Optik 124 (2013) 3571–3574

Contents lists available at SciVerse ScienceDirect

Optik

journal homepage: www.elsevier.de/ijleo

Preproduction prediction of the optical normalized reflectance transient of MOVPE-grown AlGaAs films using transfer matrix method

Jianwei Zhang[a,b], Yongqiang Ning[a,*]

[a] State Key Laboratory of Luminescence and Application, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, 3888 Southeast Lake Road, Changchun 130033, China
[b] Graduate University of the Chinese Academy of Sciences, Beijing 100049, China

ARTICLE INFO

Article history:
Received 18 June 2012
Accepted 1 November 2012

Keywords:
Normalized reflectance
In situ monitoring
Transfer matrix method
Metal–organic vapour phase epitaxy
Distributed Bragg Reflectors
Vertical cavity surface emitting lasers

ABSTRACT

This paper reported on the prediction and analysis of the optical normalized reflectance (NR) transient of AlGaAs multi layers and Distributed Bragg Reflector (DBR) using transfer matrix method (TMM). The simulation result correlated well with the measured NR transient of grown samples. Deviations of $Al_xGa_{1-x}As$ composition between the expected and real grown sample were predicted successfully. Smaller optical oscillation amplitude in the first DBR layer was predicted and could be used as an indication of Al composition. Special characteristics of NR transient within the first three pairs of DBR were also clarified using the calculated transmissivity changing with the growth thickness. TMM simulation of NR transient was thus shown to be a convenient and reliable pre-production technique, also not restricted to the AlGaAs material.

1. Introduction

With the development of metal organic vapor phase epitaxy (MOVPE), many complex multilayer devices such as VCSELs had been grown successfully [1,2]. Non-destructive in situ monitoring of normalized reflectance [3] and reflectance anisotropy spectroscopy (RAS) [4] was incorporated into the MOVPE to improve the growth technique. Many such reports appeared recently aiming at the in situ monitoring of growth rate, temperature and composition of epitaxial layer based on the reflection spectroscopy [5–8]. Parameters of grown layers gained from the NR transient were even as accurate as ex situ measurements such as SEM and TEM [9]. Thus it would be reliable if used as the pre-production technique for evaluating the information of growing layer. However, such applications had not been reported before. To apply the NR transient as the pre-production technique, simulation of the time resolved NR data of expected structure was needed to be carried out first before the costly fabrication process.

In this work, the numerical calculation of the NR transient of multilayer structures was carried out based on the transfer matrix method (TMM). Comparison between the simulated and monitored reflectance data proved that it was efficient in assessing the growth equality and calibrating the parameters during the growth process. The physical origin of some special peculiarities in the NR transient of DBR was also clarified.

2. Modeling and experimental

2.1. Modeling

Detailed description about TMM had been presented in Refs. [10,11]. The main principle of TMM was shown below.

As shown in Fig. 1, parameters n and d were the real part of the refractive index and thickness of each layer, and the subscript character indicated different layers. Signals of a, b, c, d... indicated the interfaces between these layers. Normal incidence of linearly polarized plane electromagnetic wave on the structure was assumed.

When the electromagnetic wave transmitted through the structure consisting of m layers, then the following transfer matrix could be obtained:

$$\begin{bmatrix} B \\ C \end{bmatrix} = \prod_{i=1}^{m} \begin{bmatrix} \cos\delta_i & i\sin\delta_i/n_i \\ in_i\delta_i & \cos\delta_i \end{bmatrix} \cdot \begin{bmatrix} 1 \\ n_{sub} \end{bmatrix} \quad (1)$$

Refractive index and phase change of the ith layer were expressed by n_i and δ_i. We defined $Y = C/B$ from Eq. (1). Reflectance R and transmissivity T were then calculated:

$$R = \left(\frac{n_0 - Y}{n_0 + Y}\right) \cdot \left(\frac{n_0 - Y}{n_0 + Y}\right)^* \quad (2)$$

* Corresponding author. Tel.: +86 431 86176348; fax: +86 431 86176348.
E-mail address: zcjw1985@yahoo.cn (Y. Ning).

