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PHOTOCATHODE PERFORMANCE CHARACTERISATION OF ULTRA-THIN MgO FILMS ON POLYCRYSTALLINE COPPER

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Abstract

The performance expected from the next generation of electron accelerators is driving research into photocathode technology as this fundamentally limits the achievable beam quality. The performance characteristics of a photocathode are most notably: normalised emittance, brightness and energy spread. Ultra–thin oxide films on metal substrates have been shown to lower the work function (WF) of the surface, enhancing commonly utilised metal photocathodes, potentially improving lifetime and performance characteristics.

We present the characterisation of two MgO/Cu photocathodes grown at Daresbury. The surface properties such as: surface roughness, elemental composition and WF, have been studied using atomic force microscopy (AFM), X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS). The photoemissive properties have been characterised with quantum efficiency (QE) measurements at 266 nm. Additionally, we measure the Transverse Energy Distribution Curves (TEDC) for these photocathodes under illumination at various wavelengths using ASTeC's Transverse Energy Spread Spectrometer (TESS) and extract the Mean Transverse Energy (MTE).

INTRODUCTION

The development and improvement of the next generation electron sources require photocathodes with a high quantum efficiency (QE), low emittance, fast response times and robustness. The photoemissive properties of photocathodes are governed by its surface characteristics. The surface roughness and the work function (WF) strongly influence the quantum efficiency and intrinsic emittance. Metal photocathodes are predominantly used due to their high durability and fast response time. However, their relatively high WF leads to a low QE and the requirement for UV drive lasers [1].

The mean transverse energy (MTE) is a photoemissive property which is directly dependent on the excess energy in the photoemission process, where the excess energy is the difference between the photon energy and the WF of the sample. The MTE can be approximated using the model derived by Dowell and Schmerge [2]:

$$MTE = \frac{1}{3}(h\omega - \phi) \quad (1)$$

where $h\omega$ is the incident photon energy and ϕ is the WF of the photocathode, which was measured using ultraviolet photolectron spectroscopy (UPS) (see Table 1).

One possible method of improving the performance of photocathodes is by the use of surface preparation or treatment, lowering the surface WF and increasing the QE. Surface dielectric films on metals have been shown to produce a surface with a lower WF [3] and previous works [4, 5] on MgO thin films on Ag(100) have demonstrated this reduction in WF and the potential for QE enhancement. Therefore, MgO films have the potential to improve the QE, while simultaneously increasing the robustness of the sample due to the chemical stability of MgO forming an overlayer to protect from residual gases in the system.

EXPERIMENTAL DETAILS

Sample Preparation

Two 6 mm diameter polycrystalline copper cathodes, with a surface roughness of $R_a < 30$ nm, (labeled DaCB–7 and DaCB–14) were supplied by Surface Preparation Laboratory (SPL). Cathodes were cleaned *in situ* using cycles of Ar^+ bombardment (2 keV beam energy for 20 mins) and annealing at 500 °C, with surface contamination being assessed using X-ray photoelectron spectroscopy (XPS). Once clean, an ultra-thin MgO film layer was deposited by thermal evaporation of Mg in a Chell K-Cell Miniature Knudsen Evaporation Cell in an O_2 partial pressure of 5×10^{-7} mbar. Sample preparation, QE, XPS and ultraviolet photoelectron spectroscopy (UPS) were all conducted within the same UHV system with a base pressure of 3×10^{-9} mbar.

Surface Characterisation

XPS spectra were obtained using an non-monochromated Al K_α X-ray source (1486.7 eV) and a Thermo Alpha 110 analyser. The analyser transmission function was determined experimentally using the technique described by Ruffiuex et al. [6], and the WF of the analyser was measured using the Fermi edge of Ag. Core region spectra were acquired with a pass energy of 20 eV. Analysis was conducted using CasaXPS [7].

The surface roughness of DaCB–7 was measured using atomic force microscopy (AFM) conducted at the University of Warwick using a Bruker dimension icon AFM after all other work was completed. AFM scans over 10 μm × 10 μm and smaller were repeated over two locations on the surface. The photocathode was transported under ultra high vacuum conditions, exposed to maximum pressure of 5×10^{-10} mbar during transport. This reduced surface contamination and formation of hydroxides and carbonates.

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QE measurements used a Crylas FQSS Q4 266 nm, 1 kHz pulsed laser source coupled with a ×3 beam expander and 2 mm circular aperture, and then a 2.0 OD reflective filter yielding an optical power of 19.2 μW illumination on the sample. A HV extraction electrode was placed close to the sample and a lock-in amplifier was used to measure the total yield photocurrent, thus ensuring any DC current from external source is not sampled and noise is reduced.

Energy Spread Measurement

Samples were transferred to the transverse energy spread spectrometer (TESS) under ultra high vacuum (UHV) conditions ($< 10^{-10}$ mbar). TESS captures the photoemission footprint of a photocathode when illuminated by various wavelengths of of light [8]. The mean transverse energy (MTE) is then extracted from its radial distribution [9].

