The degradation of quantum efficiency in negative electron affinity GaAs photocathodes under gas exposure

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Abstract

The influence of O$_2$, CO$_2$, CO, N$_2$, H$_2$ and CH$_4$ on the stability of the quantum efficiency (QE) of a negative electron affinity gallium arsenide (GaAs) photocathode activated with caesium (Cs) and oxygen (O) has been demonstrated for the first time under an extremely high vacuum condition, a base pressure of $1.5 	imes 10^{-11}$ mbar, where the influence of the background gas is minimized. It was found that exposure of a GaAs photocathode to N$_2$, H$_2$ and CH$_4$ does not affect the QE, whereas exposure to O$_2$, CO$_2$ and CO leads to a substantial reduction in photocathode QE. It was also found that the QE of photocathodes which have been degraded under O$_2$ exposure can be recovered to 95% of their initial QE level by the re-caesiation process, while those which have been degraded under exposure to CO and CO$_2$ can only be partly restored to 60–70% of their initial QE levels.

(Some figures may appear in colour only in the online journal)

1. Introduction

Gallium arsenide (GaAs) and GaAs-based photocathodes have in recent years been widely used as high average current and polarized electron sources in several accelerators and light sources, such as the ALICE ERL at Daresbury Laboratory (Accelerators and Lasers in Combined Experiments, an Energy Recovery Linac), the proposed International Linear Collider (ILC), and the Continuous Electron Beam Accelerator Facility (CEBAF) at the Thomas Jefferson National Accelerator Facility (Jefferson Laboratory) [1–8]. Their applications have also been found in photomultiplier tubes, high-performance image intensifiers, low-energy electron microscopes (LEEMs) and spin low-energy electron microscopy (SPLEEM) [9–13]. GaAs photocathodes have become very attractive since they can be activated to a negative electron affinity (NEA) state by depositing caesium (Cs) and oxidant layers onto their highly p-doped surfaces [13–17]. In this state, the vacuum energy level of the photocathode is located below the conduction band minimum. Consequently, electrons with a range of energies excited into the photocathode conduction band can easily escape to the vacuum. This allows the GaAs photocathodes to have a high quantum efficiency (QE), i.e. the number of the emitted electrons per incident photon, when illuminated with all photon energies greater than their band gap. A recorded QE of 35% measured at 635 nm has been reported [7]. The NEA GaAs photocathodes also offer several advantages compared to other cathodes, including high spin polarization in strained layer structures and very small thermal emittance at certain conditions [6–8, 12, 17–20].

The main limitation of NEA GaAs photocathodes is their short lifetime. Their QE deteriorates fairly quickly within an ultra-high vacuum (UHV) system with a pressure of $10^{-10}$ mbar even if they are not operated under high voltage.
or/and high power illumination [5, 12, 21, 22]. The QE dark lifetimes, i.e. the 1/e lifetime of the QE without electron emission or cathode high voltage applied, ranges from some tens of hours to several hundred hours at best, these have been reported from several laboratories [5, 23–25]. The predominant mechanism responsible for this degradation is the contamination of the cathode surface by residual gases inside the vacuum system [12, 21, 22, 26–29]. To suppress or eliminate this issue, a clear understanding of the effect of different gas species on the QE of the photocathodes is required. Previously, Wada et al. have attempted to study the influence of CO, CO2 and H2O on the stability of the GaAs photocathode [30]. They exposed the GaAs photocathode to those gas species typically found in a vacuum system, and observed that exposures to CO2 and H2O lead to a drastic decay in the QE, while an exposure to CO does not affect the QE of the photocathode. However, there is some ambiguity about these experimental results, since the experiment was carried out within a vacuum system which has a base pressure in the low-to-middle 10⁻¹⁰ Torr range (1 Torr = 1.333 mbar). At this level of vacuum, the QE of the photocathode gradually degrades due to the residual gases inside the vacuum chamber without any additional gas exposure. Therefore, it would not have been possible to observe the small effect from gas species such as CO.