0030-4026/$ – see front matter © 2012 Elsevier GmbH. All rights reserved.
http://dx.doi.org/10.1016/j.ijleo.2012.11.040

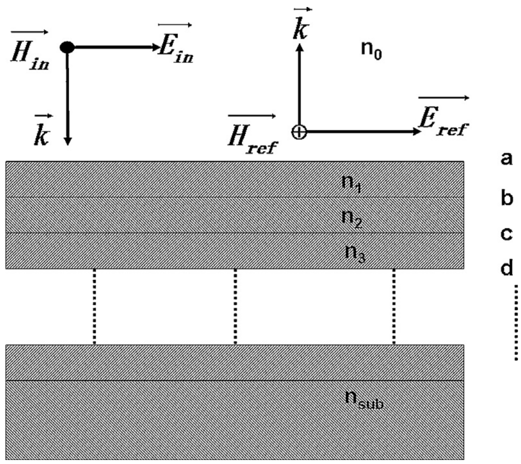

Fig. 1. Normal incidence of plane electromagnetic wave on a multilayer structure consisting of layers on an infinite thick substrate.

$$T = \frac{4 - n_0 n_{sub}}{(n_0 B + C) - (n_0 B + C)^*} \tag{3}$$

To carry out the TMM simulation of the growth process, each layer was divided into many homogeneous sub-layers, then the reflectance of grown structure after increasing by a sub-layer could represent the reflectance at corresponding growth time in the time resolved NR data. The thickness and refractive index of these layers were utilized in the simulation. Also the transmissivity of grown layers at different growth time could be obtained following this method. And it could be used for explaining the oscillation characteristics of NR transient.

2.2. Experimental

Epilayers were grown on undoped (0 0 1) GaAs substrate in a 3 × 2 planetary MOVPE reactor. The source materials included trimethylgallium (TMGa), trimethylaluminium (TMAl), and arsine (AsH_3). A pressure of 100 mbar and a growth temperature of 970 K were used to realize mirror-like surface quality. In situ measurements were performed with LayTec EpiRAS 200 spectrometer mounted above the viewport of reactor. To reduce the sensitivity of NR to the surface roughness, the monitoring photon energy $E < 3$ eV for the optical measurement was chosen.

Two samples were grown, with their SEM images shown in Fig. 2. The expected grown layers of sample 1 were GaAs buffer, $Al_{0.25}Ga_{0.75}As$, GaAs, $Al_{0.4}Ga_{0.6}As$, GaAs in turn. Sample 2 was the Distributed Bragg Reflector (DBR) mirrors, consisting of 23 pairs of $Al_{0.12}Ga_{0.88}As/Al_{0.9}Ga_{0.1}As$ layers. The designed central wavelength of DBR structure was 980 nm.

The normalized reflectance is the reflectance of grown layers divided by the reflectance of a reference sample. The reflectance of GaAs substrate R_{GaAs} was measured prior to each growth process and used thereafter as reference R_{ref}, then the NR can be defined as:

$$\mathrm{NR} = \frac{R}{R_{ref}} = \frac{R}{R_{GaAs}} \tag{8}$$

3. Results and discussion

3.1. Simulated and monitored NR transient of sample 1 at the monitoring photon energy of 2.3 eV (539.1 nm)

In Fig. 3, the simulated and measured NR transients were shown with the refractive index profile shown to identify each layer.

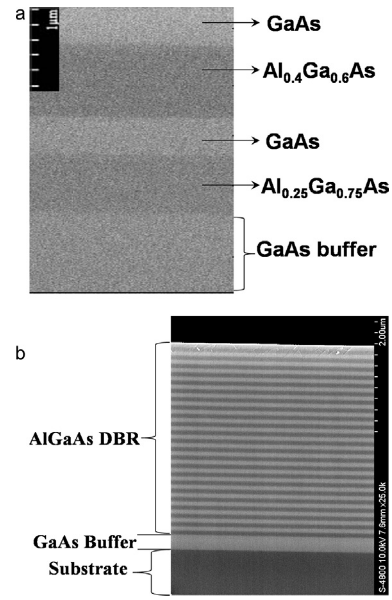

Fig. 2. SEM images of grown samples: (a) sample 1 and (b) sample 2.

