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# Non-monotonic behaviour in the mean transverse energy of electrons emitted from a reflection-mode p-GaAs(Cs,O) photocathode during its QE degradation through oxygen exposure

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#### Abstract

We present a study on the evolution of both transverse and longitudinal energy distributions due to oxygen–induced quantum efficiency degradation in the electrons emitted from a reflection–mode p-GaAs(Cs,O)-photocathode over an effective electron affinity range of  $\chi^* \sim -0.1 \text{ eV}$  to  $\chi^* \sim +0.3 \text{ eV}$  under illumination wavelengths  $\lambda = 808$ , 635 and 532 nm. For  $\lambda = 635$  and 532 nm, we found that the mean transverse energy of electrons emitted as the photocathode effective electron affinity increased has a non–monotonic character. Our proposed fitting model links the measured response to changes in the relative number of thermalised and non–thermalised electrons in the total emitted photocurrent.

Keywords: photocathode, electron source, energy spread, degradation, QE, MTE, MLE

#### 1. Introduction

Reflection–mode GaAs photocathodes are widely used as electron sources in particle accelerators [1]. Their mean transverse energy (MTE) and mean longitudinal energy (MLE) are critically important performance indicators closely related to the photocathode intrinsic emittance which is affected by several factors, including the electron affinity ( $\chi^*$ )

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and the illumination wavelength ( $\lambda$ ), both of which themselves influence the quantum efficiency (QE).

While there is an expanding range of literature presenting MTE measurement on a variety of photocathode electron sources commonly used in particle accelerators, there is little information concerning the *evolution* of MTE during photocathode degradation. Given that photocathode degradation is unavoidable during the operation of an accelerator electron source, an investigation of MTE behaviour, its evolution during photocathode degradation and the mechanisms underpinning this degradation is of key interest to the accelerator community.

To learn more about this process, we studied a GaAs photocathode activated to a state of negative electron affinity  $(\chi^* < 0)$  and then subjected to progressive degradation through precisely controlled exposure to oxygen. In so doing, we were able to simulate the typical degradation experienced by a GaAs photocathode during accelerator operations and to monitor the consequent changes in electron energy spread. By studying this behaviour at various wavelengths some way from the photoemission threshold, we found that the transverse energy component is a non-monotonic function dependent on the electron affinity which changes during the degradation process.

#### 2. Experiment description

Experiments were performed on a p<sup>+</sup>–GaAs(100) photocathode heterostructure grown by MOCVD. It consisted of a  $2.4 \,\mu\text{m}$  p–GaAs active top layer, Zn doped at a level of  $1 \times 10^{19}$  cm<sup>-3</sup>. The active layer was separated from an n–GaAs(100) substrate by a  $0.3 \,\mu\text{m}$  Al<sub>0.55</sub>Ga<sub>0.45</sub>As buffer layer, and the heterostructure was indium soldered to a molybdenum 'puck'. Prior to first use the oxide layer was removed from the active top layer using a solution of HCl in isopropanol under an inert nitrogen atmosphere [2], and the photocathode puck then transferred into vacuum using a suitcase without subsequent exposure to the atmosphere. Following an in–vacuum heat clean, it was activated in our III–V Photocathode Preparation Facility [3] (PPF) following accepted procedures [2, 4], achieving a quantum efficiency around 10.0% at 635 nm.

The photocathode was then loaded into the Transverse Energy Spread Spectrometer (TESS) which combines a photocathode holder and source electrode with a detector comprising multiple electrostatic grids, a microchannel plate (MCP) electron multiplier coupled to a phosphor screen for imaging the photoemission footprint and a high–performance CCD camera to capture the image. TESS can be used to measure both longitudinal and transverse photoelectron energy distribution curves (LEDC and TEDC respectively) depending on how the instrument is configured. A full description of the system including a schematic diagram of the experimental arrangement can be found in our previous publications [5, 6].

The photocathode was illuminated with laser light at various wavelengths and measurements made of the longitudinal and transverse energy distributions. During these measurements, the photocathode QE was progressively degraded by exposure to oxygen admitted to the vacuum chamber via a piezo–electric leak valve at a very low partial pressure (typically  $5 \times 10^{-11}$  mbar). Having noted the base pressure at the start of each experiment prior to oxygen admission, the chamber pressure was monitored continuously during the data acquisition process to record the integrated oxygen exposure.

The experiment was repeated three times with a different laser illumination wavelength ( $\lambda$ ) used for each series. In chronological order, a series of measurements was performed with  $\lambda = 532$  nm, then with  $\lambda = 635$  nm and finally with  $\lambda = 808$  nm. The photocathode was transferred from the TESS back into the PPF after each series and thermally cleaned at a temperature of about 550 °C. Once cool, the photocathode was reactivated and transferred back into the TESS for the next series of measurements. The integrated oxygen exposure levels during photocathode activations were between 0.5 and 0.7 Langmuirs (L) which significantly exceeded the oxygen exposures subsequently applied in the degradation of the photocathode in the TESS which was typically around 0.08 L.

The momentum **p** of an electron emitted from a photocathode has two components: the vector parallel to the surface normal which is the *longitudinal* momentum ( $p_{lon}$ ), and the vector parallel to the surface which is the *transverse* momentum ( $p_{tr}$ ). Consequently, total electron energy ( $\varepsilon$ ) can be represented by the sum of the longitudinal and transverse components where *m* is the electron mass:

$$\varepsilon = \left| \frac{\mathbf{p}^2}{2 \text{ m}} \right| = \frac{p_{\text{lon}}}{2 \text{ m}} + \frac{p_{\text{tr}}}{2 \text{ m}} = \varepsilon_{\text{lon}} + \varepsilon_{\text{tr}}.$$
 (1)

During TEDC measurements, the photocathode was illuminated at extremely low intensity with a tightly–focussed laser beam typically 100  $\mu$ m FWHM diameter, and its electron emission footprint recorded at some known distance from the photocathode surface. Assuming a vanishingly–small source emission spot size, and knowing both the drift distance travelled by the photoelectrons and the effective voltage through which the electrons have been accelerated (this defines their time–of–flight), the transverse electron energy distribution required to generate the observed emission footprint can be determined.

