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Enhancement of near-infrared response of InGaAs photocathode through interaction of 1064 nm light with activated surface



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ABSTRACT

To enhance the near-infrared response of InGaAs photocathodes, we proposed an improved activation recipe, including conventional Cs/O or Cs/NF₃ activation with white light and a subsequent re-activation with 1064 nm light. *In-situ* activation, re-activation and characterization were performed to investigate the relationship between the spectral response and the emitting surface. Multiple activation experiments demonstrated that reactivation illuminated by the 1064 nm light after conventional Cs/O activation effectively enhanced the spectral response in the near-infrared range, e.g., by a factor of 2–6 at 1064 nm, and this recipe had no enhancement effect on the spectral response of the Cs/NF₃ activated samples. After Cs/O activation, the surface barrier of the activation surface was further optimized through the interaction of the 1064 nm light with the activated surface during re-activation. The minor optimization of the surface barrier could significantly improve the tunneling probability of the electrons excited by the near-infrared light, as the interaction of ultraviolet-blue light with the activated surface during re-activation could not be replaced by ultraviolet-blue light, as the interaction of ultraviolet-blue light with the activated surface barrier.

1. Introduction

Since its invention, the III-V photocathode has been widely applied in photon detection, night vision, solar energy and polarized electron sources because of its high quantum efficiency, low thermal emittance, high time resolution and high spin polarization [1-4]. Compared with GaAs and multi-alkali photocathodes, InGaAs photocathodes have greater potential to extend the near-infrared (NIR) response, thus providing an approach for realizing the ultrafast and low-noise detection or imaging of extremely weak NIR light [5–8]. To lower the work function and increase the probability of photoelectron escape, the activation recipe of this vital technology has been extensively investigated in the field of photocathode research [9-12]. A negative electron affinity (NEA) photocathode can be fabricated by depositing an electropositive substance (e.g., cesium) and an electronegative substance (e.g., O2 or NF₃) on the photocathode surface to lower its surface vacuum level below the conduction band minimum in the bulk [13-15]. However, for InGaAs photocathodes, the electrons excited by the NIR photons will still have difficulty tunneling through the surface barrier, due to extremely low electron energy, leading to the low quantum efficiency of InGaAs photocathodes in the NIR range [6,16,17]. Hence, improving the NIR photoemission performance of InGaAs photocathodes has become a technical challenge that need to be overcome. So far, various activation recipes have been used to prepare NEA GaAs photocathodes, resulting in different photoemission characteristics [12,18–20]. However, few studies have explored superior activation recipes to improve the NIR response of NEA InGaAs photocathodes [21–23]. In addition, it remains to be verified whether the effect of different activation recipes on InGaAs photocathodes will exhibit the same phenomenon as GaAs photocathodes.

In the conventional activation process of InGaAs photocathodes, a white light source consisting of a tungsten halogen lamp is typically used to activate cathode samples [16,21,24,25]. By alternately switching on Cs and O sources, the photocurrent will increase in a wavy shape and the activation procedure will terminate when the photocurrent reaches the maximum value. At this time, the photocathode can be considered in the

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Received 28 December 2022; Received in revised form 5 February 2023; Accepted 13 February 2023 Available online 16 February 2023 0169-4332/© 2023 Elsevier B.V. All rights reserved. best NEA state. Unlike GaAs photocathodes, the emission performance of InGaAs photocathodes in the NIR range is much more important than in the visible range. However, because visible light can excite considerably more photoelectrons than NIR light when a tungsten-halogen lamp is used, the photocurrent excited by this white light source cannot precisely exhibit the NIR emission performance of an InGaAs photocathode. Hence, the NIR response of an InGaAs photocathode will not be optimal using the tungsten-halogen lamp light source alone during activation. To enhance the response in the NIR range, especially at 1064 nm, we proposed an improved InGaAs photocathode activation recipe, which included two steps: First, conventional Cs/O or Cs/NF3 activation was performed with a tungsten-halogen lamp to make the photocathode surface reach the NEA state. Second, additional activation, denoted as re-activation, was carried out with a 1064 nm laser. which was terminated when the photocurrent reached the new maximum. The purpose of the first activation process was to extend the long wavelength cutoff of the InGaAs photocathode to 1064 nm, allowing the accurate monitoring of the photocurrent after switching the 1064 nm light. Through the interaction of the 1064 nm light with the activated surface, the surface barrier related to the activation layer could be further optimized to enhance the NIR response. With the aid of this proposed activation recipe, the NIR performance of the InGaAs photocathode activated by the conventional Cs/O activation recipe could be effectively enhanced, helping to improve the detection efficiency of photodetectors used in laser, fluorescence, spectroscopy and other fields.

