



Determination of strain-induced valence-band splitting in GaAsN thin films from circularly polarized photoluminescence

著者	Egorov A. Yu., Kalevich V. K., Afanasiev M. M., Shiryaev A. Yu., Ustinov V. M., Ikezawa M., Masumoto Y.
journal or publication title	Journal of applied physics
volume	98
number	1
page range	013539
year	2005-07
権利	(C) 2005 American Institute of Physics
URL	http://hdl.handle.net/2241/98185

doi: 10.1063/1.1949718

Determination of strain-induced valence-band splitting in GaAsN thin films from circularly polarized photoluminescence

A. Yu. Egorov, V. K. Kalevich, M. M. Afanasiev,^{a)} A. Yu. Shiryaev, and V. M. Ustinov
Ioffe Physico-Technical Institute, Saint Petersburg, 194021 Russia

M. Ikezawa and Y. Masumoto
Institute of Physics, University of Tsukuba, Tsukuba 305-8571, Japan

(Received 1 February 2005; accepted 17 May 2005; published online 14 July 2005)

The paper studies the circularly polarized photoluminescence (PL) from dilute GaAsN alloys with nitrogen content of 1%–3.4%, grown on GaAs substrates. The room-temperature PL is found to consist of two bands whose splitting grows with increasing nitrogen content. The analysis of the PL circular polarization has shown that the PL bands originate from the splitting of light- and heavy-hole subbands, induced by an elastic strain in GaAsN layer. The dependence of the energy gap of unstrained GaAsN on the nitrogen content has been calculated using the measured light- and heavy-hole splittings. © 2005 American Institute of Physics. [DOI: 10.1063/1.1949718]

I. INTRODUCTION

GaAsN and InGaAsN alloys have recently been suggested as promising materials for near-infrared optoelectronics.¹ The merit of these materials is due to the strong bowing of the energy gap in the GaAs–GaN alloy system, which extends the spectral range of emission to 1.3 μm and beyond.^{1–4} In addition to the bowing effect, the energy gaps of GaAsN and InGaAsN layers grown on GaAs substrate can also be changed by the strain caused by a large lattice mismatch between the layer and substrate even at a low nitrogen content.^{4–6}

In this paper, we analyze the influence exerted by strain on the valence-band splitting and the energy gap in GaAsN layers grown on GaAs substrates, using circularly polarized photoluminescence (PL). The analysis of the PL circular polarization allowed us to identify the type of recombination transitions and determine the strain-induced valence-band splitting in GaAsN alloy. Based on the results obtained, we calculated the dependence of the energy gap on the nitrogen content of unstrained GaAsN.

The idea of our analysis is the following. In group III-V semiconductors, the interband absorption of circularly polarized light is known⁷ to be accompanied by electron-spin polarization. If the electrons do not lose the polarization during their lifetime, the recombination emission is also circularly polarized. The degeneracy at the Γ point of the upper valence band in group III-V semiconductors can be lifted by uniaxial strain. In the case of uniaxial compression, the top of a light-hole band, with the projections of the angular momentum $m = \pm 1/2$, lies above the heavy-hole band with $m = \pm 3/2$.⁸ The signs of the circular polarization of PL were found^{7–9} to be opposite for recombination involving light and heavy holes. In addition, the polarization sign of light-hole recombination is opposite to that of the polarization of the exciting beam. Therefore, by measuring the PL polarization signs we

can distinguish between the recombination transitions involving light or heavy holes in the strained GaAsN.

II. SAMPLES AND EXPERIMENTAL DETAILS

GaAsN layers were grown by rf-plasma-assisted solid-source molecular-beam epitaxy at 350–450 °C on semi-insulating (001) GaAs substrates. A series of 0.1- μm -thick GaAsN layers with the nitrogen content in the range of 0.01–0.034 was grown. GaAs_{0.965}N_{0.035} layer with a thickness of 1.5 μm was also grown for comparative study. The nitrogen content of the alloy and the crystallinity of the layers grown were examined by means of x-ray diffraction technique.