Data were taken with the photocathode biased at -30 V and the detector grids and MCP front plate all held at +30 V. This resulted in an overall accelerating potential of  $(60 - U_{cpd})$  V which defines the electron time-of-flight, with  $U_{cpd}$  being the contact potential difference between the photocathode and the detector. The distance between the photocathode and first grid was equal to  $23 \pm 0.5$  mm. The electron emission distribution incident on the MCP front plate was amplified by the MCP with the gain controlled by the voltage applied to the MCP back plate which was +900 V during these experiments. The amplified photoemission footprint was then accelerated onto a phosphor screen held at +3.5 kV and recorded over a period of typically 60 s using a cooled CCD camera. Details of the measurement process and the calculation to extract  $N_e(\varepsilon_{tr})$  can be found in our previous work [7].

During LEDC measurements, the TESS instrument has to be reconfigured. The grids and MCP front plate are used as a retarding–field analyser to capture electrons photoemitted by the cathode and transport them to the MCP front plate which is used as a charge collector. Photoelectrons are accelerated across the gap between the cathode and the first grid, and the transverse footprint of the photoemitted distribution is first verified to ensure that it does not exceed the working diameter of the detector assembly. To satisfy this condition, we found in our setup experiments that voltages for the first, second and third grids needed to be +64 V, +47.4 Vand +29.4 V respectively. The MCP front plate was grounded through a lock–in amplifier configured to measure the photocurrent,  $I_{\text{ph}}$ . As such, only electrons whose longitudinal energy exceeded ( $U_{\text{cpd}} - U_{\text{ph}}$ ) eV could make a contribution to the photocurrent measured by the lock–in amplifier, where  $U_{\rm ph}$  is the bias voltage applied to photocathode. The first differential of the  $I_{\rm ph}(U_{\rm ph})$  curve yields the longitudinal energy distribution curve from which  $N_e(\varepsilon_{\rm lon})$  can be extracted. Details of the  $N_e(\varepsilon_{\rm lon})$  measurement process can be found in our previous work [7].

#### 3. Results and discussion

Figure 1 shows the band structure around the  $\Gamma$ -valley for GaAs. In order to calculate the photoelectron energy in GaAs at the instant of excitation, it is necessary to take into account the degeneracy of the valence band near the  $\Gamma$ -point which arises from three sub-bands: the heavy hole sub-band; the light hole sub-band and the split-off sub-band. If the illumination photon energy is sufficiently high, then electron transitions are possible from all of these valence sub-bands to the conduction band. We used a k.p model from Pfeffer and Zawadzki [8] to calculate the conduction and valence band energies, and from this we defined the initial photoelectron energies. Our results for the (100) direction are shown in table 1 where  $\varepsilon_{e-hh}$  denotes the initial energy of the photoelectron upon transition from the heavy hole sub-band,  $\varepsilon_{e-lh}$ from the light hole sub–band, and  $\varepsilon_{e-so}$  from the split–off sub– band. Table 1 shows that under illumination at a wavelength of 808 nm, only transitions from the heavy-and light-hole sub-bands are possible, but that transitions from all three sub-bands are possible when illuminated at wavelengths of 635 and 532 nm.

Following generation in the photocathode bulk, photoelectrons lose their excess energy while establishing thermal equilibrium with the bulk material, a process known as *thermalisation* which is depicted in figure 1. In a reflection–mode photocathode, photoelectrons are generated near the emitting surface so a considerable fraction of the excited electrons can escape to vacuum before achieving thermal equilibrium. Consequently all emitted electrons can be roughly divided into two groups: *thermalised* where they have reached thermal equilibrium with the bulk before emission into the vacuum and *non–thermalised* (or '*hot*') where they have not lost all of their excess energy before emission into the vacuum [9].

Figures 2–7 show our measurements of the longitudinal  $N_e(\varepsilon_{1\text{on}})$  and transverse  $N_e(\varepsilon_{tr})$  energy distributions for the GaAs(Cs,O)-photocathode at different stages of degradation. Figures 2 and 3 show our measurements with the photocathode under illumination at  $\lambda = 808$  nm, figures 4 and 5 with  $\lambda = 635$  nm, and figures 6 and 7 with  $\lambda = 532$  nm.

Figure 2 shows changes in the longitudinal energy distribution during photocathode degradation with  $\lambda = 808$  nm. The shape of LEDCs measured with the TESS are strongly affected by local inhomogeneities of the electric field arising from the structure of the grids in the detector. This leads to smearing of the distributions which Terekhov and Orlov previously found to be close to  $\Pi$ -shaped with an exponential right side [10] into one with a bell–shaped character. To determine the positions of the vacuum level ( $\varepsilon_{vac}$ ) and the conduction band minima  $(\varepsilon_{cb})$  in the bulk, we fitted black distributions in the same manner as shown in appendix C of our previously published work [7]. This fitted curve was the convolution of a Gaussian with FWHM of 125 meV (the instrument function of our experiment) and the function  $n_e(U_{ph})$  which was defined as follows:

