Spectral transmittance and module structure fitting for transmission-mode GaAs photocathodes*

Zhao Jing(赵 静), Chang Ben-Kang(常本康)†, Xiong Ya-Juan(熊雅娟) and Zhang Yi-Jun(张益军)

Institute of Electronic Engineering and Optoelectronic Technology, Nanjing University of Science and Technology, Nanjing 210094, *China*

(Received 21 November 2010; revised manuscript received 15 December 2010)

A transmission-mode GaAs photocathode includes four layers of glass, Si_3N_4, $Ga_{1-x}Al_xAs$ and GaAs. A gradient-doping photocathode sample was obtained by molecular beam epitaxy and its transmittance was measured by spectrophotometer from 600 nm to 1100 nm. The theoretical transmittance is derived and simulated based on the matrix formula for thin film optics. The simulation results indicate the influence of the transition layers and the three thin-film layers except glass on the transmittance spectra. In addition, a fitting coefficient needed for error modification enters into the fitted formula. The fitting results show that the relative error in the full spectrum reduces from 19.51% to 4.35% after the formula is modified. The coefficient and the thicknesses are gained corresponding to the minimum relative error, meanwhile each layer and total thin-film thickness deviation in the module can be controlled within 7%. The presence of glass layer roughness, layer interface effects and surface oxides is interpreted on the modification.

Keywords: GaAs photocathode, transmission-mode, optical properties, matrix formula

PACS: 78.66.Fd, 78.20.Ci, 78.40.Fy **DOI:** 10.1088/1674-1056/20/4/047801

1. Introduction

A negative electron affinity (NEA) GaAs photocathode possesses high quantum efficiency, small dark emission and flat spectrum curve. Transmission-mode (t-mode) GaAs photocathodes are currently attracting extensive attention and have been applied in many areas such as for high performance low light level (LLL) image intensifiers, semiconductor sensitive elements and spin-polarized electron sources.[1−5] At the same time, because of the many scientific issues it has involved, the GaAs photocathode has been used for preparation technology, surface model and activation mechanism research during the last few decades.[6−8] However, there are few reports concerning the optical properties of the photocathode, such as reflectance, transmittance and absorptivity. However, the optical properties spectra contain abundant information relevant to the optical performance of each layer's material.[9]

We concentrate on the optical properties of simply configured semiconductor, where the mathematical model is built as a single-layer structure.[10−12] One advantage of the model is the feature of a short cut calculation in the context of just one film system. However, the GaAs photocathode mentioned in this paper is composed of several layers, each of which should not be ignored. The transmittance of the four-layer photocathode has been calculated and simulated by means of thin film optics. Error modification is introduced into the theoretical transmittance calculation. Compared with the experimental curve, the modification coefficient and the thickness of each layer in the photocathode are fitted corresponding to the minimum relative error. Mechanism analysis is also made.

2. Experiment

A t-mode GaAs photocathode sample was obtained with a gradient-doping structure by molecular beam epitaxy (MBE), which provides precise control of the epilayer thickness and doping concentration level. After a series of processes, the photocathode module, as shown in Fig. 1, was formed. The GaAs active layer contained several sublayers of different thicknesses and doping concentrations. They were regarded as the whole layer with the thickness

*Project supported by the National Natural Science Foundation of China (Grant No. 60678043) and the Research and Innovation Plan for Graduate Students of Jiangsu Higher Education Institutions of China (Grant No. CX09B_096Z).

†Corresponding author. E-mail: bkchang@mail.njust.edu.cn

© 2011 Chinese Physical Society and IOP Publishing Ltd

of 1600 nm since their concentrations only changed a little.

The active layer was divided into four sections with the Be-doping concentration ranging from 1×10^{18} cm^{-3} to 2×10^{19} cm^{-3}. The Be-doping concentration of the window layer was also designed as 2×10^{19} cm^{-3}. During the entire growth procedure, the growth rates of GaAs and $Ga_{1-x}Al_xAs$ were both about 1000 nm/h and the crystalline quality of the epilayer was monitored during growth via reflection high-energy electron diffraction *in situ*. All growth processes were conducted in an ultrahigh vacuum (UHV) chamber at a base pressure of 4×10^{-10} Pa at 620 °C. After the growth, the superficial GaAs cap layer was first removed with a proper selective etch. After the coating layer was applied to the surface of window layer, glass was thermocompression bonded to the full wafer structure. Following that, the substrate and the stop layer were selectively etched away to expose the active layer to perform the activation. Thus, the sample was made into a four-layer photocathode module.

