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# Morphological stability in epitaxy of monolithically integrated optical devices

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Three technological methods: LPE, MO CVD, and MBE have been compared from the point of view of their crystallization peculiarities related to the fabrication of optical monolithically integrated devices. Ignoring the dependence of the crystal growth rate on the substrate orientation, the morphological stability functions of Mullins and Sekerka for each epitaxy process have been calculated for $GaAs-Al_xGa_{1-x}As$ materials system. It has been shown that MBE is the most suitable crystallization method for monolithic optical integration, MO CVD being the method less suitable.

## 1. Introduction

Guided-wave optics and thin-film technology are the two basic elements of integrated optoelectronics. The integration of components, which comprises an optical communication system onto a single substrate offers the possibility of enhanced data rates and reduced sensivity to external disturbing influences (temperature, vibrations, etc.) [1].

In the monolithic approach to optical integration we use one materials system which works well for all optical components. The essence of this integration is that we deal with guided waves in three-dimensional channel waveguides formed by monocrystalline double heterojunction (DH) structures. A DH structure consists of a thin guiding layer with refractive index $n_1$, sandwiched between two cladding layers with refractive indices $n_2 < n_1$ (Fig. 1). By changing the shape of the surface of the DH structure guiding layer, different optical functional elements can be created.

Optical monolithically integrated devices are now made of semiconducting binary, ternary or quaternary III–V, II–VI and IV–VI compounds. Most frequently, however, they are made of $GaAs-Al_xGa_{1-x}As$, $Inp-Ga_xIn_{1-x}P_yAs_{1-y}$ and $PbTe-Pb_{1-x}Sn_xTe$ materials systems. The application of these materials systems is due to the fact that these materials allowed to achieve a good qualities of all important optoelectronic functions, like generation, guiding, modulation and detection of light, and that their operation wavelengths (Fig. 1) are well adapted to the requirements raised by the technical applications, for example, by modern glass fibres optical communication or by environmental polution measurements services.

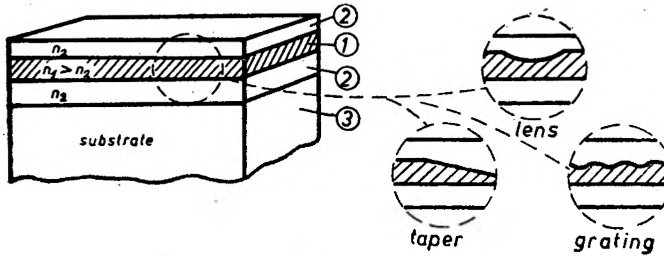
The physical parameters which define the quality of a DH structure used in optical integration are the following:

— abruptness of the heteroboundaries,

— density of surface energy states,

— crystalline perfection of the guiding and cladding layers, and

— doping according to preferentially designed spatial distribution of impurities.




| $\lambda_{oper}$ [μm] : | ≈ 0.87 | ≈ 1.1 | ≈ 8 |
|---|---|---|---|
| ① waveguide : | GaAs | $Ga_x In_{1-x} P_y As_{1-y}$ | $Pb_{1-x}Sn_xTe$ |
| ② cladding : | $Al_xGa_{1-x}As$ | InP | PbTe |
| ③ substrate : | GaAs | InP | PbTe |

**Fig. 1. A typical double heterojunction structure used as a basic construction element in monolithically integrated optoelectronics. By changing the surface shape of the guiding layer, different optical functional elements can be created. The materials system, most frequently used in practice, are indicated together with their operation wavelengths**

These parameters influence the electrical and optical confinement, the internal quantum efficiency of carriers radiative recombination, the reliability of elements and functional structures, and the light propagation losses in an integrated optical device.

There are three epitaxial growth techniques used in practice for fabrication of optoelectronic monolithically integrated devices.

Liquid Phase Epitaxy (LPE) consists of a series of growth steps from individual supersaturated melts or solutions [2]. The growth steps are connected by a series of transfer (usually sliding) steps of the substrate between the melts. LPE has proven to be a relatively simple technique giving very good quality of the DH structures, well suited to small production efforts, where scale, yield (uniformity and reproducibility), and economics play a little role in determining the optimum growth technique.

