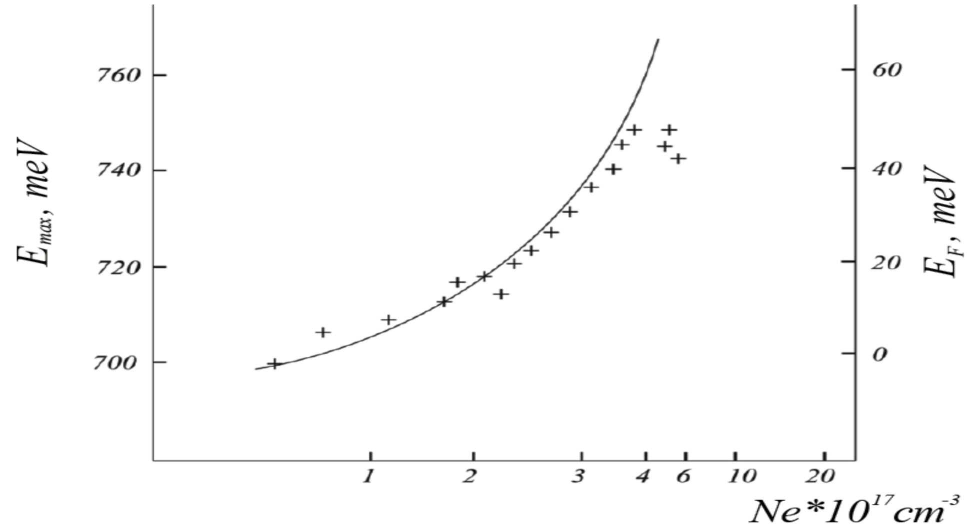

Figure4. The position of emission line maximum and level of Fermi crystalsGaSb (Se) from doping levelT=77°К

It is evident from the figures that up to concentrations of $5*10^{17}$ cm^{-3} both dependences are practically identical, and as the degree of doping increases, the influence of the impurity type appears. For gallium antimonide crystals doped with Te maximum emission from concentration shifts monotonically to the short-wavelength side with doping, saturating at N_x>$1·10^{18}cm^{-3}$, while the dependence $E^{Se}{}_{max}$ occurs at N_x>$7·10^{17}$ $^{8}cm^{-3}$ and shifts to the long-wavelength side. The behavior of the half-width of the emission lines on the carrier concentration is similar to the behavior of E_{max}. The shape of the emission line for conduction-acceptor band transitions can be described by the following expression:

$$I(E)=\rho(E)F(E)N(E) \tag{1}$$

ρ (E)-the density of states in the conduction band, assuming the conduction band to be spherical andisotropic, thenρ(E)~√E, E-kinetic energy of electrons; $F(E) = \frac{1}{exp(E-E_F)/kT}$ Fermi equilibrium function: $N(E)=A/\sqrt{E}(expA/\sqrt{E} - 1)^{-1}$ Sommerfeld multiplier for the repulsive:

$$A = \frac{2\pi}{\hbar\sqrt{2m}} \tag{2}$$

The factor (E) takes into account that transitions are associated with ionized acceptor states, i.e., with overcoming a repulsive barrier. Consequently, transitions are more probable for electrons with high kinetic energy. The results of calculations using formula (4) are shown in Figure 5. The position of the maximum and the short-wavelength wing of the emission line are practically determined by the position of the Fermi level, which is due to the sharp increase in barrier transparency with increasing electron energy. Figures 3 and 4 show the dependence of the Fermi level on the carrier concentration. The concentration is taken from direct measurements of the Hall coefficient. Indeed, as can be seen from the figure, the shift of the emission maximum with concentration toward shorter wavelengths is analogous to the change in the position of the Fermi level for $E^{Te}{}_{max}$to concentrations $1·10^{18}$ cm^{-3}, for $E^{Se}{}_{max}$ to $7·10^{17}$ cm^{-3}. At N_x>$1·10^{18}$ cm^{-3} dependence $E^{Te}{}_{max}$ is saturated, which is apparently due to the filling of the L-valley of the conduction band, since the Fermi level at a given concentration reaches the L-minima. At the same time, a contribution to radiative recombination from transitions from the L-minima should appear. This is evident in a slight discrepancy between theory and experiment in the short-wavelength wing of the spectrum at high carrier concentrations. The contribution from transitions from the L-minima was not taken into account in the calculations. The long-wavelength wing of the emission band is determined primarily by two factors: the distribution of the density of states near the bottom of the conduction band and the screening of the Coulomb potential of the impurity center. Gallium antimonide always contains a large number of natural acceptors ($P \cong 1\text{-}2·10^{17}$ cm^{-3}). Therefore, when doping with donor impurities to a level of N_x~$5·10^{17}$ cm^{-3}, a significant degree of compensation exists, resulting in “tails” in the density of states in the band gap. The presence of these tails, which we did not take into account in our calculations, is one of the reasons for the discrepancy between the experimental data and the calculation in the region of the long-wavelength wing of the spectra at N_x<$5·10^{17}$ cm^{-3}. Another factor determining the long-wavelength edge of the luminescence spectrum is the screening of the Coulomb potential of the impurity center by free electrons. The presence of a negative charge at the impurity center leads to a dependence of the probability of radiative recombination of electrons on their kinetic energy. This situation is analogous to the dependence of the transparency of the Coulomb barrier on the particle energy during tunneling. An increase in the electron concentration leads to a lowering and narrowing of the Coulomb barrier, increasing the probability of electrons with low kinetic energy passing through it. One can attempt to account for the screening of the electrostatic potential by taking the eigenvalue of the Schrödinger equation with a Hamiltonian including the electrostatic screened potential as the total electron energy E(z) in the single-electron approximation:

$$V(r) = \frac{e^2}{r} e^{-\frac{r}{r_э}} \tag{3}$$

where $r_э$ – Thomas-Fermi screening radius

$$1/r_э = (\frac{3Ne}{\pi})^{1/6} \sqrt{\frac{4me^2}{\varkappa\hbar^2}}$$

In general, the Schrödinger equation does not have a closed-loop analytical solution. However, such a solution can be obtained for large values of the screening radius $r_э > a_б$. The static screened potential in this case can be represented as:

$$1/r_э = (\frac{3Ne}{\pi})^{1/6} \sqrt{\frac{4me^2}{\varkappa\hbar^2}} \tag{4}$$

Taking into account screening in this case comes down to adding the following expression to the total energy of the electron:

$$\frac{e^2}{r} - \frac{3}{4}\frac{e^2}{r_э^2} a_б \tag{5}$$

i.e. to the shift in the electron energy reference point. One can attempt to account for the effect of screening in the same way for the case$r_э \leq a_б$, which is the case for the crystals under study. The calculation using formula (5) is shown in Figure 5:

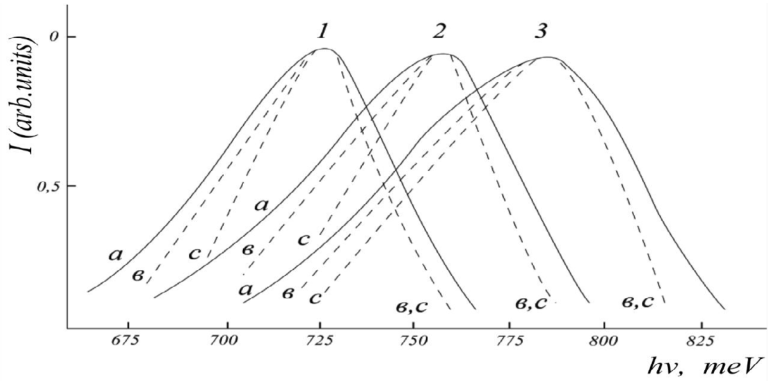

Figure5. Spectra of luminescence of n-typeGaSb,theory: a-no screening; b- subject to screening; c-experiment