2. Experimental

2.1. Preparation of photocathode material

The structure of the InGaAs photocathode samples used in this work consisted of an $In_xGa_{1-x}As$ emission layer, a distributed Bragg reflector (DBR) layer and a GaAs substrate, as shown in Fig. 1. The DBR layer was composed of alternating GaAs/AlAs sublayers, with 10 pairs of these alternating layers. The DBR served to create a Fabry–Perot cavity to



Fig. 1. Structure diagram of the grown InGaAs photocathode with the DBR structure.

enhance the photocathode spectral response [16,26,27]. The $In_xGa_{1,y}As$ emission layer was composed of a 0.4 µm-thick graded region and a 0.7 µm-thick uniform region. The In composition of the graded region increased linearly from 0.05 to 0.2 along the electron transport direction, while the uniform region was 0.2. In the growth process, metalorganic compounds trimethylgallium (TMGa), trimethylalumium (TMAl) and trimethylindium (TMIn) were used as the III group sources, while arsine (AsH₃) was used as the V group source and diethylzinc (DEZn) was utilized as the p-type dopant source. In all epitaxial layers, the p-type doping concentration was 1×10^{19} cm⁻³. The growth temperatures of the AlAs/GaAs alternating layers and the InGaAs emission layer were 660 °C and 620 °C, respectively. The epitaxial growth rate was 1 µm/h and the V/III ratio was maintained at about 50. To ensure the quality of material growth, the InGaAs photocathode sample was characterized by X-ray diffraction (XRD), a scanning electron microscope (SEM), a sputter depth profile and a spectrophotometer, as presented in Fig. S1 of Supplementary Material. It was clear that the multilayer structure of the as-grown InGaAs photocathode sample met our specific design requirements for minimum reflectivity at 1064 nm.

Before activation, a two-step surface cleaning procedure, including a surface chemical cleaning process and a heat treatment process in vacuum, was performed to clean any surface contamination, such as carbon residues and oxides. First, the ultraviolet ozone cleaning, degreasing cleaning, and etching treatments in a mixed solution of hydrochloric acid and isopropanol were performed on the sample in sequence. Then, the sample was heated in the vacuum chamber at 600 °C to obtain an atomic-level cleaned surface.

2.2. Activation and re-activation

After surface cleaning treatment, the samples were activated by Cs/O and Cs/NF₃ in the newly-developed ultra-high vacuum experimental setup for photocathode preparation and characterization, as shown in Fig. 2 [12,13]. During the activation process, the improved co-deposition activation technique was utilized in an ultrahigh vacuum chamber with a base pressure of less than 1×10^{-7} Pa, with Cs flux provided by electrically heated dispensers based on Cs₂CrO₄, oxygen produced by electrically heated dispensers containing BaO₂ [13], and NF₃ was introduced by a variable leak valve. A white light tungsten halogen lamp source with illumination of 100 lx was used as the excitation light source to activate the cathode. When the photocurrent excited by the white light source no longer increased, preliminary activation was terminated, and the white light source was replaced by a 1064 nm laser to carry out re-activation. Additional spectral response results were measured after re-activation for comparison.

2.3. Spectral response and surface analysis

The spectral responses of photocathode samples are considered vital characteristics for evaluating the enhancement effect of the preparation technology. Therefore, the spectral response curves were measured insitu by an online measurement system after preliminary activation and re-activation [12,13]. During spectral response measurement, the light source was introduced onto the sample surface by an optical fiber connected to the monochromator, where the incident light wavelength was scanned from 400 to 1100 nm with a step wavelength of 5 nm. To investigate the change in surface state after preliminary activation and re-activation, in-situ photoelectron spectroscopy measurements, including X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS), were conducted by the PHI5000 VersaProbe II system. Through XPS and UPS analysis, the changes in surface element composition and work function were directly obtained. During XPS measurement, a monochromatic Al-K α X-ray source ($h\nu =$ 1486.6 eV) was used to excite the core electrons with an X-ray spot diameter of 100 μ m. For UPS measurement, a He I (hv = 21.2 eV) ultraviolet source was used to excite the valence electrons with an



Fig. 2. Schematic illustration of the ultra-high vacuum experimental setup for photocathode preparation and characterization.

ultraviolet spot diameter of 2 mm.