Continuous-wave Ar⁺ and tunable Ti:sapphire lasers were used for PL excitation. The PL spectra were recorded along the growth axis in the backscattering configuration, using either a Ge photodiode or an InGaAsP photomultiplier. The spin polarization of electrons was created by circularly polarized light.⁷ It was monitored by measuring the degree of circular polarization of PL, which is defined as $\rho = (I^+ - I^-)/(I^+ + I^-)$, where I^+ and I^- are the right and left circularly polarized PL components, respectively. A highly sensitive polarization analyzer¹⁰ with an InGaAsP photomultiplier, a quartz polarization modulator, and a two-channel photon counter synchronized with the polarization modulator were used to measure the circular polarization of PL up to 1.4 μm with an accuracy better than 0.1%. The PL measurements were carried out at 300 or 77 K.

III. EXPERIMENTAL OBSERVATIONS

A. X-ray diffraction analysis

The crystal perfection, pseudomorphic character of growth, i.e., the degree of crystal lattice relaxation, and the composition of the ternary solid solution were determined by high-resolution x-ray diffraction (HRXD) technique. The typical HRXD rocking curves near symmetric (004) and asymmetric (115) reflections of GaAs are shown in Figs. 1(a) and 1(b). The data for a thick layer with the nitrogen content of about 0.035 and a thin layer with the nitrogen content of

^{a)}Electronic mail: m.afanasiev@mail.ioffe.ru

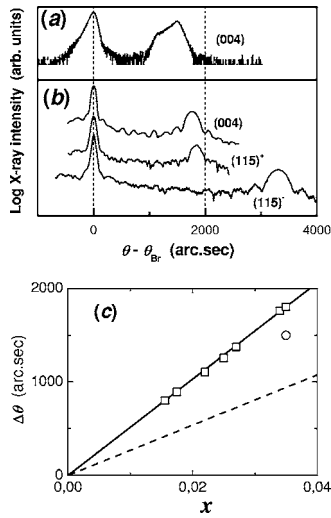


FIG. 1. X-ray rocking curves of 1.5- μm -thick $\text{GaAs}_{0.965}\text{N}_{0.035}$ (a) and 0.1- μm -thick $\text{GaAs}_{0.966}\text{N}_{0.034}$ (b) layers grown on (100) GaAs substrate, recorded near symmetric (004) and asymmetric (115) reflections of GaAs. The calculated and measured angles between peaks of HRXD rocking curves for GaAsN layers measured near symmetric (004) reflections of GaAs (c). The squares and solid line represent, respectively, the measured and calculated angles for thin elastically strained layers. The circle relates to the partly strain-relaxed 1.5- μm -thick $\text{GaAs}_{0.95}\text{N}_{0.035}$ layer. The Dashed line is the calculation for the fully strain-relaxed layers.

about 0.034 are presented in Figs. 1(a) and 1(b), respectively. The curves (115)⁺ and (115)⁻ correspond to different incidence angles of the x-ray beam $\theta_{\text{Br}} \pm \alpha$, where θ_{Br} is the Bragg angle and α is the angle between the (004) and (115) directions. The measured value of the angle between GaAsN-related diffraction peaks in (115)⁺ and (115)⁻ curves (about 1480 arc sec) for a thin layer with $x=0.034$ coincides well with the calculated value of this angle for a pseudomorphically grown layer. It evidences that the strain relaxation in the lattice of thin $\text{GaAs}_{1-x}\text{N}_x$ ($x=0.034$) does not exceed 1%, which is within the limits of the experimental accuracy. The lines in Fig. 1(c) represent the calculated angles $\Delta\theta$ between GaAs and GaAsN x-ray diffraction peaks of HRXD rocking curves near symmetric (004) reflections of GaAs for elastically strained (solid line) and fully strain-relaxed (dashed line) layers:

$$\Delta\theta = -\frac{a_{\perp} - a_0}{a_0} \text{tg}(\theta_{\text{Br}}), \quad (1)$$

where a_{\perp} is the lattice constant of the layer along the growth direction, a_0 is the GaAs substrate lattice constant, and $\theta_{\text{Br}} = 33.024^{\circ}$. The measured values of $\Delta\theta$ are shown by squares for the thin layers and by a circle for the thick layer. The strain relaxation of the thick layer leads to an increase in the lattice constant a_{\perp} , in the direction perpendicular to the layer plane, and to a reduced angle difference $\Delta\theta$ as compared with that for elastically strained GaAsN. Indeed, the measured value of $\Delta\theta$ for the thick $\text{GaAs}_{0.965}\text{N}_{0.035}$ film [circle in Fig. 1(c)] deviates significantly towards the dependence calculated for fully strain-relaxed layers (dashed line).

