# Energy band structure of $Ga_xIn_{1-x}P$ and $Ga_{x'}In_{1-x'}As_{1-y'}P_{y'}$ solid solutions

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An investigation was made of the photoluminescence of GaInP and GaInAsP solid-solution films formed by liquid epitaxy on GaAsP substrates. The photoluminescence spectra of the compositions near the point of transition from the direct to indirect band structure included exciton bands associated with both the Γ and $X$ conduction-band minima. An analysis of the photoluminescence spectra yielded the parameters of the point of transition ($E_{g\Gamma} \equiv E_{gX}$) from the direct to indirect band structure in the $Ga_xIn_{1-x}P$ ternary solid-solution system: $E_{gt} = 2.310$ eV, $x_t = 0.71$ at 77°K; $E_{gt} = 2.24$ eV, $x_t = 0.72$ at 300°K. This analysis yielded also the rate of reduction in the gap between the Γ and $X$ minima in a constant-period ($a$ = const) series of GaInAsP quaternary solid solutions: 0.2 per 1 meV reduction in the direct band gap. The results obtained were used to estimate the shortest stimulated-emission wavelengths which should be obtained from GaInAsP injection heterojunction lasers: these were 555 nm at 77°K and 617 nm at 300°K.

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"Ideal" heterojunctions between constant-period GaInAsP quaternary solid solutions were proposed in Ref. 1 and experimental realizations of this proposal were reported in Refs. 2-5. The wide direct band gap of these solid solutions made it possible to generate the shortest known injection laser wavelengths in the Ga–In–As–P system.[6,7] The very important question of the highest energies of the photons which can be generated in such lasers is discussed briefly in Ref. 7. Our task was to carry out a detailed investigation of the energy band structure of GaInP and GaInAsP solid solutions using the photoluminescence spectra emitted by these materials in the range of compositions close to the transition from the direct to indirect energy band structure.

The dependences of the band gaps on the lattice parameters of the GaInP, GaAsP, GaInAs, and InAsP ternary solid-solution systems are plotted in Fig. 1. The quaternary GaInAsP solid solutions correspond to the part of the plane bounded by these curves. The constant-period set of compositions in the GaInAsP system is represented by a vertical straight line in this diagram. The widest gap among these constant-period compositions is exhibited by those belonging to the GaInP ternary system. The quaternary GaInAsP composition with the identical direct and indirect band gaps ($E_{g\Gamma} = E_{gX}$, $\Delta E_{g\Gamma-X} = 0$) is represented in Fig. 1 by the curve joining the transition points of the dependences $E_{g\Gamma} = f(a)$ and $E_{gX} = f(a)$ exhibited by the ternary systems GaAsP and GaInP. The determination of the nature of this curve was the final task of our investigation. However, the first stage was to determine more accurately the points of transition from the direct to indirect band structure in the GaInP system because of the serious discrepancies between the published data (see Ref. 8). This was not necessary for the GaAsP system because the energy band structures of these solid solutions had been investigated thoroughly for all the compositions.[9]

## SAMPLES AND MEASUREMENT METHOD

Deliberately undoped $Ga_xIn_{1-x}P$ and $Ga_{x'}In_{1-x'}As_{1-y'}P_{y'}$ solid-solution films were grown by liquid epitaxy in which the melt was cooled from 750 to 745°C at a rate of ~2°C/min. The film thickness was 2-3 μ. The substrates were $GaAs_{1-y}P_y$ solid solutions of various compositions grown by the gas-transport method on GaAs plates oriented in the (111)A direction.

We investigated the photoluminescence emitted by these films in the temperature range 77-300°K. This photoluminescence was excited either by a DRSh-250 mercury lamp (low excitation rates) or by an LGI-21 pulsed nitrogen laser ($P = 10^3$ W/cm², $\tau$ = 10 nsec). The detector was an FÉU-79 photomultiplier.

We selected only those epitaxial ternary or quaternary solid-solution films whose natural and cleaved surfaces had a good morphology and whose photoluminescence edge bands were narrowest at 77°K. The results of earlier investigations[10-12] demonstrated that such films had the lattice parameter (period) which agreed very closely with the lattice parameter of the substrate. For example, a deviation of the composition of a $Ga_xIn_{1-x}P$ solid solution by just 1% from the value ensuring identity of the lattice constants with the substrate resulted in a strong deterioration of the structure and of the luminescence emitted by the film.

In the case of the $Ga_xIn_{1-x}P$ films grown on the $GaAs_{1-y}P_y$ substrates the constant-period condition following from the Vegard law had the simple form

$$x = 0.51 + 0.49y. \quad (1)$$

We could use this expression to find the composition x of a GaInP solid solution grown on a GaAsP substrate with a known value of y. In the present investigation we used a different method for the determination of the composition of $Ga_xIn_{1-x}P$ solid solutions. We utilized the fact that the band gap of the $Ga_{0.51}In_{0.49}P$ solid solutions grown on GaAs substrates could be regarded as well established[1)]:

$$E_{g\Gamma}(300\,°\text{K}) = 1.905 \text{ eV}^{[10-13]},$$
$$E_{g\Gamma}(77\,°\text{K}) = (1.985 + 0.007)\text{eV}^{[10,12,14]}.$$