3. Results and discussion

3.1. Activation and re-activation

Cs/O activation required 10 alternating cycles of Cs/O activation to reach the maximum value of the photocurrent peak. In each Cs/O alternating activation cycle, the O source was opened when the photocurrent decreased to 80 % of the previous peak, while the Cs source was always kept open [28]. For Cs/NF₃ activation, the NF₃ source only had to be opened once when the photocurrent of the Cs first peak reached 80 %, and activation was terminated when the photocurrent stopped increasing. The photocurrent change curves during activation for the Cs/O and Cs/NF₃ activated InGaAs photocathode samples are shown in Fig. 3(a) and (b), respectively. The sample activated by Cs/NF₃ had a higher photocurrent and faster activation velocity than the one activated by Cs/O. Through this preliminary Cs/O or Cs/NF₃ activation, the surfaces of the InGaAs samples could reach the NEA state and become responsive to the incident light at 1064 nm.

After the preliminary activation, the tungsten-halogen lamp was replaced by a 1064 nm laser to implement re-activation. During the reactivation procedure, the Cs source no longer remained open, and the Cs and O sources, or NF3 source, were switched on alternately and intermittently. The photocurrent change curves for the re-activation process are shown in Fig. 3(c) and (d). In the re-activation experiments, we focused on whether the re-activation process could improve the spectral response, especially the response at 1064 nm. Hence, the photocurrent curves were normalized to determine the increase ratio of the photocurrent, and we investigated the relationship between the photocurrent after preliminary activation and after re-activation. Herein, the increase ratio of the photocurrent during re-activation could be clearly identified by normalization. As shown in Fig. 3(c) and (d), both photocurrents decreased after opening the Cs source in the initial stage. Subsequently, the supplied O or NF₃ made the photocurrents increase again, and the following operation of the activation source repeated the above alternation process. Of note, the photocurrent reached a new higher peak only after opening the O source for the first time, while the new peak after opening the NF3 source showed little difference from the initial photocurrent. When the Cs source was opened after the O or NF3 sources were closed, both photocurrents reached the maximum. To explain the variation trend of the photocurrent during re-activation, more experiments were implemented, as shown in Fig. 3(e)-(h), wherein, the reactivation recipe was adjusted in Fig. 3(e) and (f). In the re-activation process, the first step was modified to open the O or NF3 sources, while the other steps and the 1064 nm light source remained the same.

The photocurrents in Fig. 3(e) and (f) showed completely different trends after the first opening of the O and NF₃ sources. The photocurrent of the Cs/O activated sample increased directly, while the Cs/NF₃ activated sample decreased directly, by contrast. The subsequent f photocurrent trend was the same as the previous phenomenon, as shown in Fig. 3(c) and (d). Fig. 3(g) and (h) shows that the light source was replaced by a 405 nm laser to investigate the photocurrent trend during re-activation with the short-wavelength laser. Before re-activation, the power of the laser was adjusted low enough to avoid the heating effect and the subsequent decomposition of the activation layer. Unlike the trend shown in Fig. 3(e) and (f), the opening of the O and NF₃ sources caused both photocurrents to decrease, and the final photocurrents could not be improved by re-activation.

According to the abovementioned experimental results, we concluded that re-activation behavior using the NIR light near the threshold wavelength after Cs/O activation could improve the response at 1064 nm of the InGaAs photocathode samples. Combining the phenomenon shown in Fig. 3(c) and (d), we inferred that the activation layer was in a state of excess Cs atoms after Cs/O and Cs/NF3 activation. At this time, the further absorption of Cs atoms in the activation layer would lead to a decline in the photocurrent. However, the photocurrents shown in Fig. 3(e) and (f) exhibited different trends after opening the O and NF3 sources. Because of the stronger electronegativity of the F element, the Cs atoms could more easily form dipoles with the NF₃ molecules through charge transfer, which meant that the binding rate of Cs-NF₃ was faster than that of Cs-O [12,20,29]. During the activation process, stable Cs-NF₃ dipoles could form in the activation layer with the adsorbed NF_3 molecules in the surface activation layer [11]. After the preliminary activation process, the Cs atoms and NF3 molecules on the surface were in a saturated state. In this case, the additional Cs or NF3 introduced during the initial re-activation would have no effect on photocurrent increase, regardless of the incident light conditions, and instead would result in the destruction of the optimum Cs-NF3 dipole laver.

