Structure defects and photoluminescence of epitaxial $In_xGa_{1-x}As$ films

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The methods of low-temperature photoluminescence, x-ray topography, and diffractometry were used in a study of the influence of the solid-solution composition on the structure and properties of $In_xGa_{1-x}As$ ($0.002<x<0.01$) films grown by molecular beam epitaxy.

An increase in the degree of isovalent doping of epitaxial films results in relaxation of internal stresses by formation of misfit dislocations and transformation of the photoluminescence spectrum in the range $1.3 \leq h\nu \leq 1.5$ eV, which is accompanied by nonmonotonic behavior of the intensity of the edge band and of the quantum efficiency of the photoluminescence due to changes in the number of the nonradiative recombination centers and a redistribution of the residual acceptor carbon impurity (C_{As}) with a depth across a film because of the gettering influence of the misfit dislocations.

The idea of doping GaAs single crystals in epitaxial films with the isovalent In impurity, put forward in Refs. 1-3, postulates a replacement of gallium vacancies with indium, $V_{Ga} \rightarrow [In_{Ga}]$, because these vacancies are nonradiative recombination centers.[4] This increases the luminescence efficiency in a certain range of compositions. Introduction of In creates microstresses because of the large difference between the atomic radii of In ($r_{In} = 1.44$ Å) and of Ga ($r_{Ga} = 1.26$ Å) and this in turn can generate defects and transform the existing system of point defects. These two processes are mutually related and they depend on the technology used to grow epitaxial films,[5] purity (concentration of the initially present residual C and/or Si impurities), dopant concentration, nature and magnitude of the strain fields ε governed by the epitaxial film thickness, and mismatch between the lattice parameters of the film and the substrate.

We studied how the solid-solution composition and the increasing mismatch between the lattice constants of the film and the substrate as a result of increasing the concentration of the isovalent indium impurity in epitaxial $In_xGa_{1-x}As$ ($0.002 \leq x \leq 0.01$) films grown by the molecular beam technology influenced the photoluminescence spectra and how this is related to a possible relaxation of internal mechanical stresses by formation of misfit dislocations at the interface between the film and the substrate or to the formation of additional defects.

EXPERIMENTAL METHODS

We investigated $In_xGa_{1-x}As$ ($0.002 \leq x \leq 0.01$) films grown on high-resistivity (100) GaAs substrates by the method of molecular beam epitaxy (MBE) at ~600°C. The film thickness was 1.1-1.4 μm. The density and mobility of free electrons, deduced from the Hall effect on the assumption that the Hall factor was 1, amounted to $n = 8 \times 10^{17}$ cm^{-3} and $\mu < 1000$ cm^2/(V·s) for all the investigated samples.

The steady-state photoluminescence was excited by radiation from an LG-503 argon laser which emitted at the pump wavelengths λ_p amounting to $\lambda_1 = 4880$ and $\lambda_2 = 5145$ Å (excitation density was between 5 and 50 W/cm^2) and from an He—Ne laser with $\lambda_3 = 6328$ Å (excitation density <10 W/cm^2). The recombination radiation was recorded in the 0.8 to 1.15-μm range using an uncooled FÉU-62 photomultiplier in the current-flow regime as part of an automated KSV-23 system. Measurements were carried out at temperatures of 4.2-80 K and 300 K.

The structural quality of the epitaxial films was monitored by x-ray reflection topography (Cu$K\alpha$ radiation in an asymmetric configuration, 531 reflection) and by x-ray diffractometry using a two-crystal spectrometer and Cu$K\alpha$ radiation in the $(n, -m)$ geometry. These two methods made it possible to determine the film thicknesses, the profiles of the lattice parameter across the thickness,[6] and macrobending of the system. The composition and electrophysical parameters of the batches of samples used are listed in Table I. Each batch consisted of 4-12 samples made in the same technological cycle.