The parameters of this point, together with the known en-

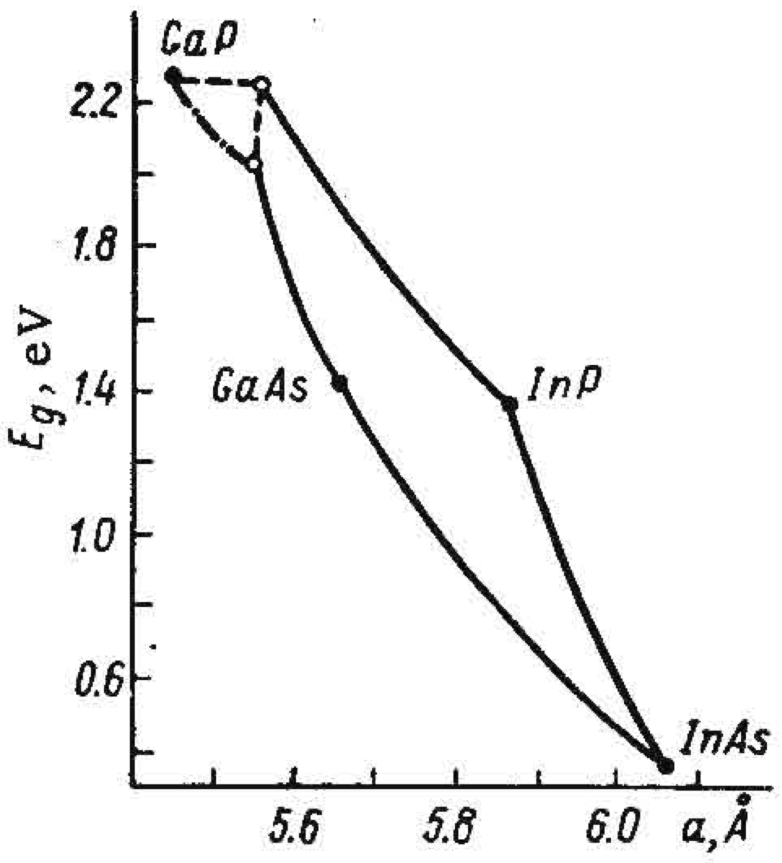


FIG. 1. Dependences of the band gap on the lattice parameter of GaInAsP solid solutions of various compositions. T = 300°K. The continuous curves correspond to the compositions with the direct energy band structure and the dashed curves to the indirect band structure; the chain curve is the transition from the direct to indirect band structure.

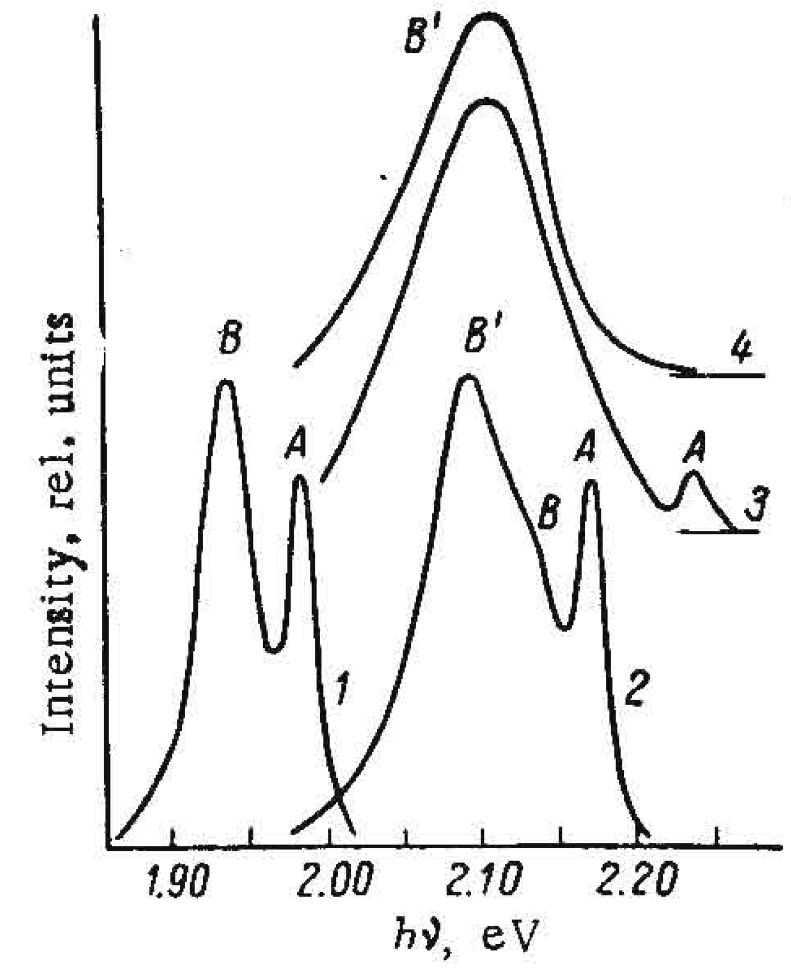


FIG. 2. Photoluminescence spectra of undoped $Ga_xIn_{1-x}P$ solid solutions of different compositions x: 1) 0.51; 2) 0.63; 3) 0.67; 4) 0.69. T = 77°K, low excitation rate.