For the Cs/O activated sample, numerous Cs-O dipoles formed and reduced the work function [30,31]. In Cs/O activation, the oxygen molecules that adsorbed on the Cs-covered GaAs surface would dissociate into atoms [32], while, after preliminary activation under white light illumination, atom vacancies were present inside the activation layer with insufficient O atoms. This inadequate structural arrangement of the activation layer could decrease the response several times in the NIR range because the low-energy photoelectrons were more susceptible to the surface barrier. During re-activation, 405 nm or other ultraviolet-blue light could effectively increase the sticking coefficient of molecular oxygen on the GaAs surface [33,34]. Furthermore, the short-wavelength photons could excite numerous electron-hole pairs near the surface,



Fig. 3. (a) and (b) Cs/O and Cs/NF₃ preliminary activation processes using while light, (c) and (d) Cs/O and Cs/NF₃ re-activation processes, starting from the Cs supply using the 1064 nm laser, (e) and (f) Cs/O and Cs/NF₃ re-activation processes starting from the O supply and NF₃ supply, respectively, using the 1064 nm laser, (g) and (h) Cs/O and Cs/NF₃ re-activation processes starting from the O supply and NF₃ supply, respectively, using the 405 nm laser.

promoting the transformation of small-size oxygen atoms to large-size oxygen ions through electron transfer from the excited electron-hole pairs. Therefore, excess oxygen ions could not fully fill the vacancies inside the activation layer, resulting in insufficient optimization of the surface barrier. During the re-activation process under 1064 nm NIR light illumination, the low-energy electron-hole pairs were excited far from the surface without ionized oxygen atoms, and excess small-size oxygen atoms could pass through the topmost Cs atoms on the surface and fill the vacancies, thus optimizing the surface barrier.

To verify the repeatability of re-activation through interaction of the 1064 nm light with the Cs/O and Cs/NF₃ activated InGaAs surface, a total of 13 experiments were conducted, including 6 Cs/O re-activation experiments and 7 Cs/NF₃ re-activation experiments. The statistical experimental results are shown in Fig. 4. The results of multiple experiments demonstrated that under the action of the 1064 nm light on the surface, re-activation after Cs/NF₃ activation showed no significant effect on the response at 1064 nm, while re-activation after Cs/O activation did effectively improve the response by a factor of 2–6 at 1064 nm.

3.2. Spectral response

To investigate the influence of re-activation on the spectral response throughout the entire wavelength range, the in-situ spectral response curves were measured, as shown in Fig. 5. Periodic oscillation of the spectral response curves in the NIR range was due to the DBR layer inserted between the substrate and the emission layer. The maximum response occurred at exactly 1064 nm, which was consistent with the purpose of the structural design [16]. As shown in Fig. 5(a), the spectral response of the sample activated by Cs/NF3 showed no obvious improvement before and after re-activation, while the NIR response of the sample activated by Cs/O significantly increased with the 1064 nm laser. Meanwhile, re-activation illuminated by the 405 nm laser showed no enhancement effect on the spectral response of the entire band for the Cs/NF₃ activated sample, as shown in Fig. 5(b). For the Cs/O activated sample, re-activation under 405 nm laser illumination showed less enhancement effect of NIR response than under 1064 nm laser illumination. Through re-activation by interaction of the 405 nm light with the Cs/O activated surface, the response at 1064 nm only showed less than twofold improvement. Combined with the results in Fig. 3(c)-(h), we inferred that, the NIR response of the Cs-O activated InGaAs photocathode could be enhanced more significantly by the re-activation



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3.3. Surface analysis

The changes in surface element composition and work function of the InGaAs photocathode samples before and after re-activation were characterized by in-situ XPS and UPS analyses. The XPS spectra after preliminary activation and re-activation are shown in Fig. 6, and the calculated surface element percentages are listed in Table 1. We observed that the O1s signal of the Cs/NF3 activated sample was ascribed to an incompletely cleaned surface. Because the low N1s peak was submerged in the Auger peak of Ga LMM, the percentage of N1s was not given. After Cs/O and Cs/NF3 re-activation, the percentages of Cs3d and O1s or F1s, all increased. The Cs/O ratio of the Cs/O activated sample changed from 1.39 to 1.29, while the Cs/F ratio of the Cs/NF₃ activated sample changed from 3.21 to 2.87.We inferred from the difference between the Cs/O and Cs/F ratios that the oxygen molecules dissociated into smaller O atoms to form dipoles in the activation layer, while fluorine was still present on the surface in the form of large N-F molecules [11]. Furthermore, as shown in Fig. 6, the Cs3d binding energy of the Cs/NF₃ activated sample was about 0.4 eV larger than the Cs/ O activated sample, demonstrating the larger electronegativity of the F element.