EXPERIMENTAL RESULTS AND DISCUSSION

X-ray topographs of our samples showed that when the mismatch between the lattice constants of the film a_1 and substrate a_2 was minimal (batches 1 and 2) the nucleation of misfit dislocations was not yet observed and the structural

TABLE I. Initial parameters of epitaxial $In_xGa_{1-x}As$ films

Batch No.	Substrate designation	Bending radius R, m	Misfit dislocations	$\Delta a/a_2$	Epitaxial film (EF)			
					EF thickness, μm	density of free carriers, cm^{-3}	mobility μ, cm^2/(V·s)	indium content x, mol.%
1 (88)	AGChP	+23	none	$9.9\cdot10^{-4}$	1.4	$8\cdot10^{17}$	<1000	0.2
2 (89)	AGChP	+30	none	$1.27\cdot10^{-3}$	1.1	$8\cdot10^{17}$	<1000	0.4
3 (90)	AGChP	−150	few	$1.48\cdot10^{-3}$	1.45	$6\text{-}8\cdot10^{17}$	<1000	0.42
4 (91)	AGChP	+152	network	$1.85\cdot10^{-3}$	1.3	$5\text{-}8\cdot10^{17}$	<1000	0.9

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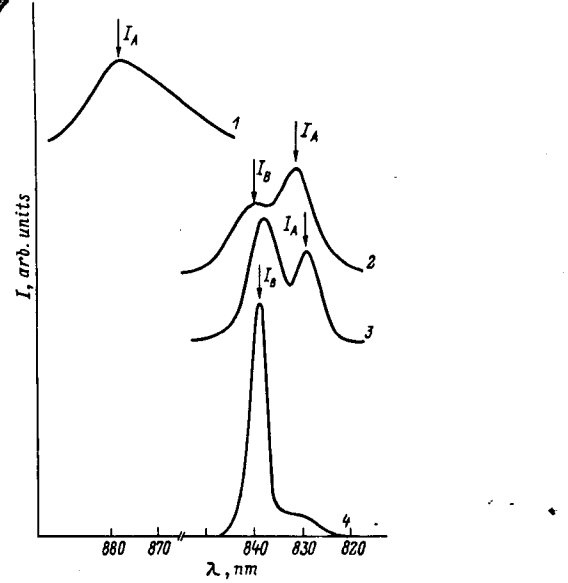

FIG. 3. Photoluminescence spectra of epitaxial $In_{0.01}Ga_{0.99}As$ films (batch 4) determined at the following temperatures (K): 1) 300; 2) 80; 3) 40; 4) 6. The photoluminescence was excited by pumping at the wavelength λ_p = 4880 Å.

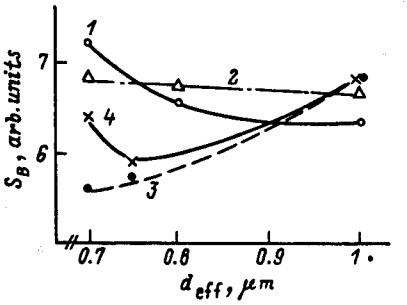

FIG. 4. Dependences of the ratio of the photoluminescence band intensities I_B/I_A on the depth of probing d_{eff} obtained for batches 1-4 (curves 1-4).

network. A nonmonotonic change in the intensity of the edge band I_A was accompanied by a nonmonotonic change in the external quantum efficiency of the photoluminescence $\eta = \int I(\lambda)d\lambda$, accompanied by a reduction in the ratio of the band intensities $S_B = I_B/I_A$ (S_B was proportional to the ratio of the concentrations of the luminescence-emitting centers). The nonmonotonic decrease in the ratio of the intensities of the photoluminescence lines (impurity and edge) with an increase in the In content could indicate a reduction in the concentration N_B of the shallow acceptors (batches 1-3) upon introduction of the isoelectronic impurity into gallium arsenide on the assumption that the capture cross section remained constant. The shallow acceptor centers involved in the radiative recombination of free carriers were usually attributed to the presence of residual (accidental) impurities (carbon and/or silicon) and such a reduction in the number of the centers could be due to changes in the acceptor (residual impurity) system because of changes in the structural quality of the films. The appearance of misfit dislocations at the film—substrate interface could result in a redistribution of the residual impurities in such a way that they would move away from the interface. The acceptor ionization energy E_A calculated from the position of the luminescence band $h\nu_B(e-A^0)$ was E_A = 27 meV at T = 4.2 K, which agreed with the ionization energy of the site carbon C_{As}. (Since the technology used in the preparation of the films belonging to different batches was the same and only the In concentration — itself of high purity — was varied, we could reject the possibility of large fluctuations in the carbon content due to the irreproducibility of the technology.)