FP oscillations of NR transient were observed due to the optical interference. The amplitude of FP oscillation was damped for each layer due to the optical absorption. The converged value of this FP oscillation was determined only by the optical constants (refractive index n and extinction coefficient k) and could be used for distinguishing the Al composition of each layer [12].

The relationship between the compositions and converged values of simulated NR were gained, as shown in Fig. 4. Also the compositions and converged values of real grown layers were shown. The composition of real grown sample was calibrated by XRD. The Al compositions for each layer were 0, 0.255, 0, 0.352, 0, while the expected one were 0, 0.25, 0, 0.4, 0. We could see from Fig. 4 that the simulated result was accurate in predicting the composition deviation. Thus it would be useful in predicting the compositions of grown layers during the growth process. In fact, the growth parameters such as the source flow etc. could be changed in real time when the average value of NR was found to be deviant during the above process.

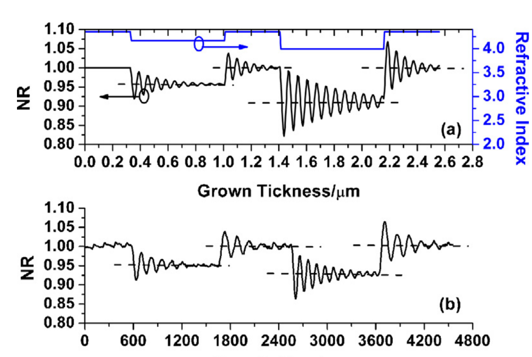

Fig. 3. (a) Simulated NR transient of sample 1, with the refractive index shown to identify different layers, corresponding layers was $GaAs/Al_{0.25}Ga_{0.75}As/GaAs/Al_{0.4}Ga_{0.6}As/GaAs$ and (b) monitored NR transient during the growth process of sample 1.

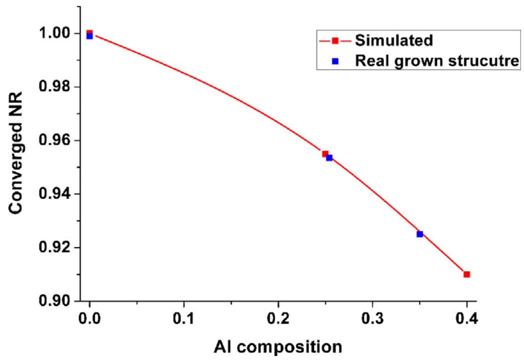

Fig. 4. Al compositions and their corresponding converged NR of grown layers, red line was the fitted relation of converged NR and Al composition from the simulation; while the blue triangle represents the results from real monitoring. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

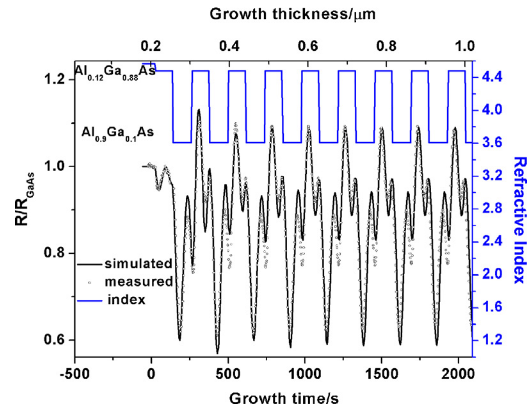

Fig. 6. Details of time-resolved normalized reflectance simulation and measurement of DBR together with a schematic refractive index profile. The line shows the simulation, the dots represent the measured data.

3.2. *Simulation and in situ monitoring of DBR structures with a central wavelength of 980 nm at monitoring photon energy of 2.6 eV (477 nm)*

Distributed Bragg Reflector (DBR) was widely used in the optoelectronic devices such as vertical cavity surface emitting lasers (VCSELs) [13], resonant cavity light emitting diodes (RCLEDs) [14], etc. A good control of the growth process of DBR would be helpful to improve the device performance.