We obtained the changes in secondary electron cut-off by measuring the UPS spectra, as shown in Fig. 7(a), and the calculated values of the work function Φ are listed in Table 1. We found that the work function of the Cs/NF₃ activated sample was lower than the Cs/O activated sample, which explained the difference in spectral response between the Cs/O and Cs/NF₃ activated samples shown in Fig. 5. re-activation after Cs/O activation slightly decreased the work function while re-activation after Cs/NF₃ activation had little effect on the work function. To describe the mechanism of interaction of the 1064 nm light with the activated surface, the energy band diagram of the InGaAs photocathode is presented in Fig. 7(b). According to Spicer's photoemission theory [35], the excited electrons must tunnel through the surface barrier to escape into the vacuum, with the tunneling probability positively related to photoelectron energy, and negatively related to the thickness and height of the surface barrier. Compared with elapsed time in activation, the short-term re-activation process will have a negligible effect on the thickness of the active layer. Hence, improvement in NIR response can be attributed to the decrease in work function with the drop in surface barrier height. Based on the double-dipole model [30,31], the surface barrier could be divided into two regions with different slopes, with region I generated by the InGaAs-Cs or InGaAs-O-Cs dipoles, and region II generated by the Cs-O or Cs-NF₃ dipoles. As shown in Fig. 7(b), E_{01} , E_{02} and E₀₃ represent the vacuum levels after Cs/O activation, Cs/O reactivation and Cs/NF3 activation, respectively. Following Cs/NF3 reactivation after Cs/NF3 activation, the vacuum level was almost unchanged, as there was no obvious increase in photocurrent and decrease in work function. Because the electrons excited by the low-energy NIR photons absorbed far from the surface would only tunnel through the lower surface barrier II to escape into the vacuum, the enhancement of NIR response by re-activation after Cs/O activation could be attributed to the minor optimization of surface barrier II [36]. Through Cs/O reactivation, the height of barrier II could be lowered to some extent. For the electrons excited near the surface by ultraviolet-blue light, most high-energy photoelectrons would tunnel through the surface barrier I to escape into the vacuum. Hence, the re-activation process had no significant enhancement effect on the spectral response in the visible range, as surface barrier I remained the same after re-activation. Hence, we determined that 1064 nm light in re-activation could not be replaced by ultraviolet-blue light, because the interaction of ultraviolet-blue light with the Cs/O activated surface could not effectively improve the structure of the activation layer, with minimal optimization of surface barrier II.

Fig. 4. The ratio of the final photocurrent to the initial photocurrent during reactivation using the 1064 nm laser, with results for 6 independent Cs/O reactivation experiments and 7 independent Cs/NF_3 re-activation experiments.

To better illustrate that re-activation changed the height of surface



Fig. 5. Measured spectral response curves of the InGaAs samples after activation and re-activation illuminated by (a) a 1064 nm laser and (b) a 405 nm laser.



Fig. 6. XPS spectra of (a) In3d, (b) Ga2p3, (c) As2p3, (d) Cs3d, (e) O1s and (f) F1s of the InGaAs samples after activation and re-activation.

barrier II rather than the height of surface barrier I, the relationship between the electron escape probability and the end height values of surface barriers I and II was simulated, as shown in Fig. 8. The electron escape probability of the InGaAs photocathode was obtained by solving the one-dimensional time-independent Schrödinger equation, based on the triangular surface barrier model using the Airy transfer matrix method [36–38]. In the simulations, the thickness values of surface barriers I and II were considered constant, with assumed values of 1 and 5 Å, respectively. Furthermore, the initial height of the surface barrier before activation was assumed to be 4.9 eV. As shown in Fig. 8(a), with a decrease in the end height of surface barrier I, i.e., $V_{\rm I}$, the electron escape probability increased in the entire energy range. Hence, if the reactivation improved the NIR response by lowering the height of $V_{\rm I}$, the response in the entire wavelength range would also be enhanced, which was inconsistent with the results in Fig. 5. However, the change in electron escape probability with different end heights of surface barrier II is shown in Fig. 8(b). We observed that a decrease in the end height of surface barrier II, i.e., $V_{\rm II}$, could only improve the escape probability of the lower-energy electrons. Meanwhile, the escape probability of the higher-energy electrons was almost insensitive to the change in $V_{\rm II}$. In

Table 1

Surface element percentages and work functions of the InGaAs samples after activation and re-activation.