In the rocking curve in Fig. 1(b) recorded for the symmetric reflection, the half-width of the peak from a nitrogen-containing layer is rather small (close to that calculated for

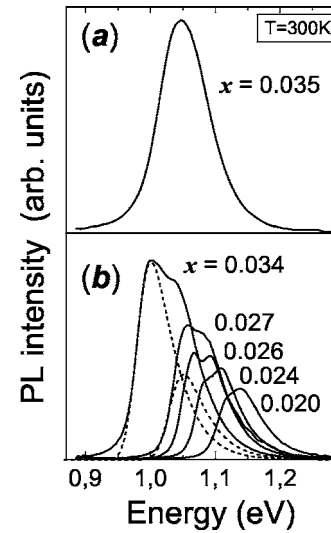


FIG. 2. PL spectra of partly relaxed (1.5- μm -thick) $\text{GaAs}_{0.95}\text{N}_{0.035}$ layer (a) and elastically strained (0.1- μm -thick) GaAsN layers with different nitrogen contents (b) grown on (100) GaAs substrate. The content of nitrogen, x , is shown near the curves. The dashed lines represent calculated curves whose sum approximates the spectrum for $x=0.034$. The curves are calculated according to formula $I(E) = C \int_0^E \exp[-(E_0 - x)^2/w^2] \exp[-(E - x)/kT] \sqrt{E - x} dx$.

the ideal case), which confirms the good homogeneity of the layer and its high crystal perfection. The presence of interference peaks in the curve indicates the planarity of the structure heterointerfaces.

Therefore, we can conclude that thin (0.1 μm) layers of GaAsN have high crystal perfection; they are elastically strained GaAsN layers grown pseudomorphically on a GaAs substrate, with a nitrogen content in the layer of up to 0.034. At the same time, a thick (1.5 μm) $\text{GaAs}_{1-x}\text{N}_x$ ($x=0.035$) layer is partly, about 40%, strain relaxed.

B. Experimental study of photoluminescence and optical orientation

Figure 2(b) shows room-temperature PL spectra of elastically strained thin GaAsN layers with different nitrogen contents. It is seen that the PL spectra shift to lower energies with increasing nitrogen content. Each PL spectrum consists of two bands. For illustration, the dashed lines represent two calculated curves whose sum approximates the PL spectrum for the sample with $x=0.034$. The energy gap between these bands increases with nitrogen content. These two bands in every PL spectrum can be interpreted as originating from the splitting of light- and heavy-hole bands in GaAsN.^{6,11,12} Indeed, the smaller lattice constant of GaAsN, as compared with that of the GaAs substrate, results in a biaxial tension of the GaAsN layer, which is equivalent to uniaxial compression along the growth axis. The uniaxial strain is accompanied by splitting of light- and heavy-hole bands⁸ as the content of N increases to become 50 meV at $x \approx 0.035$.^{4,6,12} The PL spectrum of a partly relaxed $\text{GaAs}_{0.965}\text{N}_{0.035}$ layer is shown in Fig. 2(a). It is seen that the strain relaxation results in a blueshift of the PL peak.

Spectra of PL (solid lines) and circular polarization of PL (full circles) of three thin GaAsN samples under study with nitrogen contents of 0.021, 0.027, and 0.034 are shown

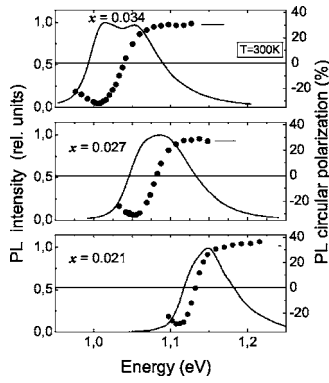


FIG. 3. PL and PL circular polarization spectra of thin GaAsN layers with nitrogen content $x=0.021, 0.027, 0.034$. Excitation energy $h\nu_{\text{exc}}=1.305$ eV ($x=0.021, 0.027$) and 1.278 eV ($x=0.034$). $T=300$ K.

in Fig. 3. It can be seen that, for all the three samples, the low-energy PL band is negatively polarized (relative to the polarization of the exciting beam), whereas the high-energy band is positively polarized.¹³

A qualitatively similar polarization spectrum (open squares) has been observed at $T=77$ K (Fig. 4). Thermalization of carriers at low temperature results in that the low-energy band peaked at 1.192 eV is predominant in the PL spectrum (solid line), and the high-energy band is represented by a weak shoulder at $1.21-1.24$ eV. The PL polarization spectrum (open squares in Fig. 4) shows that, as in the case of room temperature, the low-energy band is polarized negatively, and the high-energy band, positively. It should be noted that it is reasonable to associate the PL at $h\nu_{\text{det}} < 1.15$ eV with the density-of-states tail.¹⁴ The PL lines near 1.5 eV are associated with the emission from the GaAs barrier.