ergy band parameters of InP and GaP (see Ref. 8), enabled us to find the coefficients in the quadratic polynomial describing the dependence of $E_{g\Gamma}$ on the composition x of $Ga_xIn_{1-x}P$:

$$E_{g\Gamma}(x) = 1.414 + 0.796x + 0.656x^2 \ (77\ ^\circ K), \quad (2)$$

$$E_{g\Gamma}(x) = 1.351 + 0.727x + 0.702x^2 \ (300\ ^\circ K). \quad (3)$$

Clearly, in the range of compositions where the quasi-interband luminescence was recorded, associated with the Γ conduction-band minimum, we could use Eqs. (2) and (3) to find the composition of the investigated $Ga_xIn_{1-x}P$ epitaxial films.

## EXPERIMENTAL RESULTS AND DISCUSSION

### 1. $Ga_xIn_{1-x}P$ Solid Solutions

Figure 2 shows the photoluminescence spectra of several $Ga_xIn_{1-x}P$ samples of different compositions recorded at a low excitation rate at 77°K. The spectrum of the sample with x = 0.51 consists of a band A, due to quasiinterband transitions involving the participation of the Γ conduction-band minimum, and a band B of longer wavelength, which is due to transitions involving shallow donor and acceptor levels.[10] The photoluminescence spectra of samples with x ⩾ 0.62 exhibit – in the long-wavelength wing of the band B – an incipient new wide band B', which dominates the spectra of the samples with x > 0.65. In this range of compositions there is a reduction in the relative intensity of the band A. In the spectra of the samples with x ⩾ 0.69 the band A can be observed only at a high excitation rate (curve 4 in Fig. 2 and curve 1 in Fig. 3, both of which apply to the same sample). A comparison of these curves shows that the relative intensity of the band B' decreases as a result of an increase in the rate of excitation and the position of the maximum shifts considerably in the direction of higher energies.

At a high excitation rate (77°K) an increase in the GaP content results in a continuing increase in the intensity of the B' band in the photoluminescence spectra of $Ga_xIn_{1-x}P$ samples (Fig. 3). A shift of the band A toward shorter wavelengths is accompanied by the appearance of two new luminescence bands: a long-wavelength band X – LA ($h\nu_{max} \approx 2.27$ eV) and a weaker short-wavelength band X ($h\nu_{max} \approx 2.30$ eV), both of which shift slowly toward higher energies when x is increased. A further increase of x in the range x > 0.71 at a high excitation rate results in rapid quenching of the band A, which eventually disappears.[2)]

