Effect of Cs adsorption on the photoemission performance of GaAlAs photocathode

Xinlong Chen,¹ Muchun Jin,¹ Yugang Zeng,² Guanghui Hao,¹ Yijun Zhang,¹ Benkang Chang,^{1,*} Feng Shi,³ and Hongchang Cheng³

¹School of Electronic and Optical Engineering, Nanjing University of Science and Technology, Nanjing 210094, China

²Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, China

³Science and Technology on Low-Light-Level Night Vision Laboratory, Xi'an 710065, China

*Corresponding author: bkchang@mail.njust.edu.cn

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The effect of Cs adsorption on the photoemission performance of a reflection-mode GaAlAs photocathode in an ultrahigh vacuum chamber has been investigated. The experiments for Cs/O activation, multiple recaesiation, and degradation are performed on a GaAlAs photocathode. Meanwhile, the Cs/O activated and recaesiated photocurrent curves, degraded photocurrent curves, and spectral response curves are measured and analyzed. Besides, the performance parameters of the photocathodes are obtained by using the formula to fit with the experimental quantum efficiency curves. The results show that the Cs atoms not only make the atomically clean surface form the negative electron affinity, but also make the degraded photocathode recover to a good level. The quantum efficiency and the lifetime of GaAlAs photocathode become lower with increasing the recaesiation times. © 2014 Optical Society of America

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1. Introduction

In recent decades, III-V semiconductor negative electron affinity (NEA) photocathodes have been widely used in night vision image intensifiers, ultraviolet detection, and potential electron sources for the next-generation electron accelerators due to their high quantum efficiency, low-energy spread, low dark current emission, and fast response speed [1-4]. At present, the photodiodes are becoming increasingly important in many applications for their good sensitivity, low voltage requirement, small size, and low cost [5,6]. However, in comparison with photodiodes, the photocathodes have an obvious advantage in the applications that need detection of low light levels.

The NEA GaAlAs photocathode is a potential photocathode for the next-generation electron accelerators and marine detection due to a long lifetime and controlled threshold wavelength [7,8]. For all kinds of NEA photocathodes, the surface vacuum level should be pulled below the conduction band minimum of the bulk to form the NEA surface. The activation of a NEA photocathode surface has in large part been an art developed from the original work of alkali adsorption onto ideal metal and cleaved semiconductor surfaces [9,10]. Following the development of the activation craft, the "yo-yo" activation technology has been widely used for achieving the NEA surface [11], where Cs deposition and subsequent oxidation are repeated several times, leading to the formation of a surface dipole layer [12]. Though both of Cs and O atoms take part in the formation of the NEA surface, the Cs atoms play a leading role

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because the vacuum level at the surface could be reduced below the conduction band minimum by depositing an approximate monolayer of Cs [13].

However, there are few studies about the photoemission properties of NEA GaAlAs photocathodes so far, much less the mechanism of Cs adsorption on GaAlAs surface. In this paper, a report about the effect of Cs adsorption on the photoemission performance of a reflection-mode GaAlAs photocathode in an ultrahigh vacuum (UHV) chamber is proposed. The Cs/O activation and degradation under the illumination of white light are performed on a GaAlAs photocathode. Besides, multiple recaesiated photocurrent curves, the spectral response curves, and the degraded photocurrent curves are measured. The effects of Cs on the photoemission performance of a GaAlAs photocathode are analyzed and discussed in detail by combining with the experimental results.

2. Experiment and Results

A 2-in.-diameter reflection-mode photocathode sample was grown on the high-quality n-type GaAs(100)oriented substrate by metal organic chemical vapor deposition. The epitaxial layers consisted of a Ga_{0.21}Al_{0.79}As buffer layer and a Ga_{0.37}Al_{0.63}As emission layer; both of the GaAlAs layers were p-type zinc (Zn) doped. The Ga_{0.37}Al_{0.63}As emission layer was doped in the form of an exponential ranging from 10^{19} cm⁻³ to 10^{18} cm⁻³, which could increase the photoemission of the photocathode [14]. The total thickness of Ga_{0.37}Al_{0.63}As emission layer was 1.6 µm. The thickness of Ga_{0.21}Al_{0.79}As buffer layer was 0.5 µm, and the doping concentration was 10^{19} cm⁻³.