Let us correlate the signs of the circular polarization of PL, found in the experiment, and the types of recombination transitions. In strained semiconductors, this correlation has been studied in detail theoretically and experimentally (for a review, see Chap. 2 in Ref. 7; henceforth, we follow this review). The interband absorption of circularly polarized light (angular momentum of photons, $m_{\text{photon}} = \pm 1$) in semiconductors with a zinc-blende structure is accompanied by polarization of electrons in the conduction band.¹⁵ The spins of electrons excited from heavy- (projection of the angular momentum of holes at the Γ point, $m = \pm 3/2$) and light-hole ($m = \pm 1/2$) subbands are oppositely directed (as the quantization axis in strained GaAsN layers serves the direction of the uniaxial deformation, to which the directions of excita-

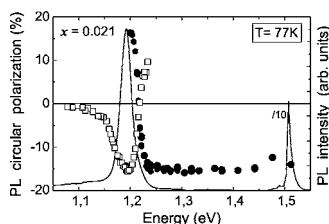


FIG. 4. PL, PL circular polarization spectra, and excitation spectrum of the PL circular polarization in $\text{GaAs}_{0.979}\text{N}_{0.021}$ layer. $T=77$ K. $h\nu_{\text{exc}}=1.57$ eV (solid line) and 1.305 eV (open squares). $h\nu_{\text{det}}=1.192$ eV (closed circles).

tion and recording of PL are parallel in our experiments). For example, the absorption of right-polarized photons ($m_{\text{photon}} = +1$) gives electrons with projections of the angular momentum $m_e = +1/2$ and $m_e = -1/2$ for the lh-c and hh-c transitions, respectively. The probability of hh-c transitions is three times that of the lh-c transitions, and, therefore, at a photon energy sufficient for simultaneous excitation of both the transitions the average spin of electrons in the conduction band has the same direction as that in the hh-c transitions, but the degree of electron polarization P_e , at the instant of generation is $P_{e0} = -50\%$. PL caused by recombination of polarized electrons is also circularly polarized. The transitions $-\frac{1}{2} \rightarrow -\frac{3}{2}$ and $\frac{1}{2} \rightarrow \frac{3}{2}$ into the heavy-hole subband are possible, with emission of σ^+ and σ^- photons, respectively. The transitions $\frac{1}{2} \rightarrow -\frac{1}{2}$ and $-\frac{1}{2} \rightarrow \frac{1}{2}$ into the light-hole subband, with emission of σ^+ and σ^- lights, are allowed. The intensities of the c-hh and c-lh transitions are related as 3:1. Therefore, the degrees of circular polarization of PL with assistance of light and heavy holes have the same absolute values, but opposite signs: $\rho_{\text{lh}} = P_e$, $\rho_{\text{hh}} = -P_e$. It should be emphasized that, in the case of excitation from both subbands, the sign of polarization of the emission involving light holes is opposite to that of the exciting light.

The PL polarization spectra in Figs. 3 and 4 were measured at simultaneous excitation of electrons from both valence subbands. Thus, the negative polarization of the low-energy PL line and the positive polarization of the high-energy PL line demonstrate unambiguously that the low- and high-energy PL bands are due to recombination involving light and heavy holes, respectively.