We shall now analyze briefly these results. A com-

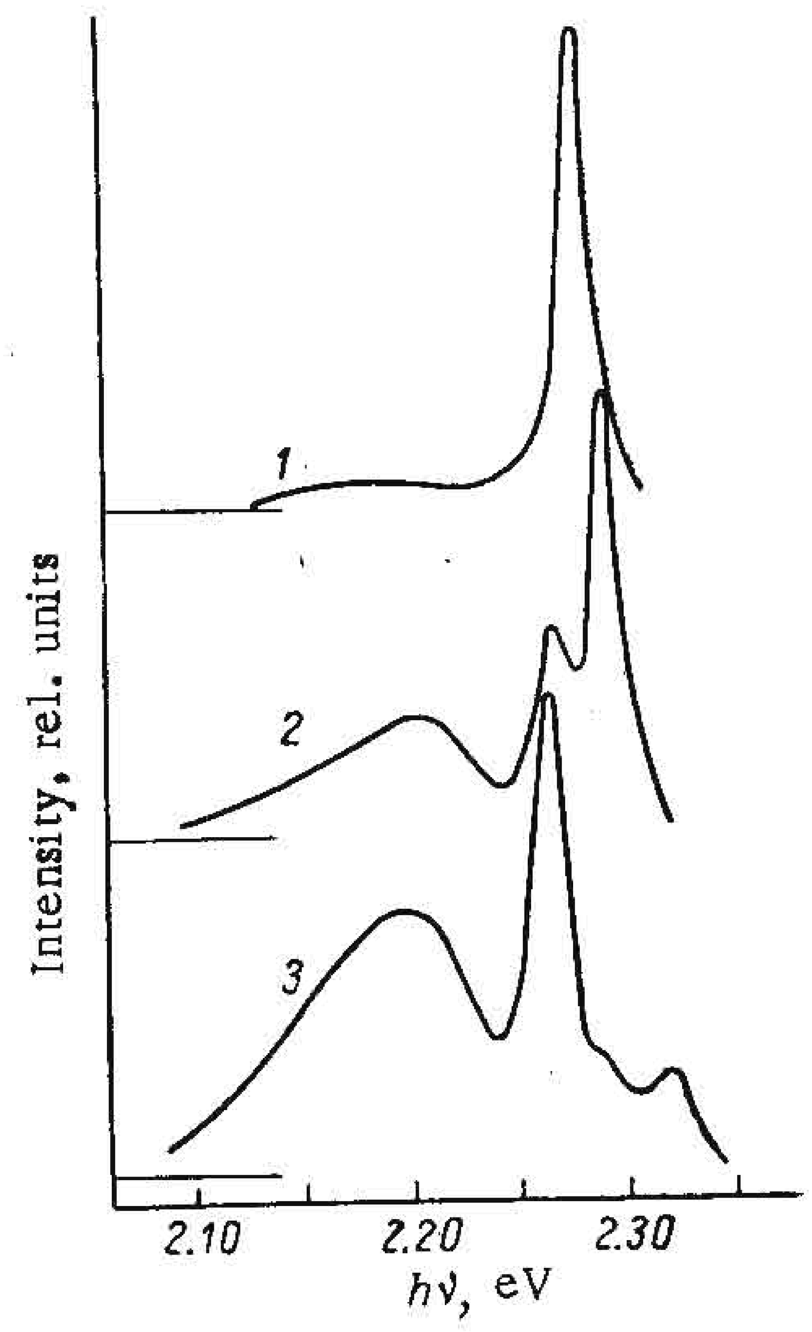


FIG. 3. Photoluminescence spectra of undoped $Ga_xIn_{1-x}P$ solid solutions of different compositions x: 1) 0.69; 2) 0.70; 3) 0.72. T = 77°K, high excitation rate.

parison with the photoluminescence spectra of GaAsP (Ref. 15) and AlGaAs (Ref. 16) solid solutions of compositions near the transition from the direct to indirect energy band structure shows that the B' luminescence band is due to interimpurity transitions involving participation of donor levels associated with the X conduction-band minimum. Clearly, the system of the edge luminescence bands (curve 3 in Fig. 3) is associated with exciton energy states, which are again related to the X minimum. The short-wavelength band X appears as a result of annihilation of free excitons without phonon participation. The appearance of this zero-phonon free-exciton band is possible in indirect-gap semiconductors on introduction of an isoelectronic impurity (in particular, as a result of formation of solid solutions) and it is due to the dissipation of quasimomentum because of the interaction with the potential of isoelectronic centers (In atoms) in the course of recombination. Optical transitions associated with the free-exciton annihilation are known to occur in the luminescence of indirect GaAsP solid solutions[9] and also AlGaP and GaInP solutions.[17,18] The X – LA band is a phonon replica of the band X (the former is due to the annihilation of an exciton accompanied by the emission of an LA phonon). Apart from g ral considerations based on the selection rules,[19] this interpretation is supported also by the size of the energy gap (27-30 meV) between the X and X – LA luminescence bands.

Additional evidence supporting this interpretation of the nature of the X and X – LA short-wavelength bands is provided by the results of our investigation of the influence

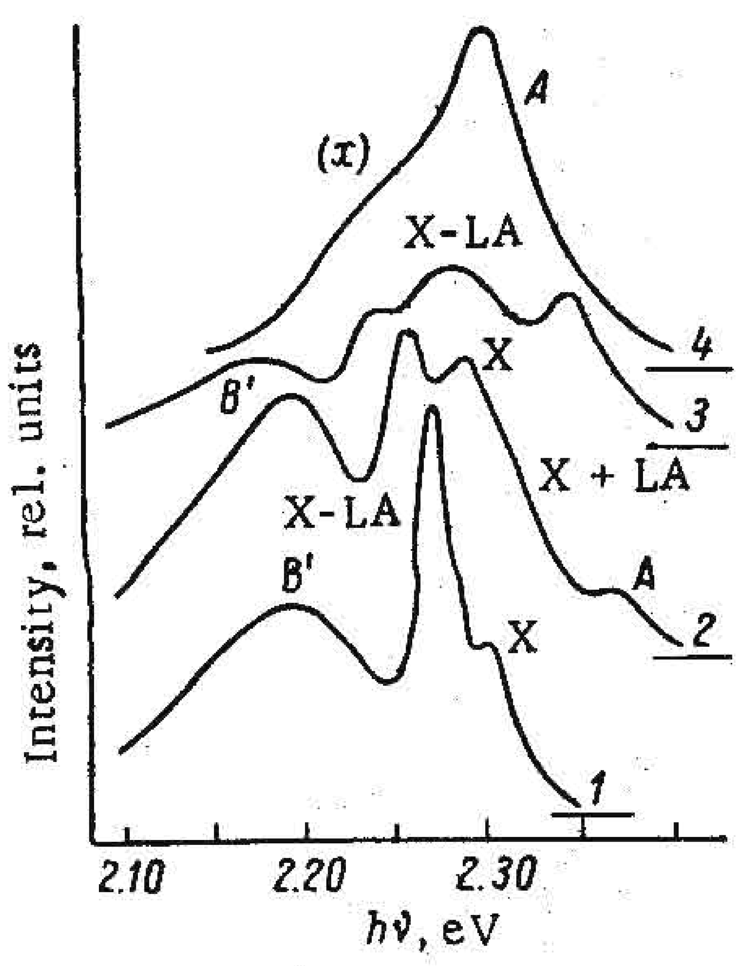


FIG. 4. Influence of temperature on the photoluminescence spectra of the undoped solid solution $Ga_{0.75}In_{0.25}P$. T (°K): 1) 77; 2) 128; 3) 179; 4) 300. High excitation rate.