Metalorganic Chemical Vapour Deposition (MO CVD) is a crystallization method, in which the constituent elements for the growth are introduced into the growth chamber in the form of gaseous metalorganic alkyl compounds, and in which the layer deposition results from thermal decomposition (pyrolysis) of these gaseous compounds and a subsequent growth reaction between

the component elements on the substrate platelet [3]. MO CVD seems to be the ideal technique for large-scale commercial heterostructure growth operations. It gives an excellent control of composition as well as of surface defects of the grown layers.

Molecular Beam Epitaxy (MBE) ia s growth method in which the constituent elements in the form of molecular beams, are introduced into the ultra-high vacuum inside the growth chamber [4] by direct evaporation from Knudsen effusion cells [5]. These molecular beams impinge on the heated substrate where surface kinetic reactions lead to crystallization of the grown layer. MBE is a highly controlled, very flexible growth method. It enables a precise control of doping and composition profiles of the grown layers. This crystallization technique is, however, still in a very exploratory development phase.

In principle it is possible to grow optical structures with sufficiently good quality, using each of the mentioned epitaxy methods. The crystal grower, however, who often has to crystallize a definite specially designed optical monolithically integrated device needs a simple criterion according to which he would be able to make a choice of the crystallization method most suitable for fabrication of this device. This is just the reason for which the three technological methods will be compared in the present work, from the point of view of their crystalization peculiarities connected with the preparation of optical monolithically integrated devices in general. The considerations will be limited to $GaAs-Al_xGa_{1-x}As$, because only for this materials system the experimental data enabling such comparison are available at present.

## 2. Comparison criterion

Let us consider the integrated devices coupled at the transmitter and receiver sides to a glass fibre optical communication link, schematically shown in Fig. 2. These devices play the role of a frequency-multiplexing light source and a frequency-demultiplexing light detector, respectively.

The first integrated device [6] consists of four distributed-feedback (DFB) heterojunction lasers [7, 8] emitting coherent electromagnetic radiation at slightly different frequencies. These lasers are coupled through four low-loss passive waveguides to the output waveguide, to which one end of the glass fibre of the communication link is connected.

The second integrated device, connected to the exit of the opposite end of the glass fibre, consists of a DH optical waveguide with a chirped-grating (grating with a variable period) [9], and of four detecting *p-n* diodes, crystallized above the waveguide at places, where the corrugation deflects different wavelengths of the transmitted electromagnetic radiation.

We can see that the appearance of many nonplanar interfaces is characteristic of these integrated circuits. The nonplanar interfaces are, however, covered by epitaxial layers which at the counter side are bounded by planar surfaces.

This means that one of the most important goals of the fabrication process of such integrated devices is to achieve uniform coverage, regardless of the substrate shape. This fact makes it possible to formulate a comparison criterion for the crystallization methods as applied to the fabrication of optical monolithically integrated devices.