glass layer 5 mm	
Si_3N_4 antireflection layer	100 nm
$Ga_{0.5}Al_{0.5}As$ window layer	1000 nm, 2×10^{19} cm^{-3}
GaAs active layer 100 nm,	2.0×10^{19} cm^{-3}
GaAs active layer 300 nm,	5.0×10^{18} cm^{-3}
GaAs active layer 500 nm,	2.5×10^{18} cm^{-3}
GaAs active layer 700 nm,	1.0×10^{18} cm^{-3}

Fig. 1. The structure of the transmission-mode GaAs photocathode sample doped with gradient.

An UV-VIS-NIR spectrophotometer was used to measure the transmittance of the sample. The equipment had three detectors, which are photomultiplier operated under the visible and ultraviolet range, InGaAs and PbS detectors are used for the near infrared region. Using the double-optical-path method, the transmittance spectrum was obtained in the range from 600 nm to 1100 nm. The reference beam propagated in one optical path with a placement of the same glass as designed in the sample. The other path was designed as a measurement beam with the sample. In the measuring procedure, incident photons entered into the GaAs layer at normal incidence and passed through the following layers. Hence, the measured data was a comprehensive transmittance of the three thin layers in the full spectrum while the glass served as an exit medium.

3. Results and discussion

3.1. Simulation results

The t-mode GaAs photocathode can be simply seen as a four-layer film structure combined with glass/Si_3N_4/$Ga_{1-x}Al_xAs$/GaAs. The spectral transmittance of the photocathode, which is a function of the wavelength, the refractive index, extinction coefficient and thickness of each layer's material, can be derived using the matrix formula in thin film optics.[13]

Suppose that $\boldsymbol{Y}$ is the characteristic matrix of these thin films and their substrate in a multilayer structure, then

$$\boldsymbol{Y} = \begin{bmatrix} B \\ C \end{bmatrix} = \left(\prod_{j=1}^{k} \begin{bmatrix} \cos\delta_j & \mathrm{i}\sin\delta_j/\eta_j \\ \mathrm{i}\,\eta_j \sin\delta_j & \cos\delta_j \end{bmatrix} \right) \begin{bmatrix} 1 \\ \eta_g \end{bmatrix}, \quad (1)$$

where δ_j is the phase difference and equal to $(2\pi\eta_j d_j \cos\theta_j/\lambda)$, η_j is the optical constant of j-th layer' material and equal to $(n_j - \mathrm{i}\,k_j)$, d_j is the corresponding thickness and θ_j represents the refraction angle, determined by the refraction law. The symbols n_j and k_j are respectively the refractive index and extinction coefficient of the material, which are all functions of incident photon wavelength. The notation η_{g} is the optical constant of the substrate. Thus, the theoretical transmittance of the multilayer structure yields

$$T_{\mathrm{the}}(\eta_j, d_j; \lambda) = \frac{4\eta_0\eta_{\mathrm{g}}}{(\eta_0 B + C) \cdot (\eta_0 B + C)^*}. \quad (2)$$

When the transmittance of the photocathode is calculated, the multilayer structure can be considered as three thin layers that are deposited on glass. Therefore, three characteristic matrixes respectively represent the Si_3N_4 antireflection layer, the $Ga_{1-x}Al_xAs$ window layer and the GaAs active layer. The fairly thick glass layer needs to be seen as an incident or exit medium of the three-layer thin film depending on the measurement method.

In order to reflect the integrated effect of the optical properties in the selected wavelength range, the integral values of the spectral transmittance is calculated by making use of the discrete-wavelength integral. The calculation formula of integrated transmittance $T_{\rm f}$ is given by

$$T_{\rm f} = \frac{1}{\lambda_2 - \lambda_1} \cdot \int_{\lambda_1}^{\lambda_2} T_{\rm the}(\eta_j, d_j; \lambda)\, {\rm d}\lambda, \qquad (3)$$

where λ_1 and λ_2 are started and stopped wavelengths in the researched waveband.

Using Eqs. (1) and (2), transmittance curves are simulated by changing the thickness and number of the module's layers. All of the simulations are operated under normal incidence. The refractive index of Si_3N_4 is fixed at 2.06 and because Si_3N_4 is not an absorptive medium, the extinction coefficient is set at zero and the thickness is designed to be 100 nm.