In Fig. 5, the measured reflectance transient during the growth of sample 2 was shown alongside a schematic cross section. Due to the limited penetration depth of incidence light and the periodic layer structure, periodicity of NR transient for each pair of layers was expected, as was shown in Fig. 5(b). Also the signal level of the whole transient slightly decreased from layer pair to layer pair due to the optical absorption.

Fig. 6 gives detail from the simulated and monitored NR transient of DBR structure, with the refractive index shown to identify each layer. The NR transient started with an increase within the layer whose refractive index was higher than the layer below, vice versa. The optical thicknesses of alternating layers were 324 nm and 284 nm at the growth temperature of 970 K. Thus only one maximum and one minimum appeared in the NR transient of each layer for DBR structure at the incident optical wavelength of 477 nm.

The amplitude of FP oscillation in simulation was larger than measured one in Fig. 6. Lower amplitude of FP oscillation indicated weaker optical interference, which was originated from the optical loss in the grown layers. Optical loss might be caused by the roughness of growing surface. This indicated that the monitoring photon energy of 2.6 eV still could not fully eliminate the influence of interface roughness.

The reflected optical field at the GaAs and $Al_{0.12}Ga_{0.88}A$ interface was very weak due to their low refractive index contrast in Fig. 6. Also the optical absorption in these two layers was very large because of their narrow energy band. Thus very slight oscillation of NR transient was expected in the first $Al_{0.12}Ga_{0.88}As$ layer, as shown in Fig. 6. This slight oscillation could be used for identifying the Al composition of the first grown layer. If the $Al_{0.9}Ga_{0.1}As$ layer was grown first, the amplitude would be much bigger because of larger refractive index contrast between the $Al_{0.9}Ga_{0.1}As$ and GaAs.

Exact periodicity of the reflectance transient started from the fourth pair of layers, as was shown in Fig. 6. To explain this peculiarity, the optical transmissivity relative to the NR transient of grown structure was calculated, as shown in Fig. 7.

Due to the optical absorption, the transmissivity of grown structure decreased rapidly with the grown thickness. The transmissivity decreased to <0.1 after the growth of the first three pairs of DBR near the substrate, thus the optical penetration depth could be estimated as three pairs of alternating layers in the DBR. The NR transient of the first three pairs of DBR could be influenced by the optical field reflected from the rough interface between the buffer layer and the first $Al_{0.12}Ga_{0.88}As$ layer. But the NR spectrum of other layers could not be influenced by this interface due to the limited penetration depth. So the NR spectrum in the first three pairs of DBR near the substrate was different from other layers. Thus the appeared characteristics of NR transient within DBR were clarified.

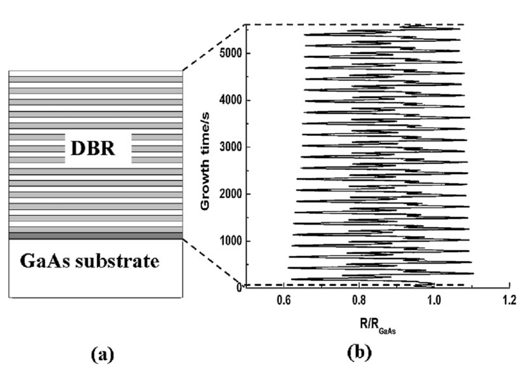

Fig. 5. (a) Schematic cross section of DBR structure and (b) corresponding NR transient at $E = 2.6$ eV (447 nm) measured during growth.

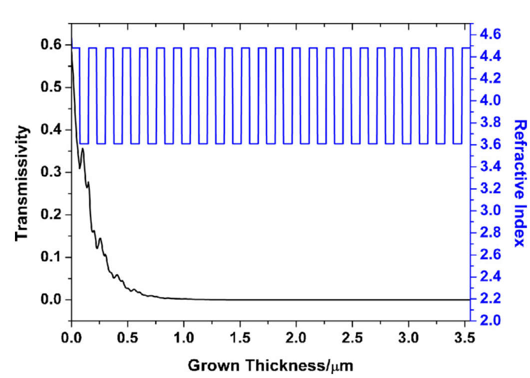

Fig. 7. Calculated transmissivity of grown structure during the DBR growth process, with the refractive index profile of grown DBR structure.