This conclusion is also confirmed by the analysis of the excitation spectrum of the PL polarization, as shown in Fig. 4 by full circles. This spectrum was measured at the peak of the low-energy PL line ($h\nu_{\text{det}}=1.192$ eV). It can be seen that the polarization of PL is negative and remains virtually constant as the excitation photon energy $h\nu_{\text{exc}}$ decreases from 1.45 to ≈ 1.23 eV. If $h\nu_{\text{exc}}$ is lowered further, the polarization of PL decreases sharply and its sign changes to positive at $h\nu_{\text{exc}}=1.216$ eV. Such a shape of the excitation spectrum can be only understood on the assumption that the low-energy PL band, at whose center the spectrum was measured, is due to recombination processes involving light holes. Indeed, the lh-c and hh-c transitions are simultaneously excited at $h\nu_{\text{exc}} > 1.23$ eV. In this case, the polarization of the c-lh recombination must be negative under excitation with σ^+ light because it is determined by the negative direction of the spin of electrons excited from the heavy-hole subband ($-\frac{3}{2} \rightarrow -\frac{1}{2}$ transition). As $h\nu_{\text{exc}}$ becomes lower than 1.23 eV, the hh-c transitions start to “switch off.” At $h\nu_{\text{exc}} < 1.216$ eV, only the electrons from the light-hole subband are excited ($-\frac{1}{2} \rightarrow \frac{1}{2}$ transition). The polarization of these electrons is positive, which gives a positive sign of the PL polarization ($\frac{1}{2} \rightarrow -\frac{1}{2}$ recombination transitions).

Thus, analysis of both PL polarization spectra and excitation spectra of PL polarization shows that the light- and heavy-hole subbands in thin strained GaAsN layers grown on GaAs substrates are split at the Γ point, with the top of the light-hole subband lying above the top of the heavy-hole subband.

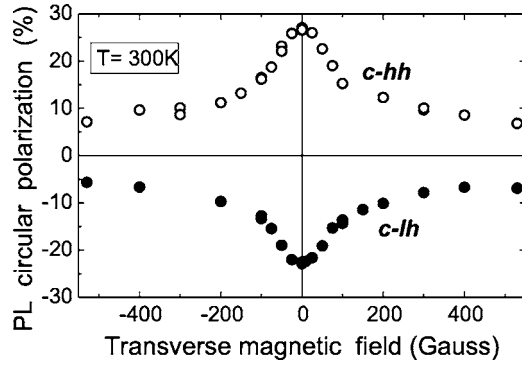


FIG. 5. Hanle effect in $\text{GaAs}_{0.979}\text{N}_{0.021}$. $T=300\text{ K}$. $h\nu_{\text{exc}}=1.305\text{ eV}$. $h\nu_{\text{det}}=1.164\text{ eV}$ (open circles) and 1.112 eV (closed circles).

The spin-related nature of the circular polarization of PL, observed in GaAsN alloys, is confirmed by its diminishing in a transverse magnetic field (Hanle effect⁷) (see Fig. 5). The magnetic depolarization curves measured at room temperature in a thin $\text{GaAs}_{0.979}\text{N}_{0.021}$ layer near PL peaks caused by a recombination of polarized electrons with light and heavy holes are shown in Fig. 5. The fact that the half-widths of both the curves are equal confirms the assumption that the polarization in both the PL bands is due to polarization of electrons in the conduction band. The Hanle effect for electrons is described by a Lorentz curve with half-width at half maximum, $B_{1/2}=\hbar/g\mu_B T_s$, where g is the Lande g factor, μ_B is the Bohr magneton, and T_s is the lifetime of the electron spin.⁷ The value of $B_{1/2}\sim 100\text{ G}$, found from the curves in Fig. 5, makes it possible to evaluate the scaled electron-spin lifetime $gT_s\sim 10^{-9}\text{ s}$.

IV. CALCULATION OF THE ENERGY GAP IN UNSTRAINED GaAsN

Figure 6 shows the energy positions of the peaks of both bands in the PL spectra [Fig. 2(b)] of samples with different contents of nitrogen. It can be seen that, as the nitrogen content increases, the spectral splitting of the PL bands grows to become 50 meV for $x=0.034$. The experimental dependences in Fig. 6 are well approximated with curves (solid lines in Fig. 6) calculated using the dispersion relation obtained in the frame of the band anticrossing (BAC) model:¹⁶

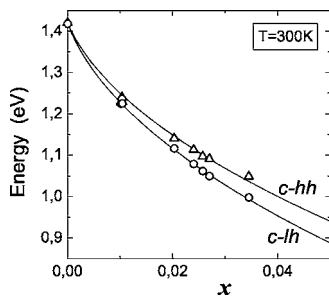


FIG. 6. Energy of the PL peaks in elastically strained GaAsN layers vs the nitrogen content. The solid lines $c\text{-hh}$ and $c\text{-lh}$ are calculated using Eq. (2).