$$N_{e}(U_{\rm ph}) = \begin{cases} N_{\rm cb} + (N_{0} - N_{\rm cb}) \cdot \frac{\varepsilon_{\rm cb} - U_{\rm ph}}{\varepsilon_{\rm cb}} & \text{for } U_{\rm ph} > \varepsilon_{\rm vac} \\ N_{\rm cb} \cdot \exp\left(-\frac{U_{\rm ph} - \varepsilon_{\rm cb}}{\varepsilon_{0}}\right) & \text{for } U_{\rm ph} \ge \varepsilon_{\rm cb} \end{cases}$$

$$(2)$$

We fitted five parameters  $N_{cb}$ ,  $N_0$ ,  $\varepsilon_0$ ,  $\varepsilon_{cb}$  and  $\varepsilon_{vac}$  to obtain the best fit between the measured LEDC and the calculated convolution. The same fitting process was duly applied to our other measured distributions, but  $\varepsilon_{cb}$  was no longer a fitting parameter. These calculations allowed us to rebuild the abscissa axis from  $U_{ph}$  to  $\varepsilon_{lon} - \varepsilon_{cb}$  and determine the position of the vacuum level for each of the measured distributions, as shown in figure 2 by arrows of the corresponding colour. The difference between  $\varepsilon_{cb}$  and  $\varepsilon_{vac}$  defines the value of effective electron affinity,  $\chi^* = \varepsilon_{vac} - \varepsilon_{cb}$ .

Figure 2 also shows that in region  $\varepsilon_{\text{lon}} < \varepsilon_{\text{cb}}$  the LEDCs are bell–shaped, while for  $\varepsilon_{\text{lon}} > \varepsilon_{\text{cb}}$  the LEDCs have an exponential character of the form  $N_e(\varepsilon_{\text{tr}}) = A \cdot \exp\left(-\frac{\varepsilon_{\text{tr}}}{\varepsilon_0}\right)$  where the value of  $\varepsilon_0$  varies from 33 meV for the black trace to 27 meV for the orange trace. An LEDC of this shape is typical for a GaAs-photocathode [4, 11]. Figure 2 also shows that as for a GaAsP photocathode [7], degradation through exposure to oxygen of a GaAs photocathode leads to a progressive shift in the position of the left–hand side of the LEDC without any significant change in the amplitude of the envelope curve. We observed this behaviour in all of our experiments.

Figure 3 shows eight transverse energy distribution curves of the 40 we measured under illumination at  $\lambda = 808$  nm. Note that in contrast to figure 2, the integrals of the distributions shown in figure 3 are not proportional to the photocathode quantum efficiency. The integral of the upper (black) curve which relates to an oxygen exposure of 0 L is equal to unity. For the purpose of clarity, the distributions related to progressive oxygen exposure have been separated vertically by setting the integral of each subsequent distribution to be  $1\frac{1}{2}$  times less than the integral of the preceding curve. The value  $1\frac{1}{2}$ has no specific meaning and has been chosen only to improve the presentation of the data. The coloured triangles denote the progressive shift in the position of the conduction band minima in the bulk. It can be seen that three upper distributions (black, red and green) correspond to the negative electron affinity (NEA) state, the blue distribution to the effective electron affinity being close to zero, and the four lower distributions (magenta, dark green, dark red and orange) correspond to the positive electron affinity (PEA) state. In the PEA state the TEDCs have exponential character with  $\varepsilon_0$  in the range 24-30 meV. For the NEA state, the TEDC character comprises two parts:

- $\varepsilon_{\rm tr} < \chi^*$  for that part to the left of the coloured triangle
- $\varepsilon_{\rm tr} > \chi^*$  for that part to the right of the coloured triangle



Figure 1. Energy band diagram for a p-GaAs(Cs,O) photocathode.

**Table 1.** Initial photoelectron energy on transition from the heavy-hole  $(\varepsilon_{e-hh})$ , light-hole  $(\varepsilon_{e-lh})$  and split-off  $(\varepsilon_{e-so})$  valence sub-bands in GaAs under illumination at specific wavelengths  $(\lambda)$ .

Wavelength, $\lambda$	$\varepsilon_{e-\mathrm{hh}}$	$\varepsilon_{e-\mathrm{lh}}$	$\varepsilon_{e-\mathrm{so}}$
808 nm 635 nm 532 nm	0.09 eV 0.43 eV 0.72 eV	0.06 eV 0.32 eV 0.59 eV	0.13 eV 0.34 eV

The TEDC shape is close to exponential in both parts, but the slopes of these exponents are significantly different, and the difference decreases progressively with the decreasing value of NEA. Such behaviour in the shape of TEDC measured for transmission-mode GaAs photocathodes has been reported by [12]. In a transmission-mode photocathode, the active layer should be sufficiently thick to permit most electrons to thermalise while they move from their point of generation to the emitting surface, so the energy distribution of electrons emitted from such photocathodes are defined by thermalised electrons. We associate deviations from the exponential form present in the lowest part of the TEDCs with errors that occur during the instrument function deconvolution procedure, as we have previously published [7] where we assert that the dynamic range for valid TEDC measurements by our method is  $1\frac{1}{2}$  orders of magnitude.

Figure 4 shows the change in longitudinal energy distributions during degradation of the photocathode for laser wavelength  $\lambda = 635$  nm. The positions of the vacuum level and the conduction band minima in the bulk were determined by the same method used for  $\lambda = 808$  nm. The black curve was measured immediately after the photocathode was transferred from the PPF to the TESS, and the red curve was measured just before the first oxygen injection. The final orange curve was measured on the morning of the day following the degradation experiment, approximately 17 hr after the last oxygen injection.