In this article, we present our recent experimental study on the influence of O2, CO2, CO, N2, H2 and CH4 on the stability of the QE of a NEA GaAs photocathode activated with Cs and O. We have exposed the photocathode to a small quantity (<0.25 Langmuirs) of these gas species under extremely high vacuum (XHV) conditions, a base pressure of 1.5 x 10⁻¹¹ mbar. To the best of our knowledge, this is the first demonstration of the influence of those gas species under this level of vacuum, where the influence of the background gas is minimized. In this experiment, we were able to demonstrate clearly the influence of these gases on the photocathode and our experimental results contradict the current assumption that CO has no effect on the QE of the NEA GaAs photocathode [30, 31].

Another interesting result found in this experiment is the possibility to restore photocathode QE after degradation. It is well known that the QE of NEA GaAs photocathodes can be recovered by depositing Cs onto a degraded surface, the so-called re-caesiation [23, 27, 30]. This technique is widely used to recover the QE and prolong the operational lifetime of an NEA GaAs photocathode. However, there have been no reports on the influence of exposure to O2, CO2, CO, N2, H2 and CH4, and the QE achievable on the subsequent re-caesiation of a photocathode. In this study, we found that the QE of the photocathodes which have been degraded under the O2 exposure can be recovered to 95% of its initial value by the re-caesiation process, while those that have been degraded under CO and CO2 exposures can be partly restored to only 60–70% of their initial values.

2. Experiments

The experiments were carried out using the photocathode preparation facility (PPF) for the ALICE accelerator at Daresbury Laboratory. The design of the ALICE PPF has been described in detail elsewhere [1]. The facility consists of three vacuum chambers: the UHV loading chamber, the UHV cleaning chamber (equipped with an atomic hydrogen source) and the XHV preparation chamber, as shown schematically in figure 1. These chambers are isolated by gate valves, and photocathodes are transferred between them using a magnetic linear transfer arm. The preparation chamber houses a rotary carousel with the capacity to hold up to six photocathodes. This carousel can be rotated into different positions for the thermal cleaning and activation of photocathodes with Cs and O2. The preparation chamber has a base pressure of 7.5 x 10⁻¹² mbar,
with the partial pressures of O₂, water vapour and CO₂ less than 10⁻¹⁵ mbar.

The photocathodes used in this experiment were a GaAs/AlGaAs hetero-structure bound to a glass base, designed by the Institute of Semiconductor Physics (ISP), Novosibirsk. The hetero-structure consists of two layers: the active layer and the buffer layer. The active electron-emitting layer is a GaAs(1 0 0) with a thickness of 2.4 μm, grown by a metal organic chemical vapour deposition (MOCVD) technique and heavily doped with Zn at a hole concentration of (1–3) × 10¹⁹ cm⁻³ to create a p-type semiconductor. The buffer layer is Al₀.₅Ga₀.₅As with a thickness of approximately 0.02 μm, doped p-type with Zn at a hole concentration of 5 × 10¹⁷ cm⁻³.

Prior to activation to the NEA state, each photocathode was chemically cleaned in a glove-box filled with dry, pure nitrogen in order to avoid surface oxidation and uncontrolled contamination. The chemical treatment involved etching in a solution of HCl (in IPA 1 : 10 by volume) for one minute, rinsing twice in pure IPA for one minute, and then blown dry with dry gaseous nitrogen. Then the photocathode was installed in a holder and transferred to the loading chamber using a nitrogen-filled air-tight transfer vessel without exposure to the atmosphere. After pumping-down the loading chamber to UHV, the photocathode was transferred to the cleaning chamber and radiatively heated from the back-side using a halogen bulb to a temperature of approximately 300 °C for 30 min to remove any chemical residue which may contaminate the preparation chamber. As soon as the process was complete and when the pressure had recovered to the 10⁻³ mbar range, the photocathode was transferred and placed onto the carousel in the XHV preparation chamber. The photocathode was then radiatively heated from the back-side using a halogen bulb to a temperature of approximately 450 °C for 30 min. The photocathode surface has to be atomically clean prior to activation through deposition of Cs and O onto its surface. Also, the cleaning temperature has to be kept as low temperature as possible to avoid damaging the photocathode. Etching the photocathode in the HCl solution under an N₂ atmosphere as described by Tereshchenko et al removes most oxides from the surface, leaving some As oxide layers which act as the protective surface layer [32]. Final heating to 450 °C to remove these As oxides leaves a clean GaAs surface. To remove other oxides such as potassium oxide, the photocathode has to be heated to above 600 °C. However, heating the photocathode to 600 °C risks surface damage since it is close to the congruent evaporation temperature for GaAs(1 0 0) of between 657 to 663 °C [33]. Exposure to high temperature also significantly increases surface roughness, which in turn increases the thermal emittance of the photocathode. Therefore, we avoid that high temperature and also expect very low level of potassium oxide found on the surface. It is known that a heavily doped p-type GaAs(1 0 0) photocathode adopts a Ga-terminated 4 × 2 surface re-construction when heat-cleaned to 520 °C [34], with surface roughness less than 1 nm over a 500 nm range [35]. While our heat-cleaning process involves a lower annealing temperature, XPS measurements have shown the absence of carbon and oxygen peaks, so we assume a clean surface with a morphology described by Benemanskaya et al [34, 35].