The optical constants of $Ga_{1-x}Al_xAs$ are related to the Al mole component, which is selected as 0.5 in this paper. As shown in Fig. 2, the refractive index and extinction coefficient of $Ga_{1-x}Al_xAs$ are functions of incident photon wavelength and provided by the preparative database of optical constants. The database has been built with the aid of interpolation of the discrete points.[14] The values of x=0.5 are calculated by interpolating between those of x=0.491 and x=0.590. The absent data out of 827 nm are interpolated using those between 725 nm and 827 nm. The initial thickness of the window layer is set to be 1000 nm.

As to the active layer, GaAs corresponds to the value of x in $Ga_{1-x}Al_xAs$ as zero and its optical constants are also provided by the database mentioned above. At present the influence of doping is not taken into account in the database. The active layer should be thick enough to absorb photons, meanwhile be thin enough to emit electrons.[15] Thus, the initial thickness is 1600 nm.

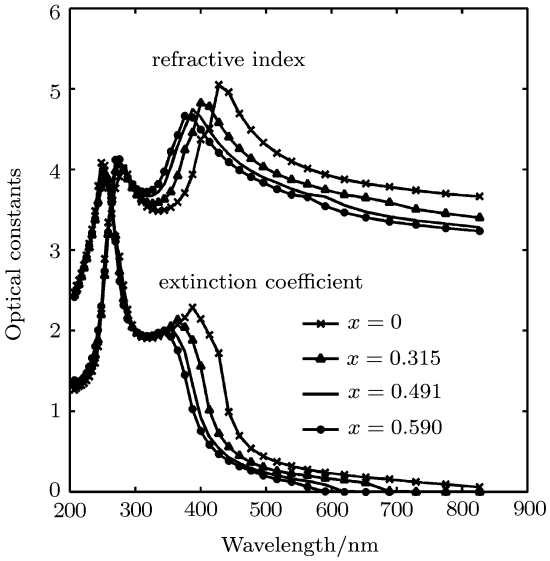
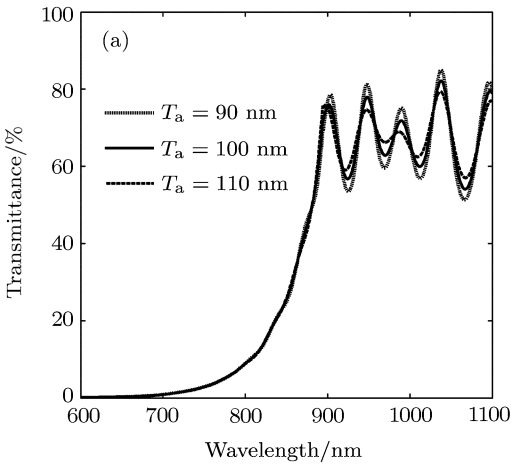
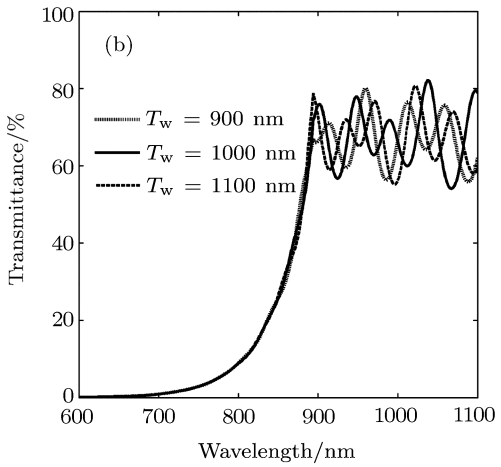


Fig. 2. The refractive index and extinction coefficient of $Ga_{1-x}Al_xAs$ material for different Al mole components x.[14]

During the simulation, the thickness of each layer varies and other parameters remain the same. The transition layer between $Ga_{1-x}Al_xAs$ and GaAs is neglected for the first run. Then, one transition layer with a thickness of 100 nm is put in the computing model, while the thicknesses of both the adjacent layers are cut down by 50 nm separately. Similarly, two transition layers that are 200 nm in total are calculated again with Eqs. (1) and (2), and the $Ga_{0.5}Al_{0.5}As$ and GaAs layers are changed to 900 nm and 1500 nm, respectively. The results are shown in Fig. 3 and Table 1.