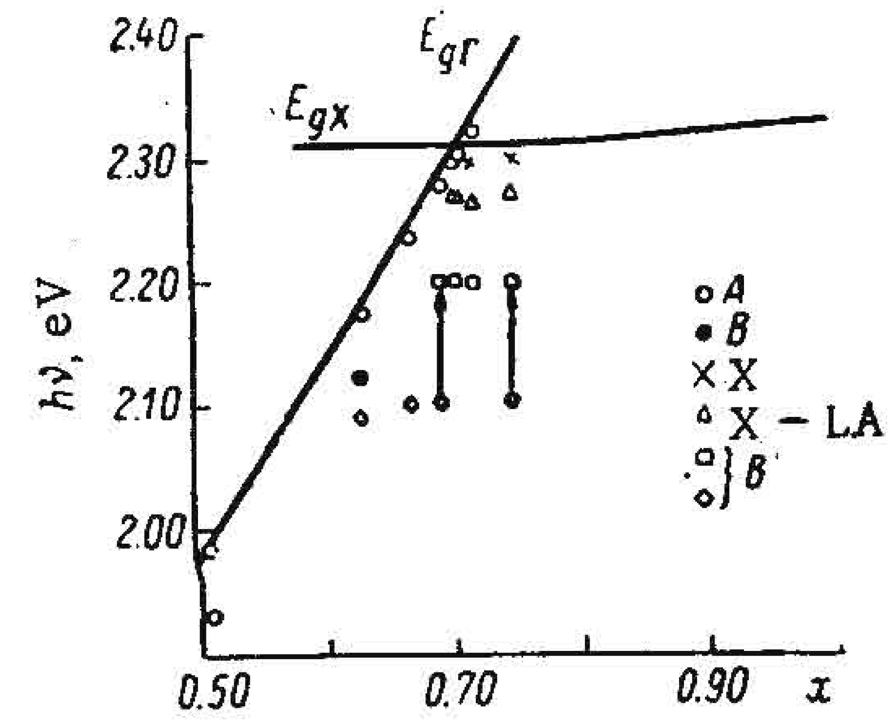


FIG. 5. Positions of the band maxima in the photoluminescence spectra of undoped $Ga_xIn_{1-x}P$ solid solutions of different compositions. T = 77°K. The curve identified as $E_{g\Gamma}$ represents calculations based on Eq. (2) and the arrows indicate the shift of the band B' on increase of the excitation rate.

of temperature on the photoluminescence spectra of a sample of $Ga_{0.75}In_{0.25}P$ (Fig. 4). We can see that an increase in temperature from 77 to 120°K increases the relative intensity of the zero-phonon band X and produces, in its short-wavelength wing, a new band X + LA, due to the annihilation of a free exciton accompanied by the absorption of an LA phonon. Above 120°K an increase in the electron density in the Γ minimum gives rise to the band A, which is due to direct transitions, although the composition of the sample is in the indirect range.

At room temperature the band A predominates (curve 4 in Fig. 4). At the same time the long-wavelength wing exhibits, at 300°K, the exciton luminescence associated with the indirect X minimum.

These results allow us to determine quite easily the position of the point of transition from the direct to indirect energy band structure in $Ga_xIn_{1-x}P$ solid solutions, namely the values of $E_{gt}$ and $x_t$. Figure 5 shows the dependences of the positions of the maxima of various luminescence bands of $Ga_xIn_{1-x}P$ samples on their composition (77°K). If we assume that the binding energy of a free exciton associated with the X conduction-band minimum is 10 meV (Ref. 19), the intersection of the dependence $E_{gX} = f(x)$ with the dependence $E_{g\Gamma} = f(x)$ calculated from Eq. (2) gives, at liquid nitrogen temperature, the values of $E_{gt}$ = 2.310 eV and $x_t = 0.71$.

An analysis of the temperature shifts of the various photoluminescence bands in the spectra represented by curves 2-4 in Fig. 4 makes it possible to estimate the values of $E_{gt}$ and $x_t$ of $Ga_xIn_{1-x}P$ solid solutions at room temperature. If we use Eq. (3), we find that the parameters of the point of transition from the direct to indirect structure at 300°K are $E_{gt}$ = 2.24 eV, $x_t$ = 0.72.

The values of $E_{gt}$ and $x_t$ are in satisfactory agreement with those deduced recently[20,21] from the absorption and electroreflectio. spectra of bulk $Ga_xIn_{1-x}P$ crystals. However, the greatest interest lies in a comparison of these results with those of a recent paper[8] reporting a study of the absorpti nd photoluminescence spectra of $Ga_xIn_{1-x}P$ films prepared, as in our study, by liquid epitaxy on $GaAs_{1-y}P_y$ substrates. The composition of the $Ga_xIn_{1-x}P$ samples, which have the same lattice parameters as the $GaAs_{1-y}P_y$ substrate, is deduced in Ref. 8 from the composition of the substrate and Eq. (1), whereas the band gap is found from the experimental absorption data. It is worth noting that the positions of the maxima of the edge photoluminescence bands of the $Ga_xIn_{1-x}P$ samples investigated in Ref. 8 are shifted by 15 meV toward lower energies compared with the values of $E_{g\Gamma}$ deduced from the experimental absorption data. This is to be expected because the absorption and photoluminescence data refer to the same thin (1.4 μ) samples without an absorbing substrate. It is shown in Ref. 22 that the effects of repeated absorption of light play an important role in such samples at 77°K and they shift the position of the edge photoluminescence band by ~7 meV toward lower energies. At room temperature this shift may be greater. The dependences $E_{g\Gamma}(x)$ obtained at 77 and 300°K and reported in Ref. 8 readily show that in the case of $Ga_{0.51}In_{0.49}P$ on a GaAs substrate the band gap is 1.965 and 1.884 eV, respectively. These two values are more than 20 meV lower than the experimental data reported in Refs. 10-14. This overestimates the coefficient in front of the quadratic term in the dependence $E_{g\Gamma} = f(x)$, which gives a higher (by 0.01) value of $x_t$ in Ref. 8, compared with that reported in the present paper.