Once the photocathode had cooled to room temperature, approximately 3 h, the NEA activation process was performed using a yo-yo technique by the co-deposition of Cs and O onto the surface [7]. Cs was evaporated from commercial Alkali Metal Dispensers (AMDs from SAES) which were operated at 2.5 A dc. O₂ was admitted into the chamber alternately through a piezo-electric leak valve. The rate of O₂ exposure was controlled so that the pressure of the chamber did not exceed 5 × 10⁻¹⁰ mbar. The cathode was illuminated from the front side to drive photocurrent emission using a low-power 635 nm laser which delivered 0.858 ± 0.005 mW, measured over a 15 hour period. This light passed through a beamsplitter with 62% transmission, and then a neutral density filter (OD = 3.0) with 0.137% measured transmission. The laser was modulated at frequency of 3 kHz using the clock signal from a lock-in amplifier, and the photocurrent measurement extracted using phase-sensitive detection. The photocathode was negatively biased to 18 V with respect to the walls of the chamber using a battery box. The QE is defined as the ratio of the number of electrons emitted per incident (rather than absorbed) photon, and is calculated from the photocurrent, I(A), using the following equation [36]:

\[
\text{QE}(\%) = \frac{Ihc}{\lambda e P} \times 100\%.
\]

where λ is the laser wavelength (m), h is Plank’s constant (6.626 × 10⁻³⁴ J s), e is the electron charge (1.6 × 10⁻¹⁹ C), c is the velocity of light (2.998 × 10⁸ m s⁻¹), and P is drive laser power (W). In this experiment, a QE of between 12–16% and 1/e dark lifetime longer than 6500 h was routinely achieved after the activation in a reproducible way. A plot of the evolution of QE and pressure of the preparation chamber during the activation is shown in figure 2 and a QE after the activation is shown in figure 3.

The QE degradation of the NEA GaAs photocathodes during gas exposure was studied after the activation and the total pressure of the preparation chamber had recovered to approximately 1.5 × 10⁻¹¹ mbar. At this point, the
showing a dark lifetime in excess of 6500 h. The test gas was then carefully injected into the chamber through the piezo-electric leak valve. During the injection, the total pressure of the chamber was increased slowly, and maintained at a constant value of approximately 1.5 × 10^{-10} mbar. The pressure was measured using an extractor gauge (Ionivac IE 514 from Oerlikon Leybold Vacuum), while the purity of the injected test gas was monitored using the RGA, not permitting a background partial pressure more than 5% for any residual oxidizer present relative to the gas under test. Gas exposures were quoted in Langmuirs (L), where 1 L = 1.33 × 10^{-6} mbar·s, and calculated as the integral of the total pressure rise over the injection time. The uncertainty of the pressure measurement and the gas exposure was 5%, based on the uncertainty of the vacuum gauge. Six gas species were tested in this work, selected because they are the main residual gases found in the vacuum system of a typical photoinjector: H₂, CH₄, N₂, O₂, CO₂ and CO. These gas species were sourced from Messer laboratory-grade research gas canisters (99.998% pure by volume). For all of the experiments, only freshly activated photocathodes were used to avoid the historic effects of the photocathode QE documented earlier.