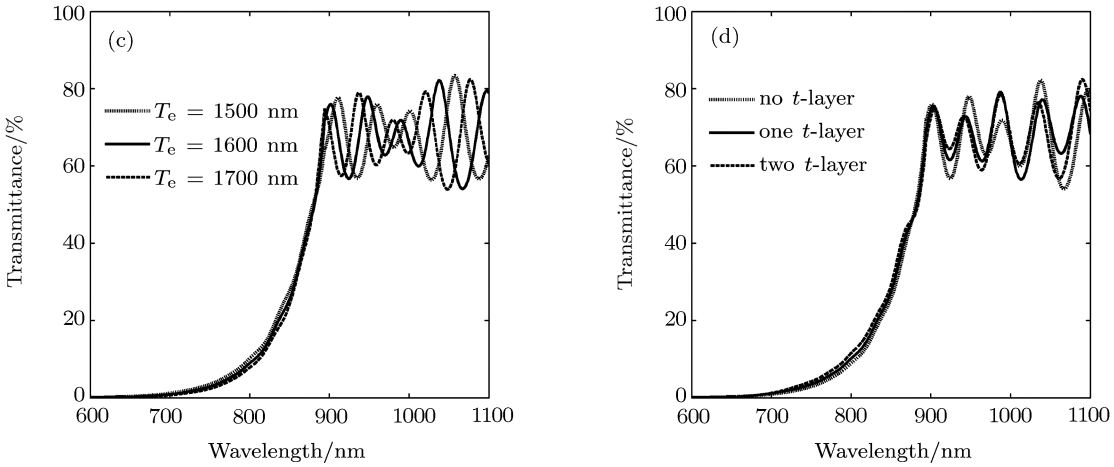


Fig. 3. Theoretical transmittance curves for a transmission-mode GaAs photocathode with different layers: (a) varied thickness of the Si_3N_4 layer, (b) varied thickness of the $Ga_{0.5}Al_{0.5}As$ layer, (c) varied thickness of the GaAs layer, and (d) varied transition layers (t-layers) with the thickness of 0, 100 and 200 nm in total separately.

The simulation results indicate that increasing thickness of the Si_3N_4 layer and the extra transition layers would lead to higher integrated transmittance. So the Si_3N_4 layer should be thin enough, the transition layers should be few enough and the other two layers should be relatively thick in order to obtain low transmittance, which benefits good photoemission. Yet the $Ga_{0.5}Al_{0.5}As$ and GaAs layers cannot be too thick for effective emission.[16] In addition, the deviating manner can be estimated by comparing the experimental curves with theoretical ones in reference with the results listed in Table 1.

Table 1. Relation between the structural parameters and transmittance of the transmission-mode GaAs photocathode.

thin film layer	various manner	short waveband	long waveband		integrated transmittance
		transmittance values	position of humps and hollows	amplitude	
Si_3N_4	increasing thickness	no change	no change	smaller	higher
$Ga_{0.5}Al_{0.5}As$	increasing thickness	no change	blue shift	irregular	lower
GaAs	increasing thickness	smaller	blue shift	irregular	lower
transition	increasing layers	bigger	no change	irregular	higher

3.2. Fitting results

Due to the inaccuracy of the thin film preparation process, the actual thicknesses of these layers in the sample would deviate from the design ones. As stated above, the spectral transmittance of the photocathode is a function of incident photon wavelength, the refractive index, extinction coefficient and the thickness of the individual layer, which are known except for the practical thickness. After setting specific initial values, we can fit each thickness in the sample in accordance with the experimental transmittance curve. Thus, the fitted thickness values and the deviations from the initial ones could be obtained.

When the curve fitting is carried out using the least squares method,[17] the relative error in measuring range is selected as controlled criterion given by

$$\Delta = \left| \frac{T_{\rm the}(\eta_j, d_j; \lambda) - T_{\rm exp}(\lambda)}{T_{\rm the}(\eta_j, d_j; \lambda)} \right| \cdot 100\%, \qquad (4)$$

where $T_{\rm exp}(\lambda)$ is the experimental transmittance data measured by the spectrophotometer. The fitting thickness of each layer can be obtained by minimizing the quadratic sum of the residual errors in Eq. (4).

The initial values are shown in Fig. 1 and the theoretical transmittance of the sample is achieved by calculating Eq. (2). Figure 4 also shows the theoretical curve. Obviously, there are big deviations of peak location and amplitude between the experimental and theoretical curves. Shown in Table 2, the relative deviation is up to 19.51% calculated by Eq. (4). Based on Table 1, the $Ga_{0.5}Al_{0.5}As$ and GaAs layers are likely thinner than the designed values in the paper.