In this study, one 10×10 mm sample cut from the as-grown sample was treated by a series of experiments. The sample was first treated in a 4:1:100 solution of sulfuric acid (98%) to hydrogen peroxide (30%) to deionized water for 2 min. After that, the sample was transferred into the UHV chamber with a base pressure of less than 10^{-7} Pa . The sample was heated under the temperature of about 650°C for 20 min. for removing the surface contamination such as carbon and oxides, and the GaAlAs sample obtained an atomically clean surface. When the sample cooled down to room temperature, the Cs/O activation for the sample was performed in the UHV chamber, in which the cesium source was kept continuous, and the oxygen source was introduced periodically [15,16]. The Cs/O activation process consists of the first Cs exposure (caesiation) and the Cs/O alternate activation. The Cs source used in the activation is in the form of a mixture of cesium chromate and zirconium-aluminum alloy powder in a thin-walled nickel tube, and the O source is in the form of barium peroxide in a nickel tube. The Cs and O atoms are released into the chamber by heating the nickel tube, and the Cs or O flux is accurately controlled by adjusting the heating current. During the activation process, the photocurrent was monitored

when the photocathode was illuminated by a halogen tungsten lamp. The photocurrent of photocathode was controlled and monitored by a self-developed multiinformation measurement system [17,18]. After the activation, the spectral response curve of reflectionmode GaAlAs photocathode was measured by the on-line spectral response measurement system.

After the Cs/O activation, the GaAlAs photocathode degraded with time under the condition of illumination in the UHV chamber, the pressure of the chamber was about 1×10^{-7} Pa. After the first degradation, the photocathode was recaesiated by applying the fresh Cs, and the recaesiated photocurrent and spectral response curves were measured. After that, the degraded curve of the first recaesiated photocathode was measured under illumination with the same intensity. The second and third recaesiated experiments were performed on the photocathode, and the corresponding degradation processes also were executed. Besides, the corresponding photocurrent curves and the spectral response curves were recorded.

The Cs/O activated photocurrent curve and the three recaesiated photocurrent curves are shown in Fig. <u>1</u>. The photocurrent curves are normalized



Fig. 1. Photocurrent curves of GaAlAs photocathodes. (a) Cs/O activated photocurrent curve. (b) Curves 1, 2, and 3 are corresponding to the first, second, and third recaesiated photocurrent curves, respectively.

using the maximum of the Cs/O activated photocurrent curve as the norm. After each activation, the degraded experiment was performed on the GaAlAs photocathode. The photocurrents of photocathodes after the degradation are similar, which could be found from the beginning photocurrents in Fig. <u>1(b)</u>. During the recaesiations, the photocurrents increase obviously in a short time, but they could not reach the maximum of the Cs/O activated photocurrent curve. Besides, we can find that the peak photocurrents and the recaesiated time decrease as the recaesiation times increase. The peak photocurrents of recaesiation are higher than the maximum photocurrent of caesiation in the Cs/O activation process.

The normalized degraded photocurrent curves of GaAlAs photocathodes are shown in Fig. 2. The 1/e of the degraded photocurrents is usually recorded as a standard to value the lifetime of the NEA photocathodes [12]. Though the preparation chamber of GaAlAs photocathode has an UHV, some residual gas molecules are difficult to be extracted by the vacuum pump. These residual gas molecules would damage the Cs-O active layer, which is the main factor causing the degradation of the photocathode. In Fig. 2, the lifetime of GaAlAs photocathode decreases as the recaesiation times increase, which could be caused by the cumulative adsorption of harmful gas molecules on the photocathode surface in the degradation process. We guess that the impurities covered on the surface may prevent the formation of dipoles, though the UHV chamber is full of Cs atoms in the recaesiation process.

Before each degradation, the spectral response curves of GaAlAs photocathodes were measured, which are shown in Fig. <u>3</u>. The spectral response curves of the recaesiated photocathodes are lower obviously than that of the Cs/O photocathode. For the recaesiated photocathodes, the first recaesiated photocathode has the largest spectral response, while the third recaesiated photocathode has the smallest spectral response. The cut-off wavelengths of four





Fig. 3. Spectral response curves of GaAlAs photocathodes. Curves 1, 2, 3, and 4 are corresponding to the spectral response curves after the Cs/O activation; the first, second, and third recaesiations, respectively.

spectral response curves are similar, which are decided by the Al composition of the emission layer.