	In3d (%)	Ga2p3 (%)	As2p3 (%)	Cs3d (%)	O1s (%)	F1s (%)	Ф (eV)
Cs/O activation Cs/O re- activation	5.11 5.37	35.34 32.48	17.79 16.70	24.26 25.61	17.50 19.84	0.00 0.00	1.173 1.145
Cs/NF_3 activation Cs/NF_3 re- activation	5.26 5.06	34.55 33.36	18.77 17.95	23.47 26.01	10.64 8.57	7.31 9.05	1.099 1.103

this case, we determined that the reason for the enhanced NIR response after re-activation came from the optimization of the surface barrier II.

4. Conclusions

In summary, an improved re-activation recipe using 1064 nm photoreaction after conventional Cs/O or Cs/NF₃ activation for an InGaAs photocathode was proposed to enhance the photoemission performance in the NIR range. Combining the photocurrent, spectral response and surface analysis results, we found that re-activation effectively improved the NIR response of the Cs/O activated InGaAs sample, and this process had no effect on the Cs/NF₃ activated sample.

Through interaction of 1064 nm light with the Cs/O activated surface, the composition and structure of the Cs–O dipole layer could be optimized by re-activation, and the height of surface barrier II further decreased, resulting in significant improvements in NIR response. Meanwhile, the ultraviolet-blue light in re-activation did not result in effective improvement of the NIR response, as it exhibited minimal optimization effect of the surface barrier. This proposed activation recipe provided an effective approach to overcome the technical challenge for improving NIR response, especially at 1064 nm of III-V group NIR photocathodes.

Data availability

Data will be made available on request.

CRediT authorship contribution statement

Ziheng Wang: Investigation, Writing – original draft. Yijun Zhang: Project administration, Supervision, Conceptualization, Writing – review & editing. Shiman Li: Data curation, Visualization. Song Tang: Validation. Junju Zhang: Formal analysis. Yunsheng Qian: Resources. Feng Shi: Project administration. Gangcheng Jiao: Software. Hongchang Cheng: Investigation, Methodology. Yugang Zeng: Investigation.



Fig. 7. (a) Secondary electron cut-off of UPS spectra for InGaAs samples after activation and re-activation, (b) energy band diagram of the InGaAs sample after different activation processes, where E_C is the conduction band minimum, E_V is the valence band maximum, E_F is the Fermi level, V_I and V_{II} are the end height of surface barriers I and II, respectively, and E_{01} , E_{02} and E_{03} represent the vacuum levels after Cs/O activation, Cs/O re-activation and Cs/NF₃ activation, respectively.



Fig. 8. (a) Theoretical electron escape probability with different values of $V_{\rm I}$, wherein the value of $V_{\rm II}$ was assumed as 0.9 eV, (b) theoretical electron escape probabilities with different values of $V_{\rm II}$, where the value of $V_{\rm II}$ was assumed as 1.2 eV. In all simulations, the assumed thickness values of surface barriers I and II were assumed as 1 and 5 Å, respectively, and the assumed initial height of the surface barrier before activation was 4.9 eV.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