## 2. GaInAsP Solid Solutions

The luminescence of quaternary GaInAsP solid solutions is largely similar to the luminescence of the ternary GaInP solid solutions with the same lattice constants. At 77°K the photoluminescence spectra of GaInAsP samples of compositions far from the indirect range consist of two bands A and B, which shift (without a separation between them) toward longer wavelengths compared with the GaInP samples with the same lattice constant.[4] The photoluminescence spectra of GaInAsP samples of compositions in the indirect range (77°K) contain, like the photoluminescence spectra of $Ga_xIn_{1-x}P$ with $x > 0.73$, an interimpurity band B' and the edge luminescence is due to the annihilation of free excitons either without phonon participation (band X) or accompanied by the emission of LA phonons (band X – LA). However, in the spectra of these samples the relative intensity of the X band is considerably greater than the intensity of the phonon replica X – LA. This is clearly due to an even greater likelihood of the dissipation of quasimomentum on lattice irregularities in the course of annihilation of indirect excitons in a multicomponent solid solution.

We shall now consider the nature of the photoluminescence spectra of GaInAsP solid solutions of compositions close to the transition from the direct to indirect energy band structure. As in the case of the photoluminescence spectra of $Ga_xIn_{1-x}P$ solid solutions, the spectra of the GaInAsP system include the bands A and X – LA, which are separated by an energy from which we can deduce the gap between the Γ and X conduction-band minima. Figure