A software program is designed for fitting each thickness of the GaAs photocathode automatically. The line of fitting I in Fig. 4 shows the first fitting results with the aid of the program. The fitting thicknesses are 83.4, 1008.7, and 1487.2 nm respectively for the Si_3N_4, $Ga_{0.5}Al_{0.5}As$ and GaAs layers. Meanwhile, the relative error reduced to 13.09%. However, the fitting method by means of the thickness variation just makes the humps and hollows sites consistent in the experimental and theoretical curves. As a result of the invalid thickness variation for the curve amplitudes, the method does not improve the fitting results further. Gradually to reach the lowest error, other factors need be considered for the adjustment of the amplitudes.

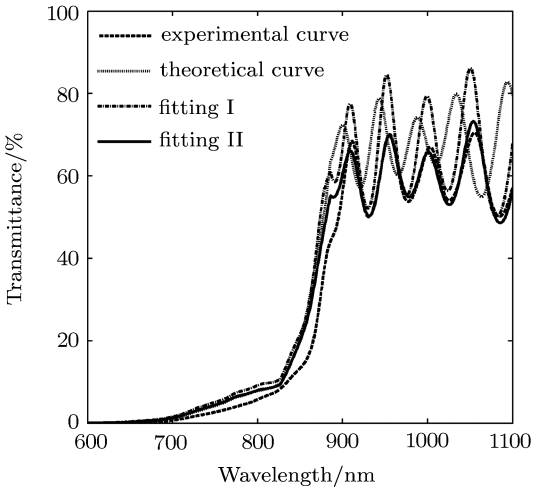


Fig. 4. The experimental and computed transmittance curves for the transmission-mode GaAs photocathode sample doped with gradient. The large error between 800 nm and 900 nm is derived from replacing light source of the spectrophotometer in the experiment.

Table 2. The fitting thicknesses and relative errors of the transmission-mode GaAs photocathode sample doped with gradient.

curve	$A_T(\lambda)$	thickness of Si_3N_4/nm	thickness of $Ga_{0.5}Al_{0.5}As$/nm	thickness of GaAs/nm	total thickness/nm	curve error[a)]/%
theoretical curve	1	100.0	1000.0	1600.0	2700.0	19.51
fitting I	1	83.4	1008.7	1487.2	2579.3	13.09
fitting II	0.88	99.5	1001.0	1500.5	2601.0	4.35
thickness error[b)]/%	/	0.50	0.10	6.22	3.67	/

[a)] The relative error between the experimental and theoretical transmittance curves.

[b)] The relative error between the designed and fitted thicknesses with modification.

According to the first fitting results, major amplitude errors are observed between the experimental curve and the fitting I curve in Fig. 4, and the experimental data are evidently less than the calculated ones in the full spectrum. The cause of deviation can be figured out between the experimental and the theoretical models. The calculation formula should be consistent with the experimental situation as far as possible. Inevitably, experimental instruments, external environments and testing technology can affect the measured results. For accuracy of fitting results, the theoretical transmittance needs to be modified by appropriate function. The modificatory formula is given by

$$T_{\mathrm{mod}}(\eta_j, d_j; \lambda) = A_T(\lambda) \cdot T_{\mathrm{the}}(\eta_j, d_j; \lambda), \qquad (5)$$

where $A_T(\lambda)$ is the dimensionless modificatory coefficient, which can be a constant or a function of wavelength. Based on the overall effect, the simplest situation is considered that $A_T(\lambda)$ is a constant lying between 0 and 1.

Fitting calculation is again made by using Eqs. (2) and (5), while $A_T(\lambda)$ is also fitted and equal to 0.88. The resulting curve is shown as the line of fitting II in Fig. 4, and the fitting thicknesses are listed in Table 2. Seen from the fitting curves, a significant consistency in phase and amplitude between experimental curve and fitting II curve can be found. The relative error

has dropped to 4.35%. In addition, the deviations of thicknesses from the designed values are smaller than those of the first fitting. The relative errors of the thicknesses are also calculated in the last row of Table 2. It demonstrates that the relative deviations of each layer and the overall thin-film thicknesses in the module are all controlled within 7%. The fitting results indicate that the Si_3N_4 and $Ga_{0.5}Al_{0.5}As$ layers scarcely change in the experiment, and the GaAs layer attenuates the thickness due to etching.