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References

- L.A. Sordillo, S. Mamani, M. Sharonov, R.R. Alfano, The interaction of twisted Laguerre- Gaussian light with a GaAs photocathode to investigate photogenerated polarized electrons, Appl. Phys. Lett. 114 (2019), 041104.
- [2] S. Karkare, L. Boulet, L. Cultrera, B. Dunham, X.H. Liu, W. Schaff, I. Bazarov, Ultrabright and ultrafast III–V semiconductor photocathodes, Phys. Rev. Lett. 112 (2014), 097601.
- [3] K. Chrzanowski, Review of night vision technology, Opto-Electron. Rev. 21 (2013) 153–181.
- [4] J.W. Schwede, T. Sarmiento, V.K. Narasimhan, S.J. Rosenthal, D.C. Riley, F. Schmitt, I. Bargatin, K. Sahasrabuddhe, R.T. Howe, J.S. Harris, N.A. Melosh, Z. X. Shen, Photon- enhanced thermionic emission from heterostructures with low interface recombination, Nat. Commun. 4 (2013) 1576.
- [5] K.J. Smirnov, V.V. Davydov, S.F. Glagolev, N.S. Rodygina, N.V. Ivanova, Photocathodes for near infrared range devices based on InP/InGaAs heterostructures, J. Phys.: Conf. Series (2018, 1038,), 012102.
- [6] V. Sachno, A. Dolgyh, V. Loctionov, Image intensifier tube (l²) with 1.06-μm InGaAs- photocathode, Proc. SPIE 5834 (2005) 169–175.
- [7] M. Laurenzis, M.L. Manna, M. Buttafava, A. Tosi, J. Nam, M. Gupta, A. Velten, Advanced active imaging with single photon avalanche diodes, Proc. SPIE 10799 (2018) 1079903.
- [8] A. Restelli, J.C. Bienfang, A.L. Migdall, Single-photon detection efficiency up to 50% at 1310 nm with an InGaAs/InP avalanche diode gated at 1.25 GHz, Appl. Phys. Lett. 102 (2013), 141104.
- [9] J.J. Scheer, J.V. Laar, GaAs-Cs: a new type of photoemitter, Solid State Commun. 3 (1965) 189–193.
- [10] M. Kashima, Y. Itokawa, T. Kanai, D. Sato, A. Koizumi, H. Iijima, T. Nishitani, Y. Honda, H. Amano, T. Meguro, The photoemission characteristics of a NEA InGaN photocathode by simultaneously supplying Cs and O₂, Appl. Surf. Sci. 599 (2022), 153882.
- [11] Z. Liu, Y. Sun, S. Peterson, P. Pianetta, Photoemission study of Cs–NF₃ activated GaAs(100) negative electron affinity photocathodes, Appl. Phys. Lett. 92 (2008), 241107.
- [12] S. Li, Y.J. Zhang, K.M. Zhang, J.Z. Zhang, J.J. Zhan, F. Shi, G.C. Jiao, H.C. Cheng, C. Feng, Comparison of activation behavior of Cs-O and Cs-NF₃-adsorbed GaAs(1 0 0)-β₂(2 × 4) surface: from DFT simulation to experiment, J. Colloid Interface Sci. 613 (2022) 117–125.
- [13] Y.J. Zhang, Y.S. Qian, C. Feng, F. Shi, H.C. Cheng, J.J. Zou, J.Z. Zhang, X. Zhang, Improved activation technique for preparing high-efficiency GaAs photocathodes, Opt. Mater. Express 7 (2017) 3456–3465.