4. Conclusions

For predicting and calibrating the growth process, a TMM simulation of the normalized reflectance transient was carried out. The NR transient calculated from the TMM is in good agreement with the measured one. Valuable information about the growth process of AlGaAs layers can be extracted from the simulated NR transient. Calibrating the growth process using simulated NR was achieved. Such a simulation is of course a convenient and precise preproduction technique, offering additional advantages over the conventional after grown analysis.

In addition, this method could be realized for any sample structures, with only the sample structure and refractive index as the key parameter. Our proposed method could also be widely applied to simulate the in situ monitoring process for any other materials. New insights regarding the simulation and analysis of growth process for optoelectronic devices are expected soon.

Acknowledgment

This work is sponsored by the National Natural Science Foundation of the People's Republic of China (60876036, 10974012, 11074247, 60876036, 61106047), and the authors would like to appreciate sincerely for this support.

References

[1] P.V. Bulaev, V.A. Kapitonov, A.V. LutetskiĬ, MOCVD-grown InGaAs/GaAs/AlGaAs laser structures with a broad-area contact, Semiconductors 36 (2002) 1065.
[2] M. Zorn, A. Knigge, U. Zeimer, MOVPE growth of visible vertical-cavity surface-emitting lasers (VCSELs), J. Cryst. Growth 248 (2003) 186.
[3] Y. Raffle, R. Kuszelewicz, R. Azoulay, G. Le Roux, J.C. Michel, L. Dugrand, E. Toussaere France, In situ metalorganic vapor phase epitaxy control of GaAs/AlAs Bragg reflectors by laser reflectometry at 514 nm, Appl. Phys. Lett. 63 (1994) 3479.
[4] S.J. Morris, J.Th. Zettler, K.C. Rose, D.I. Westwood, D.A. Woolf, R.H. Williams, W. Richter, In situ reflectance anisotropy studies of AlGaAs layers grown on GaAs (0 0 1) by molecular beam epitaxy, J. Appl. Phys. 77 (1995) 3115.
[5] H. Kawai, S. Imanaga, K. Kaneko, N. Watanabe, Complex refractive indices of AlGaAs at high temperatures measured by in situ reflectometry during growth by metalorganic chemical vapor deposition, J. Appl. Phys. 61 (1987) 328.
[6] T. Trepk, M. Zorn, J.T. Zettler, M. Klein, W. Richter, Spectroscopic ellipsometry applied for in-situ control of lattice matched III–V growth in MOVPE, Thin Solid Films 313–314 (1998) 496.
[7] M. Zorn, T. Trepk, P. Kurpas, M. Weyers, J.T. Zsttler, W. Richter, In situ monitoring and control of InGaP growth on GaAs in MOVPE, J. Cryst. Growth 195 (1998) 223.
[8] L.E. Tarof, C.J. Miner, C. Blaauw, InGaAsP compositional determination from reflectance spectroscopy of InGaAsP/InP heterostructures, J. Appl. Phys. 68 (1990) 2939.
[9] M. Zorn, K. Haberland, A. Knigge, A. Bhattacharya, M. Weyers, J.-T. Zettler, W. Richter, MOVPE process development for 650 nm VCSELS using optical in-situ techniques, J. Cryst. Growth 235 (2002) 25.
[10] M. Born, E. Wolf, Principles of Optics, Pergamon Press, Oxford, 1975.
[11] H.A. Macleod, Thin-Film Optical Filters, Institute of Physics Publishing, Bristol, 2001.
[12] C. Watatani, Y. Hanamaki, M. Takemi, K. Ono, Y. Mihashi, T. Nishimura, In situ monitoring of growth rate and composition of AlGaInP and InGaAsP by reflection measurements in MOVPE, J. Cryst. Growth 281 (2005) 227.
[13] K. Iga, Surface-emitting laser-its birth and generation of new optoelectronics field, IEEE J. Sel. Top. Quant. 6 (2000) 1201.
[14] E.F. Schubert, Y.H. Wang, A.Y. Cho, L.W. Tu, G.J. Zydzik, Resonant cavity light-emitting diode, Appl. Phys. Lett. 60 (1992) 921.