Again figure 4 shows that the LEDCs are bell–shaped in the region where  $\varepsilon_{lon} < \varepsilon_{cb}$ , while they have exponential character for  $\varepsilon_{lon} > \varepsilon_{cb}$  with  $\varepsilon_0$  close to 100 meV. The value of  $\varepsilon_0$  decreases slightly over the range  $\varepsilon_{lon} = 0.1-0.4$  eV then decreases sharply for  $\varepsilon_{lon} > 0.45$  eV. The reduction in illumination wavelength has two effects:

- (a) The initial energy of photoelectrons is increased, and consequently the path length required to fully-thermalise these photoelectrons is also increased;
- (b) The absorption coefficient of light in GaAs is increased, and consequently the mean generation depth for photoelectrons is decreased.

Both of these factors increase the fraction of nonthermalised photoelectrons emitted from a reflection-mode GaAs photocathode. A comparison of figures 2 and 4 shows that when illuminated at 635 nm, when  $\varepsilon_{lon} > \varepsilon_{cb}$  the electron emission is dominated by non-thermalised electrons. The sharp change in the slope at  $\varepsilon_{lon} - \varepsilon_{cb} \approx 0.45 \text{ eV}$  is attributed to the maximum initial electron energy which is shown to be 0.43 eV in table 1. Photoelectrons with  $\varepsilon_{lon} - \varepsilon_{cb} >$ 0.43 eV are generated through phonon absorption. The value of  $\varepsilon_0$  for the LEDCs over the range  $\varepsilon_{lon} - \varepsilon_{cb} > 0.45 \text{ eV}$  is equal to kT.

Figure 5 shows the TEDC measured under illumination at  $\lambda = 635$  nm, with the distributions normalized in the same way



**Figure 2.** Evolution of the longitudinal energy distribution curves (LEDC) for electrons emitted from a GaAs photocathode under illumination at a wavelength of  $\lambda = 808$  nm and subjected to progressive QE degradation through exposure to oxygen. The degradation drives a shift in the vacuum level ( $\varepsilon_{vac}$ ) which is annotated. The dashed arrows show the initial photoelectron energies on transition from the heavy–hole and light–hole sub–bands of the valence band.

as those presented in figure 3. As in figure 3, the distributions in figure 5 for the PEA state have an exponential form, while for the NEA state they can be approximately divided into two parts with differing slopes where:

- to the left of the coloured triangle for ε<sub>tr</sub> < χ\*, the slope increases as χ\* decreases;</li>
- to the right of the coloured triangle for ε<sub>tr</sub> > χ<sup>\*</sup>, the slope is approximately constant.

Figures 4 and 5 contain two distributions measured at the same level of oxygen exposure (dose = 0.069 L). The dark red curves in figures 4 and 5 were measured 12 and 2 min respectively after oxygen injection, while the grey curves were measured after 78 and 68 min respectively. The relaxation of photocathode properties which occurs after the end of oxygen injection can clearly be seen. We observed and documented the same relaxation progression in a previous publication [7] for a GaAsP photocathode. In LEDCs this relaxation manifests itself as only a small decrease in the vacuum level, while in TEDCs there is a significant decrease in the slope and a consequent increase in the MTE.

Figures 6 and 7 show measurements of the electron energy distributions with the photocathode under illumination at a wavelength of 532 nm. The shape and behaviour of the distributions does not differ qualitatively from the previous measurements with  $\lambda = 635$  nm, however in our measurements with  $\lambda = 532$  nm, the photocathode was already degraded to an approximate zero-state of effective electron affinity before its first exposure to oxygen. The following day,



**Figure 3.** Evolution of the transverse energy distribution (TEDC) curves for electrons emitted from a GaAs photocathode under illumination at a wavelength of  $\lambda = 808$  nm and subjected to progressive QE degradation through exposure to oxygen. The coloured triangles ( $\mathbf{V}$ ) denote the position of the conduction band minima in the photocathode bulk. The integral of the top–most black curve is equal to unity and relates to an oxygen exposure of 0 L, and the integral of each subsequent distribution is  $1\frac{1}{2}$  times less than the integral of the previous one.

the vacuum level remained below the maximum initial electron energy in the bulk. The consequence of this can clearly be seen for the orange curve in figure 7 by the change in the slope with increasing  $\varepsilon_{tr}$ . The empty triangles in this figure denote the positions of the maximum initial electron energy in the photocathode bulk, and it can be seen that the slope of the orange curve begins to increase significantly after  $\varepsilon_{tr}$  passes through the empty orange triangle. We attribute the change in slope for the blue and dark green TEDCs shown in figure 7 primarily to the same mechanism as that for the orange TEDC, though there may also be some small error present from the instrument function deconvolution procedure.

#### 4. Analysis

To analyse and interpret the data obtained and presented thus far, we constructed figure 8 which shows the dependence of effective electron affinity ( $\chi^*$ ), quantum efficiency (QE), and mean transverse energy (MTE) on the integrated oxygen exposure. The evolution of  $\chi^*$  was inferred using our measured LEDC data. The changes in QE were calculated from changes in the maximum photocurrent observed during LEDC measurements, and also from changes in the total luminosity of the phosphor screen at the detector output recorded by the CCD camera during TEDC measurements. The optical density of filters used at each illumination wavelength and changes in the CCD camera exposure time were also taken into account when assessing the QE. The MTE values were calculated from the measured TEDCs.