$$E_{-}(k) = \frac{1}{2} [E^C(k) + E^N - \sqrt{(E^C(k) - E^N)^2 + 4V^2x}], \quad (2)$$

where x is the nitrogen content, $E^C(0)=1.42\text{ eV}$ is the band gap of GaAs at room temperature, and E^N is the energy of the localized states caused by the substitutional N atoms. E^N is known to be 0.23 eV above the GaAs conduction-band edge.¹⁷ The adjustable hybridization parameter V describes the coupling between the localized states and the conduction-band states of the GaAs matrix. The fitting curves $c\text{-lh}$ and $c\text{-hh}$ in Fig. 6 have been calculated at $V=2.86\text{ eV}$ and $V=2.61\text{ eV}$, respectively. These values of V are close to $V=2.7\text{ eV}$ reported recently for similar samples.¹⁷ However, the BAC model was developed for an unstrained material and therefore the strain-induced changes of the energy bands in the films under study should be taken into account for a correct application of the BAC model.

The pseudomorphic growth of elastically strained layers leads to a biaxial strain ε_{\parallel} in the growth plane and to a uniaxial strain ε_{\perp} in the growth direction,¹⁸

$$\varepsilon_{\parallel,\perp} = \frac{a_{\parallel,\perp}}{a} - 1, \quad (3)$$

where

$$a_{\parallel} = a_0, \quad (4a)$$

$$a_{\perp} = a \left[1 - D \left(\frac{a_{\parallel}}{a} - 1 \right) \right], \quad (4b)$$

$$D^{001} = 2 \frac{C_{12}}{C_{11}}. \quad (4c)$$

Here, a_0 is the lattice constant of the substrate, and a is the lattice constant of a freestanding (unstrained) layer of the composition under study. The constant D depends on the elastic constants of the material and on the orientation of the substrate. D^{001} is the constant D for the (001) orientation of the interface.

The strain in the GaAsN layer grown on the GaAs substrate can be regarded as having two components: shear and hydrostatic. The hydrostatic component of the strain is responsible for the shift of the valence- and conduction-band edges:¹⁸

$$\Delta E_v^{\text{hy}} = a_v(\varepsilon_{\perp} + 2\varepsilon_{\parallel}), \quad (5a)$$

$$\Delta E_c^{\text{hy}} = a_c(\varepsilon_{\perp} + 2\varepsilon_{\parallel}), \quad (5b)$$

where a_v , a_c are the hydrostatic deformation potentials for the valence and conduction bands. The shear component leads, via the spin-orbit interaction, to splitting of the valence-band edge at the Γ points into the light- and heavy-hole subbands. At the (001) orientation of the substrate, the energy shifts of the light- and heavy-hole subbands with respect to the energy of heavy holes are given by¹⁸

$$\Delta E_{\text{hh}}^{\text{sh}} = \frac{1}{2} \delta E^{\text{sh}}, \quad (6a)$$

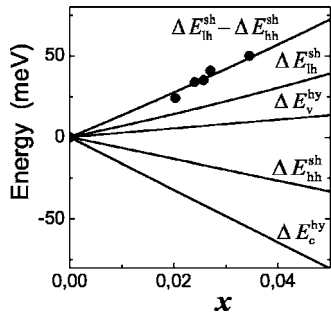


FIG. 7. Changes of the energies of the conduction-band bottom (ΔE_c^{hy}), tops of the valence band (ΔE_v^{hy}) and light- ($\Delta E_{\text{lh}}^{\text{sh}}$) and heavy-hole ($\Delta E_{\text{hh}}^{\text{sh}}$) subbands, splitting between the light- and heavy-hole subbands ($\Delta E_{\text{lh}}^{\text{sh}} - \Delta E_{\text{hh}}^{\text{sh}}$) in strained GaAsN layers vs the nitrogen content under hydrostatic and shear components of the elastic strain. The solid lines correspond to calculations and the points to the measured energy splittings of the PL bands.

$$\Delta E_{\text{lh}}^{\text{sh}} = -\frac{1}{2}\Delta_0 + \frac{1}{4}\delta E^{\text{sh}} + \frac{1}{2}\left[(\Delta_0)^2 + \Delta_0\delta E^{\text{sh}} + \frac{9}{4}(\delta E^{\text{sh}})^2 \right]^{1/2}, \quad (6b)$$

where Δ_0 is the spin-orbit splitting in the absence of strain, and δE^{sh} is the shift associated with strain. For the case in question, in which the substrate is oriented in the (001) plane, this shift is given by

$$\delta E_{001}^{\text{sh}} = 2b(\varepsilon_{\perp} - \varepsilon_{\parallel}), \quad (7)$$

where b is the potential of the shear tetragonal deformation. The conduction band at the Γ point is not affected by the shear component of the strain.