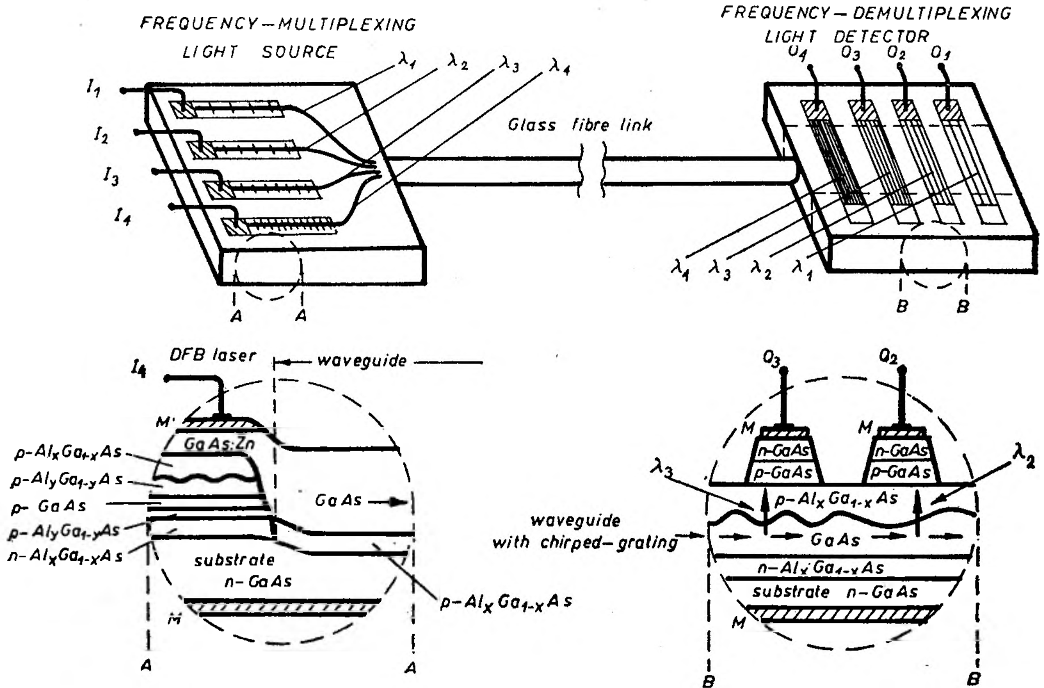



Fig. 2. Schematic drawing of a glass fibre optical communication link with two optical monolithically integrated devices, as frequency-multiplexing light source and as frequency-demultiplexing light detector. The cross-sections $A$–$A$ and $B$–$B$ of these devices show a lot of nonplanar interfaces between the epitaxial layers of these structures

According to this somewhat arbitrary, but relatively simple criterion "the best crystallization method is this one, which gives the complete smoothing of the layer covering the nonplanar interface for the smallest thickness of this layer".

This criterion suggests to use, for comparison of the crystallization methods, the morphological stability theory of MULLINS and SEKERKA [10–13] and its modification of VAN DEN BREKEL and JANSEN [14–16]. This theory deals with the stability or instability of the shape of the interface which separates a crystal from a second phase from which it is crystallizing. This second phase may be a solid, a liquid or a gas, depending upon the crystal growth process under consideration.

## 3. Morphological stability theory in epitaxy

Let us consider a planar interface separating the substrate crystal from the liquid or gaseous phase, which will be called the crystallizing phase. Suppose this interface is randomly perturbed by a collection of nonplanar optical elements of an integrated circuit, previously superimposed on it by etching or growing (Fig. 3).

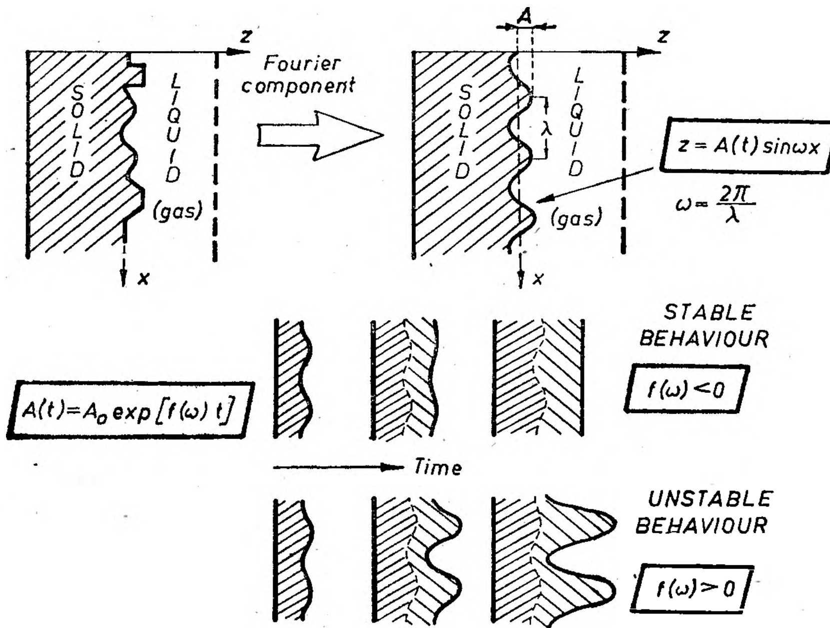



**Fig. 3. Sketch of a randomly perturbed shape of a planar solid-liquid (gas) interface, its sinusoidal Fourier component and examples of stable and unstable behaviours of this interface shape during the crystallization of a covering epitaxial layer**