Figure 8(a) summarises the behaviour of the effective electron affinity, and it can be seen that for all illumination



**Figure 4.** Evolution of the longitudinal energy distribution curves (LEDC) for electrons emitted from a GaAs photocathode under illumination at a wavelength of  $\lambda = 635$  nm and subjected to progressive QE degradation through exposure to oxygen. This degradation causes the vacuum level ( $\varepsilon_{vac}$ ) to shift progressively, and is shown using solid coloured arrows. The dashed black arrows show the initial photoelectron energies on transitions from different sub–bands of the valence band.

wavelengths the rate of increase in  $\chi^*$  slows with increasing oxygen exposure. In the initial section, the rate of change of  $\chi^*$ is  $6 \pm 1 \text{ eV/L}$  which agrees well with our previously published value of 5.7 eV/L for a GaAsP photocathode in the same configuration [7]. For our experiments with  $\lambda = 635 \text{ nm}$ , before the first oxygen exposure,  $\chi^*$  increased by 17 meV in comparison to the state immediately after photocathode transfer from the PPF. In our experiments with  $\lambda = 532 \text{ nm}$ , this increase was 73 meV. The overnight increase in  $\chi^*$  after experiments involving oxygen degradation was 290 meV for  $\lambda = 635 \text{ nm}$ and 220 meV for  $\lambda = 532 \text{ nm}$ .

Figure 8(b) shows the behaviour of the GaAs photocathode quantum efficiency during oxygen degradation. It can be seen that for  $\lambda = 808$  nm, during oxygen degradation the quantum efficiency decreased by more than four orders of magnitude, while for  $\lambda = 635$  and 532 nm respectively, the decrease in QE did not exceed  $1\frac{1}{2}$  orders of magnitude. Overnight, following our oxygen degradation experiments with  $\lambda = 635$  nm, the QE fell by a factor of 1300 times, while for  $\lambda = 532$  nm the decrease in QE was only by a factor of seven times. We note that the relative changes in QE for  $\lambda = 635$  nm and  $\lambda = 532$  nm over the oxygen exposure range 0.01-0.06 L are very close, and this is attributed to the closeness of the slope of the envelope functions for the LEDCs over the range 0.1 eV < $\varepsilon_{\rm lon} - \varepsilon_{\rm cb} < 0.4\,{\rm eV}$ . The integrated oxygen exposure required to degrade the QE by a factor of 10  $(D_{\text{ox}}^{10})$  for  $\lambda = 808 \text{ nm}$ and  $\lambda = 635 \, \mathrm{nm}$  was  $\approx 0.02 \, \mathrm{L}$  which is in good agreement with data published by [2] and [13]. For  $\lambda = 532 \text{ nm}$ ,  $D_{\text{ox}}^{10} =$ 0.055 L which corresponds to a more than twofold increase which is mainly due to the fact that in our experiments with



**Figure 5.** Evolution of the transverse energy distribution (TEDC) curves for electrons emitted from a GaAs photocathode under illumination at a wavelength of  $\lambda = 635$  nm and subject to progressive QE degradation through exposure to oxygen, annotated with filled coloured triangles ( $\mathbf{V}$ ) which denote the position of the conduction band minima in the photocathode bulk. The integral of the initial black curve for an oxygen exposure of 0 L is equal to unity, while the integral for each subsequent distribution is  $1\frac{1}{2}$  times less than the integral of the previous one.

 $\lambda = 532$  nm, oxygen was applied to a photocathode surface which had practically zero effective electronic affinity in contrast to the significant negative electronic affinity state in our other experiments.

Figure 8(b) also shows the relaxation of the photocathode properties after the conclusion of oxygen exposure which is clearly visible in the form of vertical 'segments' signifying some small degree of QE recovery. These arise because our TEDCs are measured 2-3 min after the end of each oxygen dose, then the experiment is reconfigured for the LEDC measurement, after which the original configuration is restored to re-measure the TEDC. The time between TEDC measurements at the same oxygen dose was 12-100 min. The degree of QE relaxation was maximal with  $\lambda = 808$  nm, with the magnitude of relaxation also dependent on the size of the last oxygen dose and the elapsed relaxation time. For example, with  $\lambda = 808 \text{ nm}$  and following an oxygen exposure of 0.033 L, the quantum efficiency increased by a factor of 3.3 times. The last oxygen exposure was 0.005 L and the relaxation time was 100 min, and following the total 0.049 L exposure, the quantum efficiency increased by 2.5 times, the final oxygen exposure was 0.009 L, and the associated relaxation time was 16 min.

Figure 8(c) shows changes in the MTE during photocathode degradation. For  $\lambda = 808$  nm, the MTE was seen to decrease monotonically from 60 meV to the *kT* level, while for other wavelengths, the MTE behaviour is non-monotonic. Under illumination with  $\lambda = 635$  nm, the MTE first decreased in a similar way as that seen for  $\lambda = 808$  nm, but then increased to a local maximum as the photocathode suffered degradation, and then again decreased gradually. For  $\lambda = 532$  nm, the initial



Figure 6. Evolution of the longitudinal energy distribution curves (LEDC) for electrons emitted from a GaAs photocathode under illumination at a wavelength of  $\lambda = 532$  nm and subject to progressive QE degradation through exposure to oxygen. The degradation causes the vacuum level ( $\varepsilon_{vac}$ ) to shift progressively, and is shown using coloured solid arrows. The black dashed arrows show the initial photoelectron energy values on transition from different sub-bands of the valence band.

interval with the MTE decreasing was not observed-instead MTE increased immediately to a maximum and then exhibited a gradual decrease.