A redistribution of carbon across the film thickness was checked by determining the ratio S_B in the full range of variation of x. This was done by increasing the depth of probing.[2)] The photoluminescence was excited by the λ_2 = 5145 and λ_3 = 6328 Å wavelengths. The rate of excitation was the same in each case. The probing depth d_{eff} was increased from ~0.7 μm (λ_1, at 80 K) to ~1.0 μm (λ_3, at 80 K). An increase in this depth failed to reveal a significant change in the carbon impurity concentration in samples from batches 1 and 2, but formation of misfit dislocations resulted in a redistribution of carbon with an increase in $N_{C_{As}}$ near the interface and its decrease near the surface (Fig. 4). Since a reduction in the rate of excitation due to an increase in d_{eff} could alter the ratio $S_B = I_B/I_A$ by increasing it, because of the different dependences of the edge and impurity luminescence bands on the excitation rates, we determined the values of S_B and found that an increase in the rate of excitation by a factor of 1.5-2 altered S_B by no more than 3-4%. Oblique section were formed by chemical thinning and the whole cycle of measurements at the wavelength λ_1 was repeated for an oblique section in order to determine whether such a redistribution of carbon was accompanied by a redistribution of the In isovalent impurity with depth in an epitaxial film, and also in order to exclude the influence of the rate of excitation on the qualitative estimates of the residual impurity distribution. This was done for all the samples and the results confirmed that carbon was gettered by misfit dislocations at the interface. There were no changes in the In content, which was deduced from the shift of the photoluminescence maximum (within the limits of the sensitivity of the method amounting to 0.002 eV, which corresponded to a change in the In concentration by ~1 × 10^{19} cm^{-3}).

Our results thus established that relaxation of internal stress in epitaxial $In_xGa_{1-x}As$ films on GaAs substrates occurred as a result of the mismatch between the lattice constant of the film and the substrate at a given film thickness by introducing upon increasing the isovalent dopant concentration, misfit dislocations in accordance with a mechanism that should be studied by the method of transmission electron microscopy. This was accompanied by a redistribution of the strain fields of the system (Table I) and a redistribution of the residual acceptor impurity. Bearing in mind the sign of the band broadening caused by an increase in the concentration of carbon and the relationship between the covalent radii (r_C = 0.77 Å) characterized by $r_C < r_{Ga}$, we concluded that the carbon impurity in its free state could migrate in the field of internal stresses along the field gradient toward the compressed region, in agreement with the distribution of the strain fields in the film—substrate system of samples from batches 3 and 4 (Ref. 11).

The application of the x-ray diffraction, x-ray topography, and the photoluminescence methods to the same objects made

it possible to establish clearly and unambiguously the correlation between the nonmonotonic variation in the intensity of the photoluminescence of InGaAs/GaAs epitaxial film systems grown by the molecular beam epitaxy method and changes in the dislocation structure of the interface.

We thus found that isovalent doping of $In_xGa_{1-x}As$ ($0.002 \leq x \leq 0.01$) epitaxial films grown by the molecular beam epitaxy method caused an increase, against the background of a redistribution of the strain fields and relaxation of these fields by the formation of misfit dislocations, in the intensity of the edge luminescence band and of the external luminescence efficiency ($x \leq 0.004$) and then their decrease in the range $x > 0.004$, as a result of a change in the concentration of the nonradiative recombination centers and a redistribution in the concentration of the carbon acceptor impurity in the bulk of the films, which was attributed to the gettering influence of the misfit dislocations.

[1)]The mismatch was defined as $(a_1 - a_2)/a_2$.

[2)]The probing depth was estimated from $d_{\text{eff}} = 1/\alpha_n + L_D$, where L_D is the diffusion length of the minority carriers assumed to be ≈ 0.5 μm (Ref. 9) and α_n is the absorption coefficient at the wavelength λ_n (Ref. 10).

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