Since a small arbitrary perturbation in the plane may be analysed by developing it into sinusoidal Fourier components, the development of the perturbation in time during the crystallization of a covering epilayer is simply a superposition of the developments of its Fourier components. We can assume, therefore, that the interface is stable if none of the sinusoidal Fourier components grows. If, however, any sinusoidal wave grows, i.e., its amplitude increases in time, then the interface is unstable. So, it is sufficient to assume that the interface has the form of a sinusoidal wave

$$z = A(t)\sin\omega x \tag{1}$$

in a co-ordinate system with the $z$ axis pointing toward the crystallizing phase, and the plane $z = 0$ coinciding with the planar interface (Fig. 3). The paramter $\omega$ is the spatial frequency of the perturbation, and $A(t)$ is the amplitude of

this perturbation. It is assumed that $A(t)$ is small in comparison to the wavelength $\lambda = 2\pi/\omega$.

We wish to obtain an expression for the time derivative $dA/dt$ in order to see whether the periodic perturbation grows or decays for specified crystallization conditions. To this end the growth velocity $v$ of each element of the interface should be calculated as a function of its position $x$ in terms of the suitable driving force of the crystallization process considered. The calculations differ in details for each crystallization method we are interested in, the results, however, have the following common form

$$A(t) = A_0 \exp[f(\omega)t] \tag{2}$$

where $A_0$ is the value of $A(t)$ at the beginning of the growth process (at $t = 0$).

The function $f(\omega)$ appearing in the exponent is called the stability function. The growth or decay of the amplitude of the interface perturbation depends on the sign of this function. If $f(\omega) > 0$, the amplitude $A(t)$ will grow in time and the interface will be unstable, while if $f(\omega) < 0$, $A(t)$ will decay and the interface will be stable.

It is now clear that our comparison criterion concerning the epitaxy methods mentioned above may be expressed in terms of the stability function. Now, it says that "the best crystallization method is the one, for which the stability function being negative has the largest absolute value for a given wavelength of the sinusoidal perturbation of the planar interface". It means, that for the best epitaxy method

$$f(\omega) < 0, \text{ and } |f(\omega)| = \max. \tag{3}$$

The principal physical assumptions, on which the growth rate calculations in the morphological stability theory of Mullins and Sekerka are based, are those of isotropy of bulk and surface parameters at the interface. The local thermodynamic equilibrium at all points of the interface is additionally assumed. These assumptions, however, make questionable the applicability of this theory to epitaxy. Since for the latter a strong dependence of the growth rate on crystallographic orientation of the substrate has been evidenced experimentally in many cases [17]. Thus, the growth rate becomes an anisotropic quantity. Moreover, epitaxy is a dynamic rather than an equilibrium process. In this process the kinetics of both, the mass transport and the surface process play an important role.

All these facts cause that the values of stability functions calculated for different epitaxial growth methods, according to the Mullins and Sekerka theory, should be treated only as the first approximation to the reality. Nevertheless, we believe that even a qualitative information concerning the crystallization methods may be useful for a crystal grower who must choose the most suitable growth method for fabrication of a definite optical monolithically integrated device.

It seems that for this simple purpose very important for the practice the difficult methematical formalism characteristic of an anisotropic morphological stability theory [18, 19] is useless. So, in our comparison procedure we shall deal with the relatively simple formalism of Mullins and Sekerka theory.

## 4. Stability functions for LPE, MO CVD and MBE

Stability functions depend on a number of deposition parameters, different in each of the analysed epitaxy methods. In order to make them presise first the growth velocities in each epitaxy process should be calculated.