The MTE behaviour also exhibits vertical segments characterising the relaxation processes which occur after the end of successive oxygen exposures. Figure 8(c) also shows that the relaxation processes always tend to restore the previous state. For the case of an increase in MTE following an increase in the oxygen exposure level, relaxation decreases the MTE; and in the case of a decrease in MTE following an increase in the oxygen exposure level, relaxation increases the MTE. However, relaxation cannot be simply explained by a return to some previous state of the system. Under illumination with  $\lambda = 635$  nm, during relaxation after a total oxygen exposure of 0.069 L, the quantum efficiency increased from 0.26% to 0.42% and the MTE increased from 55 to 67 meV. Based on the previous arguments, the quantum efficiency of 0.42% corresponds to an MTE of 63 meV. This implies that in addition to a decrease in the work function (see the dark red and grey curves in figure 4), there were also changes in the electron scattering and transport to the surface during emission into vacuum. We therefore associate the relaxation processes observed following oxygen exposure both to the partial desorption of adsorbed oxygen, and to the slow incorporation of adsorbed oxygen into the (Cs,O) activation layer.

We ascribe uncertainties of  $\pm 12.5 \text{ meV}$  for our electron affinity ( $\chi^*$ ) values, and relative uncertainties of  $\pm 15\%$  for QE and  $\pm 10\%$  for MTE values.

In order to understand the reasons for this non-monotonic MTE behaviour, we reprocessed the data presented in figure 8 L B Jones et al



Figure 7. Evolution of the transverse energy distribution curves (TEDC) for electrons emitted from a GaAs photocathode under illumination at a wavelength of  $\lambda = 532$  nm and subject to progressive QE degradation through exposure to oxygen. The filled coloured triangles  $(\mathbf{\nabla})$  denote the position of the conduction band minima in the photocathode bulk, and its progressive shift under degradation. The empty coloured triangles  $(\nabla)$  denote the positions of maximum initial electron energy in the photocathode bulk. The integral of the initial black curve for an oxygen exposure of 0 L is equal to unity, while the integral for each subsequent distribution is  $1\frac{1}{2}$  times less than the integral of the previous one.

to extract the dependence of the QE and MTE on the value of the effective electronic affinity,  $\chi^*$ . To accomplish this, we fitted the dependence of  $\chi^*$  to the oxygen exposure using a second-order polynomial. These fits are shown in figure 8(a)by the dashed lines whose colour corresponds to illumination wavelength,  $\lambda$ . To correctly account for relaxation phenomena at the LEDC measurement points, QE was determined from the LEDC and MTE from the TEDC measured immediately following experimental reconfiguration after these LEDC measurements. These results are shown in figure 9.

Under illumination with  $\lambda = 808$  nm, QE in the NEA region  $(\chi^* < 0)$  decreases linearly with an increase in electron affinity, but in the region of PEA  $(\chi^* > 0)$  the decrease is exponential with a slope whose characteristic energy equates to 28 meV. For shorter wavelengths, the QE decrease in the NEA region is also close to linear, while that in the PEA region has an exponential decay with a gradual decrease in slope.

The MTE behaviour is more complex. For  $\lambda = 808 \text{ nm}$ there is a progressive decrease in MTE as  $\chi^*$  approaches zero in the NEA region, ultimately reaching a value around kTwhich remains constant over the PEA region. For  $\lambda = 635$  nm the MTE first decreases from 81 meV to a minimum around 63 meV with  $\chi^* = -0.08$  eV, after which the MTE increases to a local maximum around 77 meV with  $\chi^* = 0.05$  eV, and then progressively decreases as  $\chi^*$  increases further. The MTE behaviour for  $\lambda = 532 \text{ nm}$  differs qualitatively from that for  $\lambda = 635$  nm only by the absence of the initial decrease, with an MTE maximum close to 120 meV observed in the region  $0 < \chi^* < 0.1 \, \text{eV}.$ 



**Figure 8.** Analysis and comparison of all data for our mean transverse energy measurements, with fit lines added to aid interpretation. The black curves and symbols relate to our measurements under illumination with  $\lambda = 808$  nm, the red curves and symbols to measurements with  $\lambda = 635$  nm, and the green to  $\lambda = 532$  nm.

When a reflection-mode GaAs photocathode is illuminated at a wavelength of  $\lambda = 808$  nm, thermalised electrons account for more than 95% of all emitted electrons both for the negative electron affinity state and the positive electron affinity state up to  $\chi^* \sim 150$  meV (see [9], figure 3). This implies that our data measured with  $\lambda = 808$  nm conforms to the behaviour expected for thermalised electrons.

When comparing our data with that published by other researchers, it should be remembered that MTE measurements in [11, 14–16] were obtained while illuminating the photocathode in the transmission–mode, and in this scenario thermalised electrons are emitted into the vacuum. Therefore the results of these studies should only be compared with our data obtained when illuminating the photocathode at a wavelength of  $\lambda = 808$  nm. Rodway *et al* [14] quoted the MTE of a fully–activated transmission–mode GaAs photocathode as  $100 \pm 10$  meV. According to the Heidelberg group [11, 15, 16], the fully–activated GaAs photocathode at room temperature has an MTE in the 60–100 meV range. For comparison, we have added published data from [11] denoted by a black star ( $\bigstar$ ), and [15] denoted by a grey star ( $\bigstar$ ) to figure 9(b).