3. Experimental results

The QE evolution of the photocathodes during exposures to H₂, CH₄, N₂, CO, CO₂ and O₂ at a total pressure of 1.5 × 10^{-10} mbar for approximately 30 min are shown in figure 4. No changes in QE were observed during the H₂, CH₄ and N₂ exposures. This clearly demonstrates that H₂, CH₄ and N₂ have no influence on the stability of the NEA GaAs photocathode. On the other hand, significant degradation of QE was clearly observed during the CO₂ and O₂ exposures, with only a slight degradation during CO exposure.

Comparisons of the QE degradation of the photocathodes during these exposures in Langmuir units are shown in figure 5. It is observed that O₂ shows the strongest effect since it can degrade the QE to 10% of the initial value after an exposure of only 0.025 L. Interestingly, this level of O₂ exposure is in agreement with the result reported by Pastuszka et al which stated approximately 0.021 L [27]. CO₂ has a severe effect on the photocathode, resulting in the degradation of the QE to 10% of the initial value after an exposure of approximately 0.08 L. The most interesting result is the observed effect of the CO exposure on the QE of the photocathode. In this experiment, a CO exposure of only 0.125 L reduces the QE of the photocathode to 90% of its initial value. This result is in stark contrast to the previous study by Wada et al in which CO has no influence on the QE of the NEA GaAs photocathode (even after 100 L of exposure) [30]. We explain this difference by the fact that the previous experiment was performed in a vacuum chamber which has a base pressure in the low-to-middle 10^{-10} Torr range. At that level of pressure, the QE of the photocathode will be degraded gradually due to the residual gases inside the chamber without any additional gas exposure. Therefore, it would not have been possible to observe the small degradation during CO exposure in that experiment.

To investigate the effect of the gas exposures after degradation, re-caesiation was carried out by adding Cs onto the photocathodes after the degradation. The evolution of the QE during the application of Cs to the degraded GaAs photocathodes after O₂, CO₂ and CO exposures are shown in figure 6. Since the QE degradation during CO exposure is very slow, the photocathode was thus exposed to CO up to a pressure of 2 × 10^{-10} mbar in this experiment. From the experimental results, it is seen that after adding Cs onto the surface, the QE can be restored to 95% of the initial value in the case of O₂ exposure, whereas the QE can recover only to between 60% and 70% of the initial value in the case of CO₂ and CO exposures, respectively.

4. Discussion

The results described above clearly show the influence of H₂, CH₄, N₂, CO, CO₂ and O₂ on the QE of a NEA GaAs photocathode. The NEA GaAs photocathode degrades predominantly due to the adsorption of oxygen or oxygen-containing gas species, CO₂ and CO.

Photoemission from NEA GaAs photocathodes has been described by Spicer as a three-step process [14]: the photoabsorption, the transportation of the photoexcited electrons to the vacuum interface, and the emission of electrons into the vacuum. In the final step, the photoexcited electrons have to overcome an interfacial barrier between the activating layer and vacuum. It can be expected that the lowering of this barrier will increase the electron surface escape probability and thus the QE of photocathodes. Our experimental results appear to support the existence of this interfacial barrier and can be explained by using the double-dipole model [11, 15, 37]. A schematic energy band diagram of the double-dipole model is shown in figure 7. In this model, the surface potential barrier consists of two regions with different slopes: (1) and (2) in figure 7. The barrier (1) is created by a GaAs/Cs dipole at the GaAs surface, while the barrier (2) is created by a Cs/O dipole in the activation layer. The precise nature of the adsorption of Cs and O onto GaAs is not well understood, despite much investigation. During initial adsorption, Cs does not diffuse into the bulk or form chemical compounds with the semiconductor components, but forms an amorphous layer.
Figure 4. Evolution of the photocathode QE during exposures to H₂, CH₄, N₂, CO, CO₂ and O₂ at a pressure of 1.5 × 10⁻¹⁰ mbar.

on the (100) surface, ultimately reaching a thickness of 1 monolayer [38]. So far, three preferential adsorption sites have been proposed for Cs adsorption, these being the arsenic dimer bridge D site, the gallium dangling bond site, and the arsenic T3 trench site [39]. Low binding energy bonds Cs–As and Cs–Ga and the dipole between them are both proposed for this system [40], but not yet proved experimentally.