In the LPE slider technique with confined solution vloume (Fig. 4) which has become currently the technique with the best control of thickness, morpho-

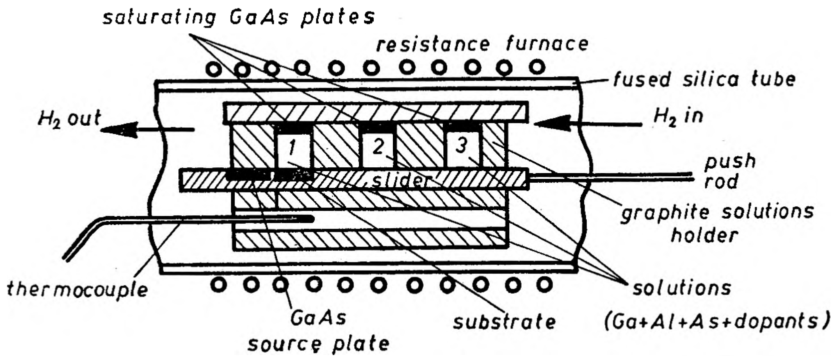



Fig. 4. The central part of a fused silica tube LPE reactor. The cross-section of the graphite crystallization crucible is shown. The Ga solutions 1, 2, 3 are confined to very small volumes by the substrate plate or slider rod from the bottom side and by the saturating GaAs plates from the top side

logy and uniformity [20], the growth process in entirely controlled by the diffusion of nutrients through the solution toward the substrate. To this case the analysis of SHEWMON [21] can be adapted.

In the considered LPE process each element of a planar solid-solution interface grows with a velocity $v$ given by the equation

$$v = \frac{D}{c_s - c_p} \left( \frac{\partial c_p}{\partial z} \right) \bigg|_{z=0} \tag{4}$$

where $c_s$ and $c_p$ are the concentrations of As atoms in the epitaxial layer of GaAs (or $Al_xGa_{1-x}As$) and in the Ga (or Ga+Al) solution at the planar interface, respectively; $D$ is here the diffusion coefficient of As in liquid gallium (or liquid Ga+As mixture). Let a perturbation of the form (1) be now superimposed on the moving interface. This causes the appearance of surface curvatures in many places of the interface and subsequently, in the case of equilibrium across the interface, gives rise to variation of the As concentration in the gallium solution

at the interface. The changed values of this concentration can be calculated by using the Gibbs-Thomson equation [22]. For small values of $A(t)$ the local curvature of the surface equals $d^2z/dx^2$, so that the concentration of As atoms at the nonplanar interface is

$$c_{\mathrm{np}} = c_L\left(1+\Gamma A(t)\omega^2 \sin\omega x\right) = f(x, t). \tag{5}$$

Here $c_L$ is the equilibrium solubility of As in gallium solution, and $\Gamma = V_0\gamma/RT$ is the capillarity constant, where $V_0$ is the molar volume of the solid phase, $\gamma$ is the surface tension of the deposited material, $R$ is the gas constant and $T$ the absolute temperature at the interface.

For LPE the variation of the solute concentration on the solution stemming from the variation of the interface shape can be in the other hand approximated by solving the Laplace's equation $\nabla^2 c_{\mathrm{np}} = 0$ for the given shape change and the boundary condition $c_{\mathrm{np}} = f(x, t)|_{z=0}$.

The general solution to Laplace's equation for a sinusoidal interface and a given gradient $G$, of the As atoms concentration in the Ga solution, at the interface is

$$c_{\mathrm{np}}(x, z) = I_0 + B_0 \exp(-\omega z)\ A(t)\sin\omega x + Gz. \tag{6}$$

Now, we should like to choose the constants $I_0$ and $B_0$ so that this equation be reduced to Eq. (5) along the surface given by Eq. (1). This is accomplished to first order in $A(t)$ if $I_0 = c_L$ and $B_0 = c_L\Gamma\omega^2 - G$ or if

$$c_{\mathrm{np}}(x, z) = c_L + (c_L\Gamma\omega^2 - G)\ A(t)\sin\omega x\ \exp(-\omega z) + \mathrm{G}z. \tag{7}$$

The linear growth rate is then given by Eq. (4) as

$$v_{\mathrm{np}} = v_p + \frac{dA}{dt}\sin\omega x = \frac{D}{c_e - c_p}\left[G + (G\omega - c_L\Gamma\omega^3)A(t)\sin\omega x\right]. \tag{8}$$