It follows from Eqs. (6a) and (6b) that the energy gap of strained GaAsN is determined by the energy at which lies the top of the light-hole subband. Thus, the low-energy band in the PL spectra of the GaAsN layer in Fig. 2(b) corresponds to a radiative recombination of an electron and a light hole, and the high-energy band, to the recombination of an electron and a heavy hole, which was confirmed in Sec. III A by the analysis of the polarization of these PL bands.

Using Eqs. (3)–(7), we calculated how the shift of energy levels and the energy gap between the tops of the heavy- and light-hole bands depend on the nitrogen content in the layers under study. The constants $a_c = -7.17$ eV, $a_v = 1.16$ eV, $C_{11} = 1.18 \times 10^{12}$ dyn/cm², $C_{12} = 0.54 \times 10^{12}$ dyn/cm², $\Delta_0 = 0.34$ eV, and $b = -1.7$ eV for GaAs were taken from Ref. 18. The effect of incorporated nitrogen on the elastic constants and the spin-orbit splitting Δ_0 was disregarded. The results of the calculation are shown in Fig. 7 by solid lines. The experimental energy gaps between the PL bands (Fig. 2) are shown in Fig. 7 by full circles. The good agreement between the theoretical ($\Delta E_{\text{lh}}^{\text{sh}} - \Delta E_{\text{hh}}^{\text{sh}}$ curve) and experimental energy gaps suggests that the model described above and the constants of gallium arsenide can be used to evaluate the band splitting in thin strained GaAsN layers.

The energy gap in thin strained GaAsN layers grown on GaAs substrates corresponds to the energy of an electron-hole transition involving a light hole (fitting curve c -lh in Figs. 6 and 8). Calculations of the energy gap for unstrained

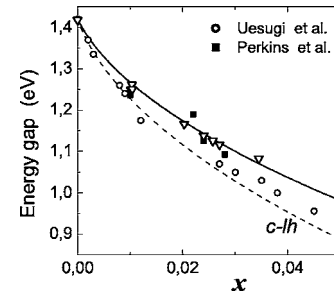


FIG. 8. The forbidden band gap in elastically strained GaAsN layers on GaAs (dashed line) and in freestanding (unstrained) GaAsN alloy (open triangles) vs the content of the incorporated nitrogen, x . The solid line is calculated using Eq. (2) with $V = 2.42$ eV. \circ and \blacksquare : experimental data for thin (see Ref. 19) and thick (see Ref. 20) GaAsN layers grown on GaAs, respectively.

(freestanding) GaAsN were performed for all the measured energy-gap values. The calculated data are presented by open triangles in Fig. 8. The solid line in Fig. 8 is the fitting curve calculated according to Eq. (2) (BAC model) with $V = 2.42$ eV. The energy gap of freestanding GaAsN steadily decreases from 1.42 to 1.08 eV as the nitrogen content grows from 0% to 3.4%. The difference in energy gap between the freestanding and elastically strained $\text{GaAs}_{0.966}\text{N}_{0.034}$ is as much as 90 meV. That is a quarter of the total energy-gap reduction.

In addition to our results, Fig. 8 shows the experimental data obtained by Uesugi *et al.*¹⁹ and Perkins *et al.*²⁰ on thin and thick GaAsN layers grown on GaAs substrates, respectively. It can be seen that the data obtained by Uesugi *et al.* on thin layers (open circles) are in a good agreement with our experimental dependence of the energy gap (dashed line), whereas the results of Perkins *et al.*, obtained on thick (relaxed) layers (full squares), well coincide with our calculated curve (solid line). This coincidence confirms also the adequacy of the approach we chose.

In conclusion, we observed two bands in PL spectra of elastically strained ternary GaAsN solid solutions grown on GaAs substrates. As demonstrated by an analysis of their circular polarizations, these bands are associated with electron-hole transitions involving light and heavy holes. The energy gap between the peaks of these bands corresponds to the calculated splitting of light- and heavy-hole subbands induced by the elastic strain of the GaAsN layer. The dependence of the fundamental energy gap on the nitrogen content for unstrained GaAsN alloys has been calculated, taking into account the effect of the elastic strain on the position of the energy bands.