Our experimental results show that exposure to O₂ and O-containing species leads to a reduction in the QE of the photocathodes. This is because when oxygen bonds with the NEA GaAs photocathode surface (as atoms, molecular oxygen, or part of some other species), they accept electrons from Cs atoms and are chemisorbed inside the activation layer [11]. We propose that this changes the chemistry of the Cs/O layers and weakens the dipole in the activation layer region. As a result, the height of the surface potential barrier in region (2) and the vacuum level increase. Moreover, the adsorbed oxygen can effectively penetrate the activation layer and directly interact with the GaAs surface, thus forming surface oxides, as has been reported [11]. Consequently, the width of barrier region (1) increases and the barrier profile for a degraded photocathode will evolve as shown by the dashed line in figure 7.

After the degradation, the dipole in region (2) can be restored by the re-caesiation process. The additional Cs atoms on the degraded photocathode surface strengthen the Cs/O dipole in the activation layer, so that the vacuum level will be reduced again as shown by the dotted line in figure 7. However, the re-caesiation process has little influence on the barrier region (1). In other words, the surface oxides formed during the degradation process cannot be removed by the re-caesiation process alone. Therefore, the QE of the degraded photocathode cannot be fully recovered to its initial value in this way, only 95% of the initial value after the O₂ exposure in this experiment.

In these experiments, we also found that O₂ has the strongest effect on the QE of the photocathode because it directly reacts with the Cs/O layer and weakens the dipole in the activation layer. Conversely, CO was found to have little
effect on the QE. Wada et al suggested that this may be related to the fact that CO does not react with pure Cs clusters [30]. Moreover, given that CO has a high dissociation energy of 11.10 eV [30], we believe that CO bonds with the activation layer through molecular adsorption rather than dissociating to create carbon and free oxygen. An earlier study [41, 42] on the absorption of CO on GaAs suggests that CO adsorbs molecularly on the surface rather than dissociating into C and O. However, the adsorption of CO through covalent bonding perturbs the local electronic structure, subtly changing the chemistry of the Cs/O activated surface. This weakens the dipole in the activated layer, resulting in the low degradation rate observed. For the case of CO2, because the dissociation energy of CO2 into CO and O is low, 5.44 eV [30], it is hence expected to dissociate and provide oxygen and CO to the activation layer. Therefore, the QE degrades much faster than when exposed to CO alone, but slower than when exposed to O2. Our experimental results on the re-caesiation after CO and CO2 exposures are also in good agreement with the model. CO is adsorbed molecularly, penetrates the activation layer and directly interacts with the GaAs substrate. As a result, the restored QE of only 70% was obtained after the re-caesiation process. The lower restored QE in the case of CO exposure suggests that the interaction of CO causes a greater increase in the width of barrier region (1) compared to oxygen. In the case of CO2 exposure, CO2 is dissociated to CO and O which both can penetrate the activation layer and interact with the GaAs substrate. Therefore, the restored QE of only 60% was obtained after the re-caesiation process.

These experimental results have aided the understanding of the degradation mechanism for NEA GaAs photocathodes. However, a more complete understanding of the nature of degradation requires further studies of the electronic structure of contaminated NEA GaAs photocathodes under XHV conditions which include the use of additional oxygen-containing contaminants such as NO and NO2, and these are now under way. While it is beyond the present scope of this work, future studies should be underpinned by the development of a quantitative model linking QE to oxygen adsorption.

5. Conclusions

We have demonstrated the influence of H2, CH4, N2, CO, CO2 and O2 on the QE of a NEA GaAs photocathode at extremely low residual gas pressure. It was found that H2, CH4 and N2 have no effect on the QE of the photocathodes, whilst substantial reductions of the QE have been observed during the exposures to O2, CO2 and CO. It was also found that the QE of the photocathodes which have been degraded under the O3 exposure can be recovered to 95% of its initial value by the
re-caesiation process, while those have been degraded under CO and CO₂ exposures can be partly restored to only 60–70% of their initial values.

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