3.3. Mechanism analysis

A comparison of the experimental transmittance with the theoretical value demonstrates that the data analysis has to be performed by taking the module structure into account. In the analysis of film parameters, there are three main factors ought to be considered including glass surface roughness, a non abrupt substrate-film interface and the presence of surface contaminants such as oxides.[18] These factors are all irrelevant to wavelength, which causes the experimental transmittance to decline in the whole waveband. Thus a constant coefficient should be chosen to modify the theoretical formula, as shown in Eq. (5).

According to the fabrication technology of the photocathode sample and test method of the transmittance, the effect from the glass layer is evident, but was not considered in the theoretical calculation. In a realistic model, the photocathode deposits on the glass substrate. The reflection of the glass and the glass–Si_3N_4 interface needs careful consideration. The simulation is shown in Fig. 5, where the reflectance and transmittance spectra of glass in the sample can be calculated. Both the spectra are anomalous lines and mutually complementary due to the absence absorption.

The integral average should be used to indicate the results. The integral transmittance is obtained from Eq. (3) for single layer, and the integral reflectance is equal to $(1 - T_f)$. Suppose that the thickness of the glass layer is 5 mm and the refractive index is 1.49. The integral results are obtained as 93.73% for transmittance and 6.27% for reflectance in the range from 600 nm to 1100 nm. So the constant coefficient of glass' influence can approximately be 0.94, which is recorded as A_1.

In addition, the photocathode sample consists of four layers. So, the interfacial effect exists between the different layers, such as the adjacent layers of glass–Si_3N_4, Si_3N_4-$Ga_{0.5}Al_{0.5}As$ and $Ga_{0.5}Al_{0.5}As$-GaAs.[19,20] Due to the discrepancy of material performance, inter-diffusion of different kinds of particles has been observed. Specifically, a similar interface makes Al-ion diffuse gradually between the $Ga_{0.5}Al_{0.5}As$ and GaAs layers. It should be taken into account that in one or more $Ga_{1-x}Al_xAs$ subdivision layers x value changes with thickness, varying between 0 and 0.5. Thus some new interfaces form in the module. When incident photons reach these interfaces, complex process of reflection and transmission would take place. The global transmittance will be changed according to A_2 in proportion. The number of electrons, which are generated in the $Ga_{1-x}Al_xAs$ layer and flow into the GaAs layer, is formulated by integrating the local electron concentrations throughout the GaAs layer. An explicit representation of the detailed process of logic reasoning and numerical calculating is shown in Ref. [21].

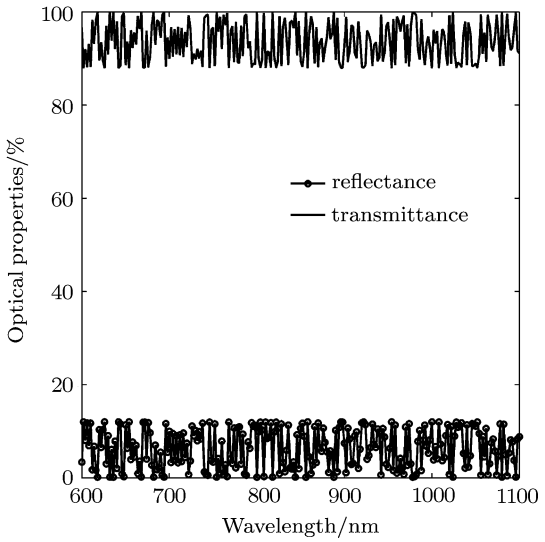


Fig. 5. The reflectance and transmittance curves of the glass layer in the transmission-mode GaAs photocathode sample.

The experimental transmittance is also measured after a series of process flows, among which the influence outside is inescapable. Obviously, the outermost layer (the active layer of GaAs) is oxidized easily by ambient air. So the alteration of the thickness and component produces an effect on the transmittance curve. Let the ratio of the change be registered as A_3.

Based on an overall consideration of various factors, the experimental values are less likely than the computational ones. It is reasonable that the transmittance multiplies a fitted coefficient for error com-

pensation. The modificatory coefficient should be calculated with the following expression

$$A_{\mathrm{T}}(\lambda) = A_1 \cdot A_2 \cdot A_3. \tag{6}$$

In this way, more accurate thicknesses can be obtained after the modificatory process. The fitting results listed in Table 2 indicate this.