It is evident that taking into account the screening broadens the long-wavelength wing of the spectrum. The best agreement between the experimentally observed spectra of GaSb(Te) and the calculated results is observed at Hall concentrations of (6-8) 10^{17} *cm*$^{-3}$; i.e., when the role of the density of states “tails” is already relatively small, and the contribution of transitions from the L-minimum is still insignificant. Returning to Figs. 2 and 3, we note a significant difference in the behavior of the concentration dependences of Emaxfor crystals doped with tellurium and selenium. In the case of GaSb(Te), E^{Te}_{max} monotonically shifts to the short-wavelength side similarly to E_F up to concentrations of the order of $1{\cdot}10^{18}$ *cm*$^{-3}$, and saturates with a further increase in the concentration. This behavior at high concentrations is due to the fact that the Fermi level reaches the L-minima of the conduction band (the Te level, located below the L-minima for these concentrations is degenerate) and due to the higher density of states in them(m_2*=0,29m_0) the growth of the Fermi level should slow down. Moreover, at such electron concentrations, the effect of narrowing of the band gap with doping is already significant. Saturation can be explained in this case by the simultaneous action of two factors: a weak increase in the Fermi level and a decrease in the band gap with doping. In GaSb(Se) crystals, the resonant levels of selenium associated with the L-minima, as will be shown in the next paragraph, are located 33 meV below the bottom of the L-minimum of the conduction band. Consequently, saturation of the concentration dependence of E^{Se}_{max}, associated with the filling of the resonant states with Ie, should occur at lower concentrations N_x than for GaSb(Te) crystals. However, it is evident from Fig. 4 that the dependence of E^{Se}_{max} on N_x passes through a maximum at $N_x \sim 7{\cdot}10^{17}$ *cm*$^{-3}$ and then shifts to the long-wavelength side with a further increase in N_x. Therefore, it can be concluded that the decrease in the band gap with doping should be significantly greater than for crystals in GaSb(Te). The stronger decrease in the band gap with doping may be due to the fact that selenium in gallium antimonide produces a center that differs from the hydrogen-like center, and for such a center, the dependence of the change in the band gap width with doping can be significantly stronger than for the hydrogen-like center.

Conclusion

In this paper, the photoluminescence of gallium antimonide (GaSb) crystals doped with selenium (Se) and tellurium (Te) was studied, and the following conclusions were drawn from the obtained data:

1. The photoluminescence of gallium antimonide crystals doped with selenium and tellurium was determined by the transition from the conduction band to the second ionized state of the natural acceptor in the concentration range ($1{*}10^{17}$ – $8{*}10^{18}$ *cm*$^{-3}$ и $1.8{*}10^{17}$ – $2{*}10^{18}$ *cm*$^{-3}$) for selenium and tellurium.

2. The shape and position of the emission maximum line are determined by the Coulomb potential of the impurity, additional minima in the conduction band, and associated resonant impurity states.

3. It was established that the difference in the photoluminescence spectra of gallium antimonide crystals with different donor impurities (selenium, tellurium) is due to the influence of resonant impurity levels associated with the conduction band minima.

4. It was noted that the shift of the long-wavelength emission edge toward higher energies with increasing concentration is explained by the influence of impurity concentration fluctuations.

Acknowledgement, conflict of interests

The authors declare that they have no conflict of interest.

The contribution of the authors

Egemberdieva S. – drafted the manuscript, read and edited it, and formulated the key goals and objectives.

Kushkimbayeva B. – collected data, conducted the analysis, and approved the final version of the article for publication.

Kusherbayeva M. – read and edited the manuscript, providing valuable critical comments, prepared illustrations, and wrote and edited the text.

Keikimanova M. – processed and interpreted the data.