Figure 9(b) shows firstly that our data are consistent with published data, and secondly that the MTE is **not** an unambiguous monotonic function of the photocathode effective



**Figure 9.** The dependence of quantum efficiency (QE) and mean transverse energy (MTE) on effective affinity ( $\chi^*$ ), with solid lines added to guide the eye. The black curve and triangles ( $\blacktriangle$ ) relate to our measurements under illumination at  $\lambda = 808$  nm, the red curve and circles ( $\bullet$ ) to  $\lambda = 635$  nm, and the green curve and squares ( $\blacksquare$ ) to  $\lambda = 532$  nm. The additional data shown in (b) is that from Pastuszka *et al* [11] denoted by the black star ( $\bigstar$ ), Orlov *et al* [15] denoted by the grey star ( $\bigstar$ ), and Bazarov *et al* [19] by the coloured dashed horizontal lines.

electron affinity. Thus, the GaAs photocathode used by [15] had an MTE of 59 meV at the effective electron affinity of  $\chi^* = -0.155$  eV, and that used by [11] had an MTE of 77 meV at  $\chi^* = -0.145 \,\text{eV}$ . The reasons for these differences may relate to the physical and chemical roughness of the photocathode surfaces, as suggested by [17]. Differences in the value of effective electron affinity obtained after activation also lead to differences in the observed MTE values. The monotonic decreasing behaviour for the MTE of emitted thermalised electrons during photocathode degradation in figure 9(b) also corresponds to the data presented by [16] and [15] in which an additional electrode was used to suppress the low-energy portion of the emitted electrons which, as can be seen by figure 6, is equivalent to oxygen degradation in our experiments. In work published by [18], the MTE of reflection-mode GaAs photocathodes were measured under illumination with a photon energy close to that of the band gap. The observed emission was dominated by thermalised electrons with an MTE of  $63 \pm 11$  meV, and its subsequent monotonic decrease under photocathode degradation also agrees well with our data.

When a photocathode is illuminated in reflection-mode with light whose photon energy significantly exceeds the band gap, both thermalised and non-thermalised electrons are emitted. In this case, the observed MTE essentially depends on the initial energy of the photoelectron and the average distance from the generation point to the emitting surface. Both of these quantities are determined by the wavelength of the incident light. The MTE dependence on the illumination wavelength for reflection-mode GaAs photocathodes was studied by [19] for which the authors obtained MTE values of  $113 \pm 8 \text{ meV}$  for  $\lambda = 532 \text{ nm}$ ,  $81 \pm 6 \text{ meV}$  for  $\lambda = 633 \text{ nm}$  and  $33 \pm 3 \text{ meV}$  for  $\lambda = 808 \text{ nm}$ . These values are denoted by coloured dashed horizontal lines in our figure 9(b), and it can be seen that in the range  $0 < \chi^* < 0.1 \text{ eV}$ , our data are in good agreement.

Matsuba *et al* [20] measured the MTE of three reflectionmode GaAs photocathodes under illumination with  $\lambda = 785$ and 544 nm. For  $\lambda = 785$  nm they observed MTE in the range 31 to 37 meV, and for  $\lambda = 544$  nm MTE were in the range 57–60 meV. Their results for  $\lambda = 785$  nm agree with our data and the rest of that published by other groups, but their MTE values obtained for  $\lambda = 544$  nm are approximately half that of both our results for  $\lambda = 532$  nm and those published by [19]. The reason for such low MTE values at the green wavelength range is unclear and cannot be explained by the high positive value for the effective electronic affinity, since according to their data [20], the QE of their GaAs photocathode at  $\lambda = 785$  nm was 1%–5% which indicates that a state of negative effective electronic affinity was achieved.

If we assume that the ensemble of emitted electrons consists of two groups containing  $N_1$  and  $N_2$  electrons with mean transverse energy for each group of MTE<sub>1</sub> and MTE<sub>2</sub>, then it is easy to show that the mean transverse energy of the entire electron ensemble is given by:

$$MTE = \frac{N_1}{N_1 + N_2} \times MTE_1 + \frac{N_2}{N_1 + N_2} \times MTE_2.$$
 (3)

In our experiments, the two groups of electrons are thermalised and non-thermalised electrons, denoted by  $N_{\text{th}}$  and  $N_{\text{non-th}}$ respectively. Changes in the number of thermalised electrons emitted as a function of effective electronic affinity are proportional to changes in the quantum yield for  $\lambda = 808$  nm.

$$N_{\rm th}(\chi^*) = C_1 \times \, \text{QE}\left(\chi^*, \, \lambda = 808 \text{ nm}\right) \tag{4}$$

where  $C_1$  is a constant. Our data shows that for  $\lambda = 808$  nm, changes in the MTE as a function the progressive shift in effective electronic affinity are due to the dominance of thermalised electrons in the photoemission distribution:

$$MTE_{th}(\chi^*) = MTE(\chi^*, \lambda = 808 \text{ nm}).$$
 (5)

Figure 9(a) shows that for  $\lambda = 635$  nm and  $\lambda = 532$  nm, emission at  $\chi^* > 0.1$  eV is dominated by non-thermalised electrons while the number of thermalised electrons is determined by QE as shown by  $\lambda = 808$  nm, therefore:

$$N_{\text{non-th}} (\chi^* > 0.1 \text{ ev}, \lambda = 635, 532 \text{ nm})$$
  
=  $C_{2,3} \times \text{QE} (\chi^* > 0.1 \text{ ev}, \lambda = 635, 532 \text{ nm})$  (6)

$$MTE_{non-th} (\chi^* > 0.1 \text{ ev}, \lambda = 635, 532 \text{ nm}) = MTE (\chi^* > 0.1 \text{ ev}, \lambda = 635, 532 \text{ nm})$$
(7)



**Figure 10.** Dependence of quantum efficiency (QE) and mean transverse energy (MTE) on effective electron affinity  $(\chi^*)$ . Red curves and circles (•) correspond to illumination with  $\lambda = 635$  nm. Green curves and triangles ( $\mathbf{V}$ ) correspond to illumination with  $\lambda = 532$  nm. The symbols represent our experimental data and the solid lines the result of fitting using equation (3). Dashed lines represent the non-thermalised electron group, and the dotted lines represent the thermalised electron group.