3. Analysis and Discussion

As we know, the degradation of an NEA photocathode in the UHV chamber is mainly related to residual gasses such as CO_2 , H_2O , CO, and so on. The residual gas molecules would be adsorbed on the photocathode surface and damage the original NEA surface. In order to study the degradation mechanism of a GaAlAs photocathode, we assume degradation of the photocathode is mainly caused by the adsorption of residual gasses, though there have been a lot of impact factors on the degradation of NEA photocathodes such as the ion-back-bombardment, the operation at high voltage, and the photocathode heat load generated by high-intensity illumination [<u>19</u>].

Based on the research of Whitman, the assumption about the activity region is mentioned [20,21]. The active layer of an NEA GaAlAs photocathode is constituted by a lot of activity regions. Once one activity region of an active layer adsorbs a harmful gas molecule, the region would never form the dipole and lose the talent of photoemission. We assume the number of activity regions per unit area is n. When the vacuum pressure is p, the collision times of harmful gas molecules on the photocathode surface per unit area and per unit time are cp; c is the collision coefficient. The adhesion coefficient of harmful gas molecules on the photocathode surface under the unit of light intensity is A; the light intensity is H. At time t, the ratio of the residual activity region per unit area and the total activity region per unit area is $\theta(t)$.

According to the assumption above, the photocurrent of the photocathode with time I(t) is

$$I(t) = I_0 \theta(t), \tag{1}$$

Fig. 2. Degraded photocurrent curves of GaAlAs photocathode. Curves 1, 2, 3, and 4 are corresponding to the degraded photocurrent curves after the Cs/O activation, the first, second, and third recaesiations, respectively.

where I_0 , namely I(0), is the initial photocurrent before degradation, and $\theta(0) = 1$.

 $\theta(t)$ is obtained by recurring:

$$\theta(1) = 1 - \frac{AHcp\theta(0)}{n} = 1 - \frac{AHcp}{n}, \qquad (2)$$

$$\theta(2) = \theta(1) - \frac{AHcp\theta(1)}{n} = \theta(1)\left(1 - \frac{AHcp}{n}\right) = [\theta(1)]^2.$$
(3)

So

$$\theta(t) = [\theta(1)]^t = \left(1 - \frac{AHcp}{n}\right)^t,\tag{4}$$

$$I(t) = I_0 \theta(t) = I_0 \left(1 - \frac{AHcp}{n} \right)^t.$$
(5)

From Eq. (5), it could be found that the changed trend of photocurrent degrading with time is close to the exponential form, and the degraded formula could be described by

$$I(t) = I_0 \, \exp\left(-\frac{t}{\tau}\right). \tag{6}$$

The degraded photocurrent curves in Fig. 2 conform to the degraded rule of Eq. (6). Combined with Eqs. (5) and (6), the lifetime τ could be obtained:

$$\tau = -\frac{1}{\ln\left(1 - \frac{AHcp}{n}\right)} \approx \frac{n}{AHcp} \quad \frac{AHcp}{n} \ll 1.$$
(7)

According to Eq. (7), it could be found that the lifetime is proportional to the number of activity regions on the surface. The better vacuum pressure would make the lifetime of the photocathode increase undoubtedly. The lifetime is inversely proportional to the collision and adhesion coefficients, which depends on the harmful gasses in the UHV chamber. We have proved that the lifetime would decrease with an increase of the incident light intensity; the harmful gas molecules may adsorb on the photocathode surface under the high intensity illumination [7].

Based on the analysis above, we can conclude that shortening of lifetime with increasing of recaesiation times is induced by decreasing of activity regions on the surface. As mentioned above, some impurity molecules adsorbing on the photocathode surface could not be removed in the following recaesiation and degradation processes. Then the activity regions would further decrease as the accumulating of impurity molecules in the process of multiple recaesiations and degradations. We think the degraded surface also could form the dipole after applying the fresh cesium, and the dipoles only form on the activity regions. According to the dipole formation process, Zou *et al.* has put forward an equation to explain the evolution of photocurrent curves of the first

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Cs exposure on the atomically clean GaAs(100) surfaces [22]:

$$\frac{I'(t)}{I_s} = 1 - \varphi(t) = 1 - \left(1 - \frac{N_a}{N_t}\right)^t,$$
(8)

where I'(t) is the photocurrent at time t, I_s is the total photocurrent increase during the process of Cs exposure, N_a is the rate of dipoles formation, denoting the number of dipoles formed per unit photocathode surface area and per unit time. N_t is the final total number of dipoles formed per unit area, and $\varphi(t)$ represents the coverage, which is the percentage of a monolayer covered by Cs atoms at time t.