The first term in Eq. (8) represents the growth rate in the absence of any interface shape perturbation. The development of a shape perturbation of the interface relative to the mean position of the latter is given by the second terms as

$$\frac{dA}{dt} = \frac{D}{c_s - c_p}(G - c_L\Gamma\omega^2)\omega A(t). \tag{9}$$

From Equations (2) and (9) the following form of the stability function for LPE is evident

$$f(\omega)_{\mathrm{LPE}} = \frac{D}{c_s - c_p}(G - c_L\Gamma\omega^2). \tag{10}$$

The first term of the right-hand side of this equation may be interpreted physically as being due to an increase in the concentration gradient in front of the *hills* on the perturbed interface. The second term is due to the concentration

gradients along the surface which cause solute transport and so tend to smooth out the sinusoidal distrurbance.

In MO CVD growth technique in which TMG [$Ga(CH_3)_3$], TMA [$Al(CH_3)_3$] and $AsH_3$ are used as Ga, Al and As sources, respectively (Fig. 5), the ratio of

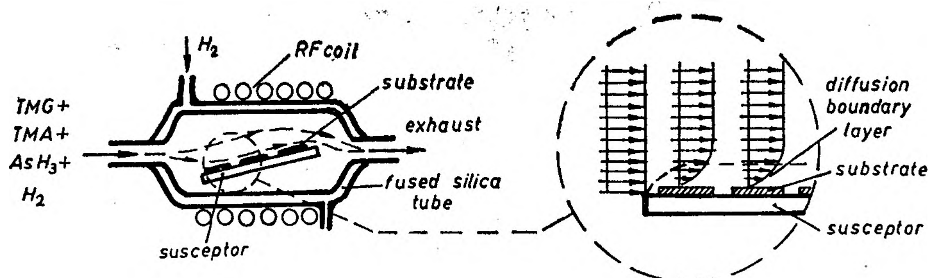



Fig. 5. Schematic drawing of the fused silica tube crystallization reactor for MO CVD process. The cross-section shows the development of diffusion boundary layer over the substrates, holder plate indicating laminar and turbulent gas flow regimes [17]

As to Ga (or Al+Ga) in the input gas stream is always larger than one for the most favourable growth conditions [17, 23]. The growth rate of the epitiaxial layer is therefore found to be linearly proportional to the metalorganic component flow rate.

The temperature dependence and the magnitude of growth rate are typical of the behaviour expected for mass transport limited growth kinetics. It has also been evidenced [24] that the organometallic molecules may be decomposed in the diffusion boundary layer near the substrate, which causes that the actual species diffusing to the interface may be the GaAs (or $Al_xGa_{1-x}As$) molecules.

These facts justify the adaptation of the van den Brekel and Jansen analysis of morphological stability the isothermal growth process of polycrystalline Si CVD [14–16] to the case of GaAs (or $Al_xGa_{1-x}As$). This gives the following formula for the stability function in our case of the MO CVD

$$f(\omega)_{\mathrm{MO\,CVD}} = \frac{Mk_D}{\varrho N}\left[\frac{G-\Gamma\omega^2}{1+\mathrm{Nu}(\tan h\omega\delta/\omega\delta)}\right] \tag{11}$$

where $M$ and $\varrho$ are the molecular weight and the specific density of the deposited layer, respectively, $N$ — the Avogadro number, $G$ — the concentration gradient in the diffusion boundary layer of thickness $\delta$, $\mathrm{Nu} = (k_D\delta)/D$ is the Nusselt number, $D$ — the diffusion coefficient of the reactive compound, and $k_D$ — the mass transfer coefficient for the epitaxial growth reaction. The capillarity constant is here equal to $(\gamma c_G V_m)/kT$, where $\gamma$ is the surface tension of the deposited material, $c_G$ — the equilibrium concentration of the reactive compound at the planar interface, $V_m$ — the molar volume of the solid, and $k$ — the Boltzmann constant.

The MBE growth process is dominated by mass transport defined by the evaporation rate from the effusion sources, generating molecular beams in the

ultra-high vacuum of the growth chamber (Fig. 6). The surface processes, like adsorption, surface diffusion, and incorporation into the growing epitaxial layer play, however, a crucial role in this growing process.