4. Conclusions

A t-mode GaAs photocathode has been prepared with gradient-doping by means of MBE, and the transmittance curve of the sample was measured in the spectral range from 600 nm to 1100 nm with a spectrophotometer. The theoretical transmittance was calculated and simulated based on the matrix formula in thin film optics. The simulation results demonstrate that fewer transition layers, a thinner Si_3N_4 layer and higher thicknesses of the $Ga_{0.5}Al_{0.5}As$ and GaAs layers should obtain low transmittance, which favours photoemission for GaAs photocathodes. In addition, the experimental and computational curves are fitted with primitive and modificatory formulas, respectively. The result indicates that a reduction in the transmittance relative error emerges from 19.51% to 4.35% after modifying. The thicknesses of the Si_3N_4, $Ga_{0.5}Al_{0.5}As$ and GaAs layers are obtained respectively as 99.5, 1001.0, and 1500.5 nm, meanwhile the deviations between the designed and fitted thicknesses are controlled below 7%. The result demonstrates that the presence of glass layer roughness, interface effects of layers and surface oxides claim a modificatory term to draw in calculation for error compensation. The term can be a constant or function needed fitting as in different cases. The results and methods in this paper are significant for the constructional design and spectral fitting of t-mode GaAs photocathodes.

Acknowledgments

The authors are grateful to Shi Feng, Guo Hui, Hu Cang-Lu, Xu Xiao-Bing and Cheng Wei of Key Laboratory of Low Light Level Technology of COSTIND for their assistance with the experiment.

References

[1] Liu Z, Sun Y, Peterson S and Pianetta P 2008 *Appl. Phys. Lett.* **92** 241107

[2] Zhang Y J, Chang B K, Yang Z, Niu J and Zou J J 2009 *Chin. Phys.* B **18** 4541

[3] Mulhollan G A, Subashiev A V, Clendenin J E, Garwin E L, Kirby R E, Maruyama T and Prepost R 2001 *Phys. Lett.* A **282** 309

[4] Liu Z, Machuca F, Pianetta P, Spicer W E and Pease R F W 2004 *Appl. Phys. Lett.* **85** 1541

[5] Ding H B, Pang W N, Liu Y B and Shang R C 2005 *Acta Phys. Sin.* **54** 4097 (in Chinese)

[6] Moré S, Tanaka S, Tanaka S, Fujii Y and Kamada M 2000 *Surf. Sci.* **454–456** 161

[7] Spicer W E and Herrera-Gomez A 1993 *Proc. SPIE* **2022** 18

[8] Su C Y, Spicer W E and Lindau I 1983 *J. Appl. Phys.* **54** 1413

[9] Witit-anun N, Rakkwamsuk P and Limsuwan P 2008 *Proc. SPIE* **6793** 67930J

[10] Zhang X J, Ma H L, Li Y X, Wang Q P, Ma J, Zong F J and Xiao H D 2006 *Chin. Phys.* **15** 2385

[11] Larruquert J I, Méndez J A, Aznárez J A, Tremsin A S and Siegmund O H W 2002 *Appl. Opt.* **41** 2532

[12] Lévêque G and Villachon-Renard Y 1990 *Appl. Opt.* **29** 3207

[13] Tang J F, Gu P F, Liu X and Li H F 2006 *Modern Optical Thin Film Technology* (Zhejiang: Zhejiang University Press) pp. 20–32 (in Chinese)

[14] Aspnes D E, Kelso S M, Logan R A and Bhat R 1986 *J. Appl. Phys.* **60** 754

[15] Du X Q *Study of GaAs Photocathode with High Performance* (Ph. D. Thesis) (Nanjing: Nanjing University of Science and Technology) (in Chinese)

[16] Yang Z, Zou J J and Chang B K 2010 *Acta Phys. Sin.* **59** 4290 (in Chinese)

[17] Jung C and Rhee B K 2002 *Appl. Opt.* **41** 3861

[18] Yu P Y and Cardona M 1996 *Fundamentals of Semiconductors* (Berlin: Springer)

[19] Kawashima Y, Ide T, Aoyagi S and Kudo M 2004 *Appl. Surf. Sci.* **231** 800

[20] Baek J, Kovar D, Keto J W and Becker M F 2006 *Appl. Opt.* **45** 1627

[21] Watanabe K, Nakamura Y, Ichikawa M, Kuboya S, Katayama R and Onabe K 2009 *J. Vac. Sci. Technol.* B **27** 1874