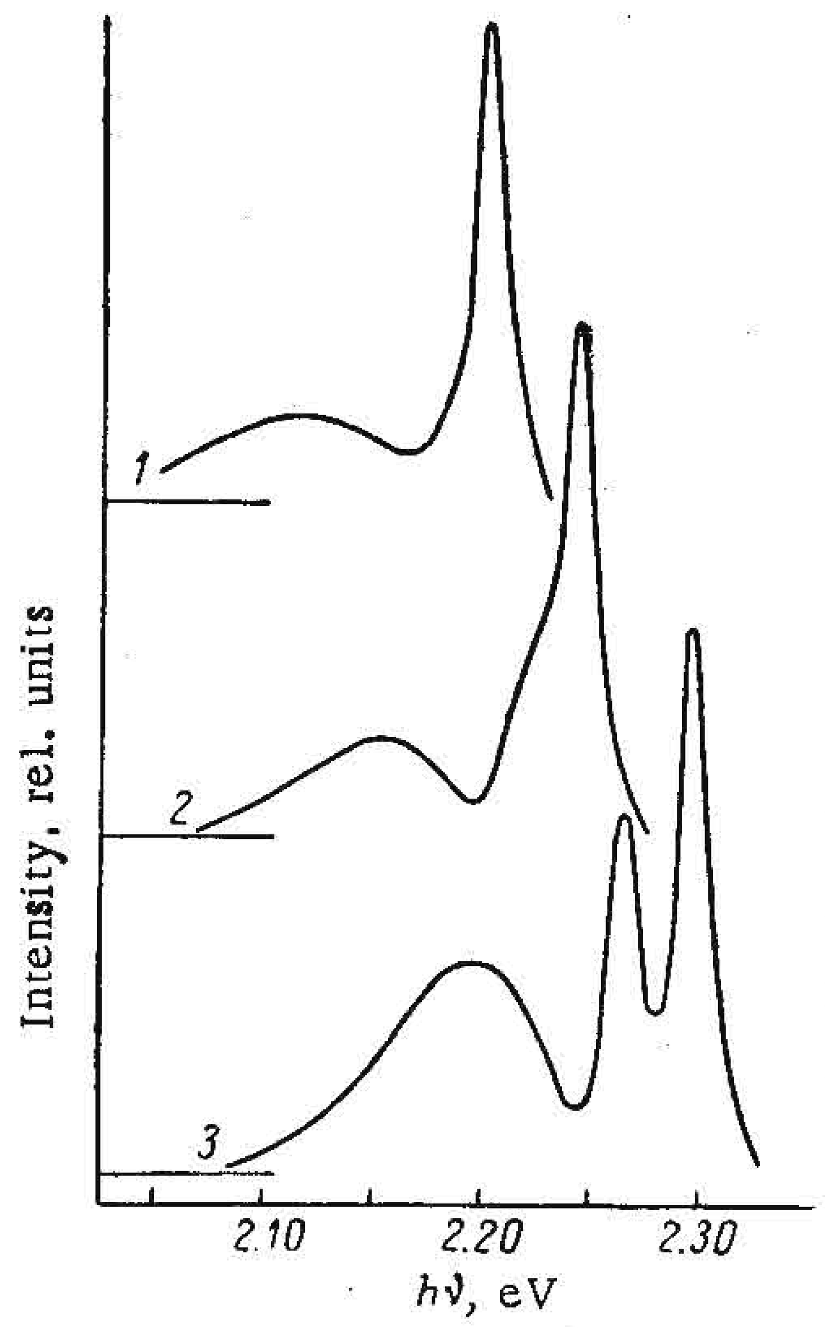


FIG. 6. Photoluminescence spectra of constant-period $Ga_{x'}In_{1-x'}As_{1-y'}P_{y'}$ solid solutions of compositions near the transition from the direct to indirect energy band structure. T = 77°K; high excitation rate. Arsenic content y': 1, 2) $y'_1 > y'_2 > 0$; 3) $y'_3 = 0$ ($Ga_{0.71}In_{0.29}P$).

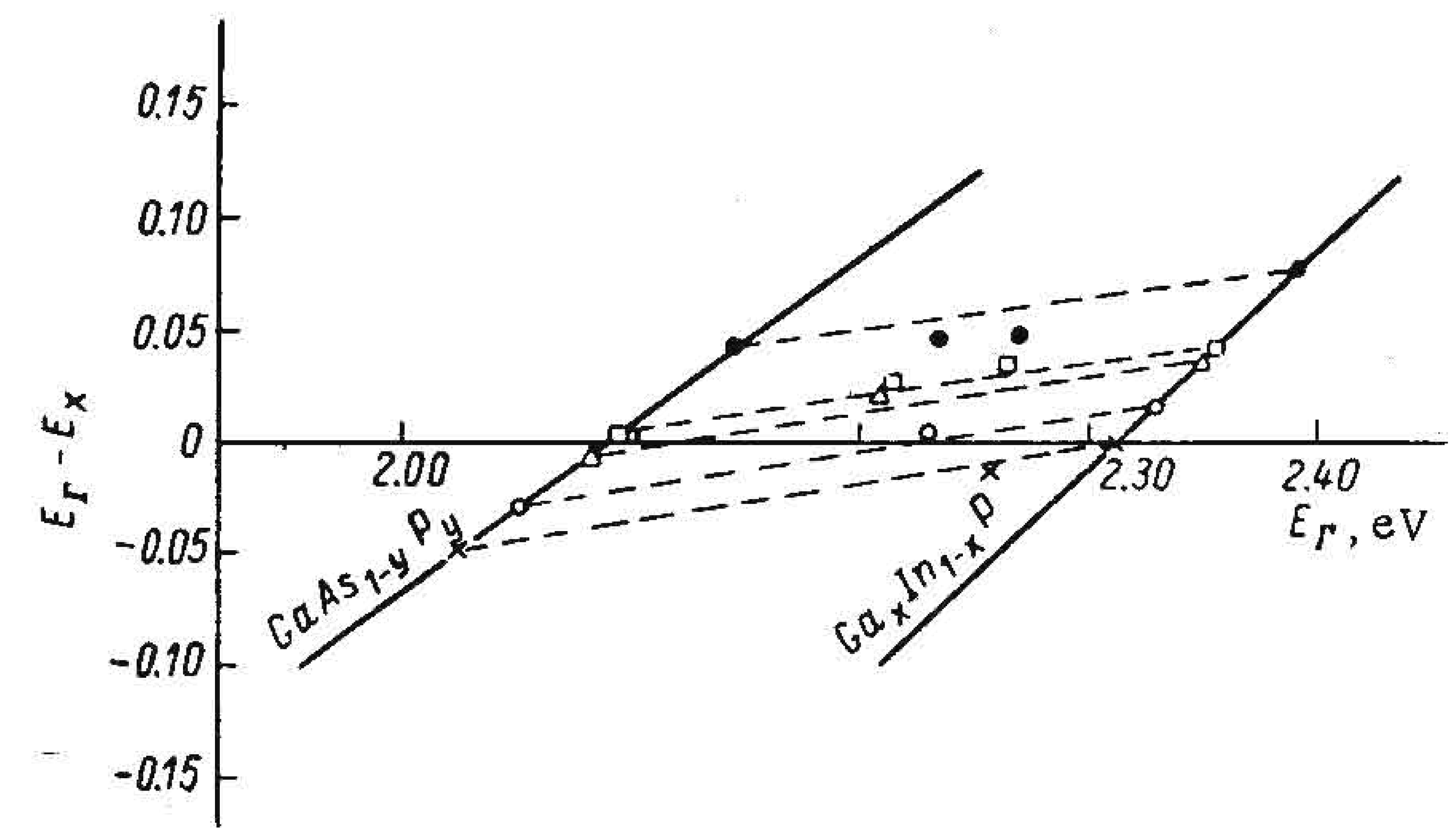


FIG. 7. Dependences of the energy gap between the Γ and X minima on the direct band gap of GaAsP (based on Ref. 9) and GaInP (based on Fig. 5) solid solutions. T = 77°K; the same symbols are used for the investigated constant-period GaInP and GaInAsP samples and the positions of the points in the case of GaAsP are calculated on the basis of the data of Ref. 9 using Eq. (1).