The growth rate or the velocity of the growing solid-gas interface of MBE GaAs (or $Al_xGa_{1-x}As$) films is entirely controlled by the flux density of the Ga

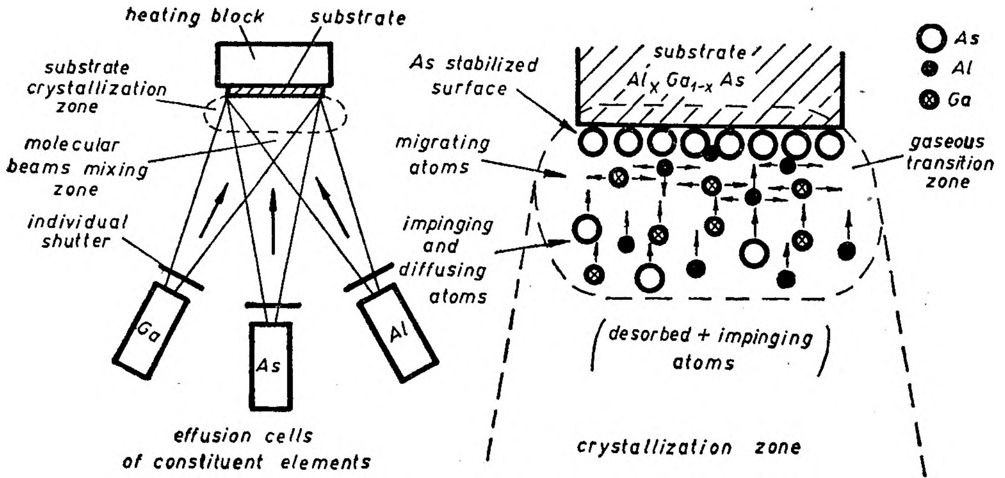



Fig. 6. Schematic illustration of the basic elements of a simple MBE apparatus (left side), and of the main physical processes appearing during the MBE process in the immediate vicinity of the substrate surface (right side)

(or Ga and Al) beam impinging on the substrate surface, because the group III elements have sticking coefficients to GaAs substrates with values close to unity, at typical MBE growth temperature (450°C–620°C) under As-stabilized growth conditions. This implies that nearly all Ga (or Ga and Al) atoms incident on the GaAs substrate surface get incorporated into the growing epitaxial layer, despite the fact that after being adsorbed they still undergo numerous migration and rearrangement [4].

The problem of morphological stability in MBE has not been analysed yet at all. So, no numerical and analytical data for this crystallization method can be found in the literature. In order to make possible the estimation of the stability function for this case, the following assumptions should be made:

— At the solid-gas interface there exists a thin gaseous transition layer extending into the high vacuum environment up to few atomic layers of the crystallizing material. In this layer the atoms impinging on the substrate, migrating over its surface and desorbing from the substrate create a gas mixture with a quasi-equilibrium concentration of group III atoms equal to $c_p$.

— The group III atoms diffuse across the transition layer toward the interface with a diffusion constant $D$, giving rise to the growth rate similar to the atoms of As in the LPE growth process.

— The Shewmon's analysis can be applied to the MBE case, after some modification.

— An additional term reflecting the stabilizing effect of the surface migration [12] of group III atoms should be added to the stability function for MBE, with a negative sign and with a $D_s$ surface diffusion coefficient.

On the basis of these assumptions the following form can be assigned to the stability function for MBE

$$f(\omega)_{\mathrm{MBE}} = \frac{D\omega}{c_p}\left(G - c_p\Gamma\omega^2 - c_p\frac{D_s}{D}\Gamma\omega^2\right). \tag{12}$$

We can now compare the three epitaxy methods by evaluating their stability functions expressed by the equations (10), (11) and (12). Using the values given in Table for the respective physical quantities the results shown in Fig. 7 have been obtained for spatial frequencies $\omega$ corresponding to corrugation periods of GaAs $-\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ DFB lasers.