Different with the dipole-forming mechanism of Cs adsorption on the cleaned GaAlAs(100) surface, the Cs atoms also could react with the oxide molecules on the degraded surface in the recaesiation process. However, the evolution of the recaesiated photocurrent curves also obeys the photocurrent evolution of the first Cs exposure on the cleaned surface.

In Fig. <u>1(b)</u>, the photocurrents almost have no change in the beginning dozens of seconds because the Cs source needs a period of preheating before releasing the Cs atoms. So the preheating stage is wiped from the photocurrent curves in Fig. <u>4</u>. In Fig. <u>1(b)</u>, it could be found that the beginning photocurrent values of three photocurrent curves are similar, which shows that the number of residual dipoles on the degraded surfaces is almost the same. For better discussion of photocurrent curve evolution, the beginning photocurrent values are subtracted from the three photocurrent curves in Fig. <u>1(b)</u>. Besides, the photocurrent curves, which are shown in Fig. <u>4(a)</u>, are normalized using the maximum of the first recaesiated photocurrent curve as the norm.

Equation (8) is used to fit the photocurrent curves of the recaesiated photocathodes; the fitting photocurrent curves are shown in Fig. 4. We set $B = N_a/N_t$, the fitted B for three recaesiated photocathodes, which are 1/70, 1/62, and 1/55, respectively. During all recaesiation processes, the evaporation velocity of Cs is constant, so the rates of dipoles formation N_a are similar to the three recaesiated GaAlAs photocathodes. The ratio of N for the three recaesiated photocathodes is 70:62:55. The result shows that the dipoles formed on the activity regions decrease with the recaesiation times and also proves that more dipoles are helpful in increasing the lifetime of a photocathode.

Quantum efficiency is the direct parameter to evaluate the photoemission performance of photocathodes, which is influenced by the minority carrier diffusion length, the back interface recombination velocity, and the surface electron escape probability. The quantum efficiency formula of the exponentialdoping reflection-mode GaAlAs photocathodes could be obtained by solving 1D continuity equations with the given boundary conditions [7]. The formula is



Fig. 4. Experimental (solid lines) and theoretical (dashed lines) photocurrent curves of (a) the first recaesiation, (b) second recaesiation, and (c) third recaesiation.

$$Y_{\rm RE} = \frac{P(1-R)\alpha_{hv}L_D}{\alpha_{hv}^2 L_D^2 - \alpha_{hv}L_E - 1} \times \left[\frac{N(S-\alpha_{hv}D_n)\exp(L_E T_e/2L_D^2 - \alpha_{hv}T_e)}{M} - \frac{Q}{M} + \alpha_{hv}L_D\right],$$
(9)

where

$$N=\sqrt{L_E^2+4L_D^2},$$

$$\begin{split} S &= \mu |E| + S_v, \\ L_E &= \mu |E| \tau = \frac{q |E|}{k_0 T} L_D^2, \\ M &= (ND_n/L_D) \cosh(NT_e/2L_D^2) \\ &+ (2 \; SL_D - D_n L_E/L_D) \sinh(NT_e/2L_D^2), \\ Q &= SN \; \cosh(NT_e/2L_D^2) \\ &+ (SL_E + 2D_n) \sinh(NT_e/2L_D^2), \end{split}$$

In Eq. (9), P is the surface electron escape probability, R is the reflectivity on the surface of photocathode, α_{hv} is the absorption coefficient of the emission layer, L_D is the electron diffusion length in the emission layer, S_v is the back interface recombination velocity, T_e is the thickness of the emission layer, D_n is the electron diffusion coefficient, τ is the minority carrier lifetime, and L_E is the electron drift length under the built-in electric field E. The average distance of photoelectrons moving toward the surface in the exponential-doping GaAlAs layer is defined by the electron diffusion and drift length L_{DE} [23]:

$$L_{DE} = \frac{1}{2} \left(\sqrt{L_E^2 + 4L_D^2} + L_E \right), \tag{10}$$

$$E = -\frac{k_B T}{q} \cdot \frac{\ln(N_0/N_s)}{T_e},\tag{11}$$

where k_B is the Boltzmann constant, T is the absolute temperature, q is the electron charge, N_0 is the doping concentration at the back interface of emission layer, and N_s is the doping concentration at the surface.

The quantum efficiency curve can be obtained by transforming the spectral response curve according to the following equation [24]:

$$Y(hv) \approx 1.24 S_{\lambda}/\lambda,$$
 (12)

where Y(hv) is quantum efficiency and S_{λ} is the spectral response value of the corresponding wavelength λ .