ACKNOWLEDGMENTS

This study was partially supported by the Program of the Russian Ministry of Science and Technology, Russian Foundation for Basic Research, and Nanoscience Special Project of the University of Tsukuba. The authors are grateful to K.V. Kavokin and X. Marie for useful discussions.

¹M. Kondow, T. Kitatani, S. Nakatsuka, M. C. Larson, K. Nakahara, Y. Yazawa, M. Okai, and K. Uomi, IEEE J. Sel. Top. Quantum Electron. **3**, 719 (1997).

- ²L. Bellaiche, S. H. Wei, and A. Zunger, *Appl. Phys. Lett.* **70**, 3558 (1997); P. R. C. Kent, G. L. W. Hart, and A. Zunger, *ibid.* **81**, 4377 (2002).
- ³A. Yu. Egorov, A. E. Zhukov, and V. M. Ustinov, *J. Electron. Mater.* **30**, 477 (2001).
- ⁴Y. Zhang, A. Mascarenhas, H. P. Xin, and C. W. Tu, *Phys. Rev. B* **61**, 4433 (2000).
- ⁵M. H. Ya, W. Z. Cheng, Y. F. Chen, and T. Y. Lin, *Appl. Phys. Lett.* **81**, 3386 (2002).
- ⁶A. Yu. Egorov, E. S. Semenova, V. M. Ustinov, Y. G. Hong, and C. Tu, *Semiconductors* **36**, 981 (2002).
- ⁷*Modern Problems in Condensed Matter Sciences*, edited by F. Meier and B. Zakharchenya (North-Holland, Amsterdam, 1984), Vol. 8.
- ⁸G. L. Bir and G. E. Pikus, *Symmetry and Strain-Induced Effects in Semiconductors* (Wiley, New York, 1974).
- ⁹M. I. Dyakonov and V. I. Perel, *Fiz. Tekh. Poluprovodn. (S.-Peterburg)* **7**, 2335 (1973).
- ¹⁰V. D. Kul'kov and V. K. Kalevich, *Instrum. Exp. Tech.* **24**, 1265 (1981).
- ¹¹V. K. Kalevich, M. Ikezawa, A. Yu. Shiryaev, A. Yu. Egorov, V. M. Ustinov, and Y. Masumoto, *Nanostructures: Physics and Technology*, Proceedings of the 11th International Symposium, St.-Petersburg, Russia, 23–28 June 2003 (Ioffe Physico-Technical Institute, St. Petersburg, Russia, 2003) (unpublished), pp. 277–278; <http://edu.ioffe.ru/conf/nano2003/nano2003pgm.html>
- ¹²V. K. Kalevich, M. Ikezawa, A. Yu. Shiryaev, A. Yu. Egorov, V. M. Ustinov, and Y. Masumoto, Abstracts of the Physical Society of Japan, 2003 Autumn Meeting, Okayama, Japan, 20–23 September 2003 (unpublished), Vol. 58, p. 4, p. 574.
- ¹³First observation of optical orientation in thin GaAsN layers was reported in Refs. 11 and 12.
- ¹⁴I. A. Buyanova, W. M. Chen, G. Pozina, J. P. Bergman, and B. Monemar, *Appl. Phys. Lett.* **75**, 501 (2002).
- ¹⁵Polarization of photoexcited holes can be neglected because of their fast spin relaxation in the valence band (Ref. 7).
- ¹⁶W. Shan, W. Walukiewicz, J. W. Ager III, E. E. Haller, J. F. Geisz, D. J. Friedman, J. M. Olson, and S. R. Kurtz, *Phys. Rev. Lett.* **82**, 1221 (1999).
- ¹⁷W. Walukiewicz, *Physica E (Amsterdam)* **20**, 300 (2004).
- ¹⁸M. P. C. M. Krijn, *Semicond. Sci. Technol.* **6**, 27 (1991).
- ¹⁹K. Uesugi, N. Morooka, and I. Suemune, *Appl. Phys. Lett.* **74**, 1254 (1999); *J. Cryst. Growth* **201/202**, 355 (1999).
- ²⁰J. D. Perkins, A. Mascarenhas, Y. Zhang, J. F. Geisz, D. J. Friedman, J. M. Olson and Sarah R. Kurtz, *Phys. Rev. Lett.* **82**, 3312 (1999).