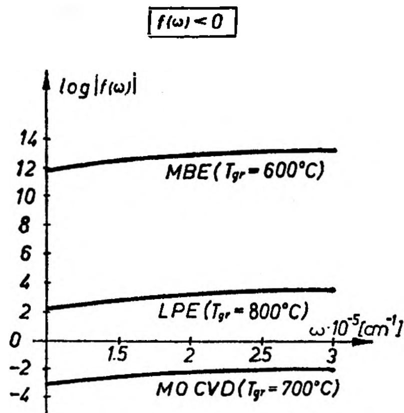



Fig. 7. Stability functions of the three considered epitaxy methods, evaluated by using the values of the physical quantities given in Table

| Quantity | LPE | MO CVD | MBE |
|---|---|---|---|
| $D\,[\mathrm{cm^2 \cdot s^{-1}}]$ | $4\cdot10^{-5}$ | 1.76 | $10^4$ |
| $D_s\,[\mathrm{cm^2 \cdot s^{-1}}]$ | | | $10^3$ |
| $\Gamma\,[\mathrm{cm}]$ | $7.8\cdot10^{-8}$ | | $7.8\cdot10^{-8}$ |
| $\Gamma\,[\mathrm{cm^{-2}}]$ | | $2.23\cdot10^8$ | |
| $C_s$ (at %) | 50 | | |
| $C_p$ (at %) | 2.4 | | 0.1 |
| $C_L$ (at %) | 2.5 | | |
| $G\,[\mathrm{cm^{-1}}]$ | 1 | | $10^{-2}$ |
| $G\,[\mathrm{cm^{-4}}]$ | | $2.1\cdot10^{15}$ | |
| $k_D\,[\mathrm{cm \cdot s^{-1}}]$ | | 11.73 | |
| $\delta\,[\mathrm{cm}]$ | | 0.3 | |
| Nu | | 2 | |
| $M\,[\mathrm{g}]$ | | 145 | |
| $\varrho\,[\mathrm{g \cdot cm^{-3}}]$ | | 5.316 | |

We see that all crystallization methods being considered are characterized by negative stability functions. It means that they are stable. The absolute values of the stability function is, however, the largest for MBE process, which means that just this growth process is the most stable and thus the most suitable for preparation of monolithically integrated optical devices.

## 5. Conclusions

The estimated stability functions for LPE, MO CVD and MBE can be used for comparison of these three epitaxy growth techniques, from the point of view of their application to the fabrication process of monolithically integrated optical

devices. This comparison is relatively simple, it refers, however, solely to the phenomenological crystallization aspects and neglects such important factors like structural perfection or electrical and optical parameters of the grown layers. So, it gives useful information for the crystal grower but says little to the physicist or engineer designing the integrated devices.

The values of the respective physical quantites given in Table can be used to evaluate the stability functions for the considered epitaxy methods. From the plots of the stability functions shown in Fig. 7 it can be concluded that the most stable growth technique, i.e., the most suitable for optical monolithic integration is the MBE technique and that the less stable (the less suitable) is the MO CVD technique. This conclusion is in agreement with the observed experimental facts, that the layers covering the corrugations of DFB lasers optical resonator are always grown by MBE [25] or by LPE [26]. So far, no information concerning a successful crystallization of such covering layers by using tne MO CVD technique have been published.

A more exact analysis of the morphological stability in epitaxy requires complicated theoretical considerations and intensive experimental investigations of this problem, taking into account the anisotropic features of growth velocity of the epitaxial layers.

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**Морфологическая устойчивость кристаллов монолитно интегрированных оптических устройств**

Сравнены три технологических метода: LPE, MO CVD и MBE с точки зрения их кристаллизационных особенностей, связанных с изготовлением монолитно интегрированных оптических устройств. Для каждого процесса, проходящего в кристаллах системы GaAs$-$Al$_x$Ga$_{1-x}$As вычислены функции морфологической устойчивости Муллинса и Секерки, причем не была учтена зависимость темпа роста кристалла от ориентации подложки. Доказано, что для монолитного оптического соединения наиболее подходящим методом кристаллизации является метод MBE, наименее же подходящим - метод MO CVD.

*Перевела Малгожата Хейдрих*