The experimental and theoretical quantum efficiency curves of GaAlAs photocathodes are shown in Fig. <u>5</u>; the theoretical curves are obtained by fitting to the experimental curves using Eq. (9). In the fittings, the α_{hv} is measured by the nkd-system, and the value of D_n is approximately set to be 10 cm²/s for Ga_{0.37}Al_{0.63}As [25]. Besides, the values of T_e and R are 1.6 µm and 0.3, respectively.

The fitting performance parameters of quantum efficiency for the Cs/O activated, the first, second, and third recaesiated photocathodes, are shown in Table. <u>1</u>. We can find that fitting S_v and L_{DE} is similar for four photocathodes because they are decided by the intrinsic property of the photocathode material. It also could be found that the difference of surface electron escape probability P is evident



Fig. 5. Experimental (solid lines) and theoretical (dashed lines) quantum efficiency curves of exponential-doping reflection-mode GaAlAs photocathodes. Curves 1, 2, 3, and 4 are corresponding to the Cs/O activated photocathode; the first, second, and third recaesiated photocathodes, respectively.

for the four photocathodes, which is determined by the evolution of the surface barrier.

The band structure and surface barrier of the exponential-doping reflection-mode GaAlAs photocathode are shown in Fig. 6. For the exponential-doping reflection-mode GaAlAs photocathode, the incident photons are absorbed mainly in the GaAlAs emission layer. The excited photoelectrons near the back interface would thermalize rapidly at the bottom of conduction band. Then they would diffuse and drift to the surface under the role of the built-in electric field and emit into a vacuum after crossing the surface barrier. The excited photoelectrons at the near surface would directly cross the surface barrier and emit into a vacuum. For the Cs/O activated photocathode, the surface barrier based on the double-dipole model comprises two approximately straight lines with different slopes, namely, barrier I and II, which decides the surface electron escape probability [12]. Some low-energy photoelectrons could not cross the surface barrier when the barrier is high. On the other hand, the amount of photoelectrons crossing the barrier would decrease when the thickness of the barrier increases.

In comparison with the surface barrier of the Cs/O activated photocathode, both the thickness and height of the surface barrier for the degraded photocathode increase, as shown in Fig. <u>6</u>. We think the surface barrier of the recaesiated photocathode would not strictly comprise two approximately straight lines with different slopes because there

Table 1. Fitting Parameters of Quantum Efficiency for Cs/O Activated: the First, Second, and Third Recaesiated Photocathodes

Photocathode	$S_v/({ m cm/s})$	Р	$L_{ m DE}/\mu{ m m}$
Cs/O activated	1×10^{6}	$0.48 \\ 0.34 \\ 0.3 \\ 0.27$	0.81
First recaesiated	1×10^{6}		0.81
Second recaesiated	1×10^{6}		0.8
Third recaesiated	1×10^{6}		0.81



Fig. 6. Band structure and surface barrier of the exponentialdoping reflection-mode GaAlAs photocathode. CBM, conduction band minimum; VBM, valence band maximum.

are residual oxides of Cs on the photocathode surface. The forming mechanism of fresh Cs adsorption on the degraded photocathode is difficult, so we use one approximately straight line with a slope to represent the surface barrier of the recaesiated photocathode. It could be concluded that the thickness and height of surface barrier for the recaesiated photocathode would increase as the increasing of recaesiation times.

4. Conclusion

In this paper, we have studied the effect of Cs adsorption on the photoemission performance of a reflection-mode GaAlAs photocathode in the UHV chamber. The Cs/O activation, recaesiation, and degradation experiments have been performed on a GaAlAs photocathode. We have obtained and analyzed the Cs/O activated photocurrent, the recaesiated photocurrent, the degraded photocurrent, and the spectral response curves. The performance parameters of GaAlAs photocathodes are obtained by fitting with the experimental quantum efficiency curves. The results show that the Cs atoms could make the degraded photocathode recover to a good level, but the quantum efficiency and lifetime of the GaAlAs photocathode decrease with the increase of recaesiation times. Besides, the variations of quantum efficiency curves for the Cs/O activated and recaesiated photocathodes are decided by the evolution of the surface barriers. This work helps us to understand the mechanism of Cs adsorption on the photocathode surface and the effect of Cs on the photoemission performance of a GaAlAs photocathode. Some results and discussions of this study may be useful references for studying other III-V NEA photocathodes.

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