where  $C_2$  and  $C_3$  are constants. In order to estimate the number of non-thermalised electrons emitted and their mean transverse energy for  $\chi^* < 0$ , we fitted the dependencies QE ( $\chi^*$ ) and MTE( $\chi^*$ ) over the range  $0.08 < \chi^* < 0.3 \text{ eV}$  using an exponent and a straight line respectively, and extrapolated the obtained dependences for all  $\chi^*$ .

$$N_{\text{non-th}}(\chi^*, \lambda = 635, 532 \text{ nm}) = C_{2,3} \times (A_{1,2} \times \chi^* + B_{1,2})$$
(8)

$$MTE_{non-th}(\chi^*, \lambda = 635, 532 \text{ nm}) = \exp(A_{3,4} \times \chi^* + B_{3,4})$$
(9)

where  $A_1$  to  $A_4$  and  $B_1$  to  $B_4$  are results of the fitting process. Combining equations (3)–(5), (8) and (9), we can fit our QE ( $\chi^*$ ) data for illumination with  $\lambda = 635$  nm and  $\lambda = 532$  nm with only one pair of parameters, either  $C_1$  and  $C_2$  or  $C_1$  and  $C_3$ , and then calculate MTE( $\chi^*$ ). The result of this fit is shown in figure 10.

Figure 10 demonstrates that our calculation results (shown by the solid lines) are in qualitative agreement with our experimental data (denoted by the symbols). The quantitative difference in MTE behaviour between the calculated and experimental data is explained by inaccuracies related to the decomposition of thermalised and non-thermalised electrons into their respective groups, and the subsequent determination of MTE for each of these electron groups. Nevertheless, the calculations we have performed provide an explanation for the non-monotonic MTE behaviour with a progressive change in  $\chi^*$ . For negative values of  $\chi^*$ , thermalised electrons make up a significant fraction of the emitted electron distribution. For example, at  $\chi^* = -0.1$  eV, the fraction of thermalised electrons with  $\lambda = 635$  nm is about 55%, and falls to about 35% for  $\lambda = 532$  nm. As  $\chi^*$  increases, the MTE of the thermalised electron group decreases (MTE<sub>th</sub>, shown by dotted lines in figure 10), which leads to a decrease in MTE for the entire distribution. However at the same time, the fraction of thermalised electrons in the distribution also decreases rapidly, driving the overall MTE towards that for the group of non-thermalised electrons (MTE<sub>non-th</sub>, shown by dashed lines in figure 10) and thus drives MTE growth. The decrease in MTE<sub>non-th</sub> arising from the progressive increase in  $\chi^*$  drives the decrease in MTE from its local maximum. As we have already indicated, the MTE of a photocathode depends on many parameters and is not an unambiguous function of  $\chi^*$ , and hence we are not asserting that we have de-convoluted the universal dependencies encapsulated within MTE( $\chi^*, \lambda$ ).

Changes in the proportions of thermalised and nonthermalised electrons in the emitted distribution will naturally lead to a similar non-monotonic behaviour of the MLE with increasing  $\chi^*$ . Our experimental setup has a broad instrumental function when measuring LEDC to the extent that the error in determining the MLE exceeds the magnitude of this non-monotonic behaviour. Nevertheless, from the shape of the LEDCs in figures 4 and 6 and their evolution with increasing  $\chi^*$ , coupled with the principle of energy conservation in the photoemission process, we can conclude that the MLE behaviour should also be non-monotonic.

When the photocathode is operating as an electron source for a particle accelerator, the photocathode degradation rate and the maximum power output of the laser illumination light source determine the accelerator operational period before photocathode replacement becomes necessary. MTE is an important and intrinsic photocathode parameter, but it cannot be controlled during accelerator operation-photocathode degradation dictates the level of MTE in the injected electron bunch. Therefore knowledge of the MTE behaviour during photocathode degradation is crucial. Our data show that when using a reflection-mode GaAs photocathode whose initial  $\chi^* = -0.1 \text{ eV}$  under illumination with  $\lambda = 635 \text{ nm}$ , degradation in the quantum efficiency by a factor of 10 during accelerator operations which shifts the effective electron affinity to zero (see figure 9(b) for  $-0.1 < \chi^* < 0 \text{ eV}$ ) will lead to an increase in photocathode MTE from an initial value 65 meV with high photocathode QE to an ultimate value of 77 meV with low QE at the end of accelerator operations.

Degradation of the photocathode in an accelerator photoemission electron gun is driven by several mechanisms, some of which impact in a similar fashion to oxygen exposure on the photocathode. While ion back–bombardment is a clear exception to this, we believe that our data illustrate a crucial pathway for MTE changes during the degradation of a GaAs photocathode, and this mechanism must be taken into account when planning the operation of an accelerator which relies on the consistent delivery of a high–quality, high–brightness electron beam.

#### 5. Conclusions

We have measured both the transverse and longitudinal energy distribution curves for a reflection-mode GaAs photocathode which was subjected to progressive degradation by exposure to oxygen. Our experiments were performed for three illumination wavelengths with  $\lambda = 808$ , 635 and 532 nm. Following each period of oxygen exposure, some *relaxation* was observed in the photocathode properties which was evident as recovery in the QE. We associate this relaxation process firstly with the partial desorption of adsorbed oxygen, and secondly with the slow incorporation of adsorbed oxygen into the (Cs,O) activation layer.

Our experiments have shown that during degradation of a reflection-mode GaAs photocathode, the MTE can both decrease and increase, with the level of this increase being dependent on the illumination wavelength. This nonmonotonic MTE behaviour is explained by changes in the relative population of the thermalised and non-thermalised groups in the photoemitted electron distribution, and we have provided a model which fits our experimental results.

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