- [14] Y. Sun, R.E. Kirby, T. Maruyama, G.A. Mulhollan, J.C. Bierman, P. Pianetta, The surface activation layer of GaAs negative electron affinity photocathode activated by Cs, Li, and NF₃, Appl. Phys. Lett. 95 (2009), 174109.
- [15] Y. Sada, T. Meguro, Study on work function and corresponding electron emission during NEA activation of GaAs surfaces, Appl. Surf. Sci. 513 (2020), 145699.
- [16] Z.H. Wang, Y.J. Zhang, S.M. Li, S. Li, J.J. Zhan, Y.S. Qian, F. Shi, H.C. Cheng, G. C. Jiao, Y.G. Zeng, Enhancement of near-infrared response for GaAs-based photocathode with laminated graded-bandgap structure: theory and experiment, J. Mater. Res. Technol. 19 (2022) 2008–2017.
- [17] Y. Wang, F. Shi, W.W. Sha, J.J. Zhang, Reseach on structural design and preparation technology of InGaAsInP photocathodes, Proc. SPIE 10846 (2018) 108462M.
- [18] J. Biswas, E.D. Wang, M.J. Gaowei, W. Liu, O. Rahman, J.T. Sadowski, High quantum efficiency GaAs photocathodes activated with Cs, O₂, and Te, AIP Adv. 11 (2021), 025321.
- [19] J.K. Bae, L. Cultrera, P. DiGiacomo, I. Bazarov, Rugged spin-polarized electron sources based on negative electron affinity GaAs photocathode with robust Cs₂Te coating, Appl. Phys. Lett. 112 (2018), 154101.
- [20] N. Chanlek, J.D. Herbert, R.M. Jones, L.B. Jones, K.J. Middleman, B.L. Militsyn, J. High stability of negative electron affinity gallium arsenide photocathodes activated with Cs and NF₃, Phys. D: Appl. Phys. 48 (2015), 375102.
- [21] L.E. Bourree, D.R. Chasse, P.L.S. Thambana, R. Glosser, MBE-grown InGaAs photocathodes, Proc. SPIE 4796 (2003) 1–10.
- [22] M.C. Jin, X.L. Chen, G.H. Hao, B.K. Chang, H.C. Cheng, Research on quantum efficiency for reflection-mode InGaAs photocathodes with thin emission layer, Appl. Opt. 54 (2015) 8332–8338.
- [23] K.J. Smirnov, V.V. Davydov, Y.V. Batov, InP/InGaAs photocathode for hybrid SWIR photodetectors, J. Phys.: Conf. Series 1368 (2019), 022073.
- [24] L. Chen, Y. Shen, X.D. Yang, M.C. Jin, S.M. Liu, L.Z. Duan, S.Q. Xu, Research on Cs/ O activation process of near-infrared In_{0.53}Ga_{0.47}As photocathodes, J. Alloys Compd. 831 (2020), 154869.
- [25] M.Z. Yang, M.C. Jin, Photoemission of reflection-mode InGaAs photocathodes after Cs, O activation and recaesiations, Opt. Mater. 62 (2016) 499–504.
- [26] W. Liu, Y.Q. Chen, W.T. Lu, A. Moy, M. Poelker, M. Stutzman, S.K. Zhang, Recordlevel quantum efficiency from a high polarization strained GaAs/GaAsP superlattice photocathode with distributed Bragg reflector, Appl. Phys. Lett. 109 (2016), 252104.
- [27] Z.H. Wang, Y.J. Zhang, S.M. Li, S. Li, J.J. Zhan, Y.S. Qian, F. Shi, H.C. Cheng, G. C. Jiao, Y.G. Zeng, Temporal response of laminated graded-bandgap GaAs-based photocathode with distributed Bragg reflection structure: model and simulation, Chin. Phys. B 31 (2022), 098505.
- [28] Y.J. Zhang, K.M. Zhang, S.M. Li, S. Li, Y.S. Qian, F. Shi, G.C. Jiao, Z. Miao, Y. L. Guo, Y.G. Zeng, Effect of excessive Cs and O on activation of GaAs(100) surface: from experiment to theory, J. Appl. Phys. 128 (2020), 173103.
- [29] S. Pastuszka, M. Hoppe, D. Kratzmann, D. Schwalm, A. Wolf, A.S. Jaroshevich, S. N. Kosolobov, D.A. Orlov, A.S. Terekhov, Preparation and performance of transmission-mode GaAs photocathodes as sources for cold dc electron beams, J. Appl. Phys. 88 (2000) 6788–6799.
- [30] C.Y. Su, W.E. Spicer, I. Lindau, Photoelectron spectroscopic determination of the structure of (Cs, O) activated GaAs (110) surfaces, J. Appl. Phys. 54 (1983) 1413–1422.
- [31] D.G. Fisher, R.E. Enstrom, J.S. Escher, B.F. Williams, Photoelectron surface escape probability of (Ga, In)As : Cs–O in the 0.9 to 1.6 μm range, J. Appl. Phys. 43 (1972) 3815–3823.
- [32] K.V. Toropetsky, O.E. Tereshchenko, A.S. Terekhov, Energy threshold of Csinduced chemisorption of oxygen on a GaAs(Cs, O) surface, JETP Lett. 88 (2008) 520–523.
- $[{\bf 33}]$ T. Guo, The adsorption of Cs and O2 on a clean GaAs(110) surface under light illumination, J. Vac. Sci. Technol. A 7 (1989) 1563–1567.
- [34] K.A. Bertness, P.H. Mahowald, C.E. McCants, A.K. Wahi, T. Kendelewicz, I. Lindau, W.E. Spicer, Photoenhancement mechanism for oxygen chemisorption on GaAs (110) using visible light, Appl. Phys. A 47 (1988) 219–228.
- [35] W.E. Spicer, A. Herrera-Gómez, Modern theory and applications of photocathodes, Proc. SPIE 1993 (2022) 18–33.
- [36] J.J. Zou, B.K. Chang, Z. Yang, Y.J. Zhang, J.L. Qiao, Evolution of surface potential barrier for negative-electron-affinity GaAs photocathodes, J. Appl. Phys. 105 (2009), 013714.
- [37] K.L. Jensen, D. Finkenstadt, D.A. Shiffler, A. Shabaev, S.G. Lambrakos, N. A. Moody, J.J. Petillo, Analytical models of transmission probabilities for electron sources, J. Appl. Phys. 123 (2018), 065301.
- [38] W. Liu, E. Wang, Monte Carlo modeling of thin GaAs photocathodes, J. Appl. Phys. 126 (2019), 075706.