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Селен (Se) және теллур (Te) қосылған галий антимониді (GaSb) кристалдарының фотолюминесценциясы

Аңдатпа. Бұл жұмыс галлий антимонидінің кристалдарындағы фотолюминисценцияны зерттеуге арналған негізгі жұмыстардың қысқаша шолуын, сондай-ақ аймақтық құрылымның ерекшеліктерін және қоспа күйлерінің классификациясын қарастырады. (10^{17}-$9{\cdot}10^{17}$ $см^{-3}$. $1{,}8{\cdot}10^{17}$-$2{\cdot}10^{18}$ $см^{-3}$) концентрациядиапазонында селен мен теллур үшін легирленген галлий антимонидінің фотолюминисценциясы берілген. GaSb фотолюминесценциясы қос табиғи акцептордың өткізгіштік аймағынан екінші иондалған күйіне өту арқылы анықталатыны көрсетілген. Шығару сызығының максимумының пішіні мен орны қоспаның кулондық потенциалымен, өткізгіштік аймағындағы қосымша минимумдармен және олармен байланысты резонанстық қоспа күйлерімен анықталады.Құрамында әртүрлі донорлық қоспалар (селен, теллур) бар галлий антимониді кристалдарының фотолюминесценция спектрлеріндегі айырмашылықтар өткізгіштік зонаның L-минимумдарымен байланысты резонанстық қоспалар деңгейлерінің әсерінен болатыны анықталды. GaSb фотолюминесценция спектрлерінің пішінінің теориялық есептеулері жүргізілді. Максимумның орналасуы және қысқа толқынды сәулелену сызықтарының қанаты іс жүзінде Ферми деңгейінің орналасуымен анықталады бұл электрондардың энергиясының жоғарылауымен тосқауыл мөлдірлігінің күрт өсуіне байланысты. Мақалада жұмыс нәтижелерінің практикалық маңыздылығы сонымен қатар жартылай өткізгіш құрылғылардың әртүрлі түрлерінде оптикалық және фотолюминесценциялық құбылыстарды кеңінен қолданумен байланысты екені келтірілген.

Түйін сөздер: фотолюминисценция, қоспалар, кристалл, рекомбинация, концентрация, деңгей, энергия

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Фотолюминисценция кристаллов антимонида галлия (GaSb) легированных селеном (Se) и теллуром (Te)

Аннотация. В данной работе рассматриваются краткий обзор основных работ, посвященных исследованию фотолюминисценций в кристаллах антимонида галлия, а также особенности зонной структуры и классификация примесных состояний. Приведены фотолюминисценция антимонида галлия, легированных селеном и теллуром в диапазоне концентраций (10^{17}-$9\cdot10^{17}$ *см*$^{-3}$. $1{,}8\cdot10^{17}$-$2\cdot10^{18}$ *см*$^{-3}$) для селена и теллура соответственно. Показано что фотолюминисценция GaSb определяется перепадами из зоны проводимости на второе ионизованное состояние двойного природного акцептора. Форма и положение максимума линий излучения определяется кулоновским потенциалом примеси, дополнительными минимумамизоны проводимости и резонансными примесными состояниями связанными с ними. Выявлено, что различие спектров фотолюминисценций кристаллов антимонида галлия,содержащих различные донорные примеси (селен, теллур) обусловлен влиянием резонансных примесных уровней связанных с L–минимумами зоны проводимости. Проведены теоретические расчеты формы спектров фотолюминисценций GaSb. Положение максимума и коротковолновые крыло линий излучения практически определяется положением уровня Ферми что обусловлено резким возрастанием прозрачности барьера при увеличений энергий электронов.Практическое значение итогов работы в статье связано также с широким использованием оптических и фотолюминисцентных явлений в различных типах полупроводниковых приборов.

Ключевые слова: фотолюминисценция, примеси, кристалл, рекомбинация, концентрация, уровень, энергия

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