6 shows the photoluminescence spectra of three constant-period samples of GaInAsP solid solutions of compositions near to this transition. The presence of arsenic (spectra 2 and 3) is indicated by a considerable shift of the band A toward longer wavelengths compared with its position in the case of $Ga_{0.71}In_{0.29}P$ (curve 1). The reduction in the separation between the bands A and X − LA in the photoluminescence spectra of samples 2 and 3 shows that quaternary $Ga_{x'}In_{1-x'}As_{1-y'}P_{y'}$ solid solutions become "more direct" as the As content is increased. An analysis of the nature of the photoluminescence spectra of several sets of constant-period GaInAsP solid solutions of compositions near the transition from the direct to indirect energy band structure makes it possible to determine the configuration of the line representing this transition, discussed in the introduction (Fig. 1).

The continuous lines in Fig. 7 represent the dependences of the energy gap between the direct and indirect minima $\Delta E = E_{g\Gamma} - E_{gX}$ on the band gap (forbidden band width) $E_{g\Gamma}$ of the GaAsP and GaInP solid solutions. The dependence describing the GaAsP system is calculated on the basis of the results of Ref. 9 and the dependence $\Delta E = f(E_{g\Gamma})$ for the GaInP solid solutions is based on our measurements, plotted in Fig. 5. The results of similar measurements on the GaInAsP solid-solution system are represented in Fig. 7 by points between the continuous lines. The constant-period solid solutions grown on GaAsP substrates of the same composition are represented by the same type of symbol. We can see that they are located near the almost parallel lines (shown dashed) connecting the corresponding points of the constant-period GaAsP and GaInP solid solutions. Since in the case of the GaAsP and GaInP solid solutions in a narrow range of compositions near the point of transition from the direct to indirect energy band structure the dependences $E_{g\Gamma} = f(a)$ can be regarded as linear, the results obtained indicate that in the case of the GaInAsP solid solutions with a fixed gap $\Delta E$ (including those with $\Delta E = 0$) the band gap should vary linearly with the lattice parameter. Therefore, the boundary between the direct and indirect GaInAsP solid solutions (Fig. 1) can be represented quite accurately by a straight line.[3] On the basis of Fig. 7 we may also conclude that the rate of change of the gap between the Γ and X minima in the constant-period GaInAsP samples in the direct composition range is approximately 0.2 per 1 meV of reduction in the direct band gap.

## 3. Estimates of the Shortest Stimulated Emission Wavelengths of GaInAsP Heterojunction Lasers

We shall obtain these estimates on the basis of the data on the parameters of the shortest-wavelength lasers in which the thoroughly investigated AlAs−GaAs system is used. At 77°K ($\lambda_{st}$ = 619 nm, jth = 300 A/cm² − Ref. 23) the active region of such lasers consists of the $Al_{0.37}\cdot Ga_{0.63}As$ solid solutions, in which the gap between the Γ and X minima clearly does not exceed 10 meV (Ref. 16). On the other hand, it is known that normal operation of a heterojunction laser requires an electron confinement barrier of ~7 kT. A similar energy band configuration can be obtained using $Ga_{0.71}In_{0.29}P$−GaInAsP heterojunctions in which the active region has a band gap 50 meV smaller than the corresponding band gap of the emitter. If we assume that the shift of the stimulated emission energy relative to $E_g$ in the active region is 20 meV at 77°K, we find from Sec. 2 and Fig. 5 that the shortest stimulated-emission wavelength is $\lambda_{min} \approx 555$ nm (77°K).

At 300°K the minimum stimulated-emission wavelength of AlGaAs heterojunction lasers is 668 nm (jth = 110 kA/cm²), as reported in Ref. 24. The gap between the Γ and X minima in the active region of these lasers is at least 65 meV and the barrier for electron confinement should be 150 meV. Similar conditions can be achieved also in $Ga_{0.70}In_{0.30}P$−GaInAsP heterojunctions. The shortest stimulated-emission wavelength calculated for these junctions is 617 nm (this value is obtained after allowing for the shift of the stimulated-emission line by 40 meV relative to $E_g$).

Thus, Ga−In−As−P heterojunction lasers can clearly emit right up to the green part of the spectrum at 77°K and the shift of the short-wavelength limit compared with the AlAs−GaAs lasers is 240 meV. The corresponding shift at 300°K is somewhat smaller (160 meV) because a higher energy barrier is needed at the heterojunction to ensure efficient electron confinement at higher temperatures.

Nevertheless, injection lasers made of GaInAsP solid solutions and operating at room temperature should emit throughout all the red and some of the orange wavelengths.

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1) The value 7 meV represents the difference between the value of $E_{g\Gamma}$ and the position of the photoluminescence edge maximum in the case of relatively pure n-type samples at 77°K.

2) The composition of the samples in which this happens can be found from Eq. (3) and the position of this band maximum at room temperature.

3) In general, the discrete coordinates on this line can be found from Eq. (7) because they represent the points of intersection of the $a$ = const curves for samples with the same lattice constant (dashed lines in Fig. 7) and the abscissa.


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