EXPERIMENTAL RESULTS

Table 1: QE and WF of Substrate and Thin Film Sample, The Error for the WF is ± 0.1 eV

Sample	Substrate WF (eV)	QE	MgO Film WF (eV)	QE
DaCB–7	4.9	1×10^{-6}	3.7	7.0×10^{-5}
DaCB–14	5.0	8×10^{-7}	3.2	4.5×10^{-5}

Both polycrystalline copper substrates underwent the same surface preparation and *in situ* cleaning. Sample preparation was continued to the point at which XPS showed no surface contamination from adventitious carbon or oxidation. Following this, the QE of each substrate was measured to be 1×10^{-6} and 8×10^{-7} for DaCB–7 and DaCB–14 respectively. Previously reported copper QE values are significantly higher [1]. The WF of both samples (Table 1) were measured and fall within reference Cu values [10], albeit on the high end. The photon energy at 266 nm is 4.66 eV, lower than the measured WF of both Cu substrates. Consequently, a low QE is expected due to the Fermi distribution at room temperature.

Figure 1 shows the post-deposition surface elemental composition of both photocathodes, DaCB–7 and DaCB–14. In regards to DaCB–7, the Cu $2p_{3/2}$ (Fig. 1a) has a peak position of 932.6 eV and a FWHM of 1.3 eV, suggesting a clean copper surface free from any significant oxidation from the O_2 exposure during deposition. The lineshape of the O 1s (Fig. 1e) suggests two components contributing to the core region spectrum; MgO and Cu_2O. The MgO film is stoichiometric as there is no observable peak at the binding energy (BE) 1303 eV in the Mg 1s spectrum, Fig. 1c. Using the method outlined by Cumpson (2000) [11], we estimate the MgO film thickness to be 0.24 nm ± 0.02 nm.

Similar features are also observed in the DaCB–14 XPS spectra. The copper substrate has remained relatively oxide-free following deposition and no observable Mg metallic component is present in the Mg 1s, Fig. 1d. A slight asymmetry in the O 1s shown in Fig. 1f once again suggesting a trace amount of Cu_2O, although relatively this is significantly less prominent than for the previous sample. We estimate the film thickness to be thicker at 0.71 nm ± 0.07 nm.

Figure 2 shows the measured MTE for both DaCB–7 and DaCB–14 photocathodes under different illumination wavelengths. All data points have an experimental error of ± 10 %. Overall, the data shows the direct dependence of the MTE on the excess energy as predicted by Dowell [2]. The plotted lines represent the MTE approximation, given in Eq. (1).

At 266 nm the MTE was 374 meV and 507 meV for DaCB–7 and DaCB–14 respectively. Previous work on copper photocathodes reported values of 137.2 meV and 162.6 meV for two different Cu polycrystalline samples [12].

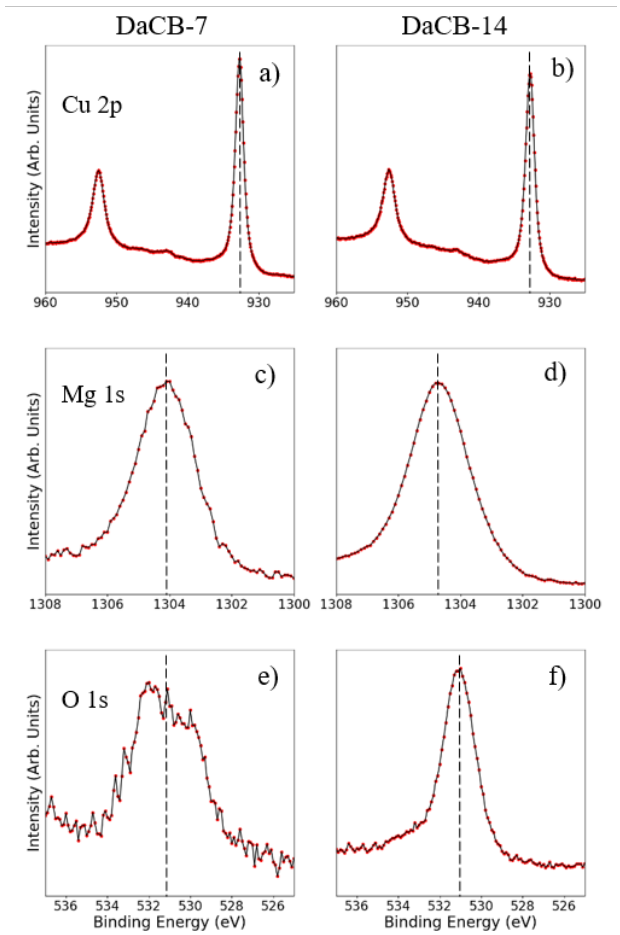

Figure 1: XPS core region scans of DaCB–7 (a, c and d) and DaCB–14 (b, d and f) following ultra-thin film deposition. XPS spectra were acquired for the core regions; Cu 2p (a, b), Mg 1s (c, d) and O 1s (e, f), showing a ultra-thin film MgO layer on the Cu substrate.

At threshold emission both samples achieved a MTE value of 28 meV and 26 meV, commensurate with the minimum energy determined by the temperature of the system (k_bT), 25 meV at room temperature. The two photocathodes varied

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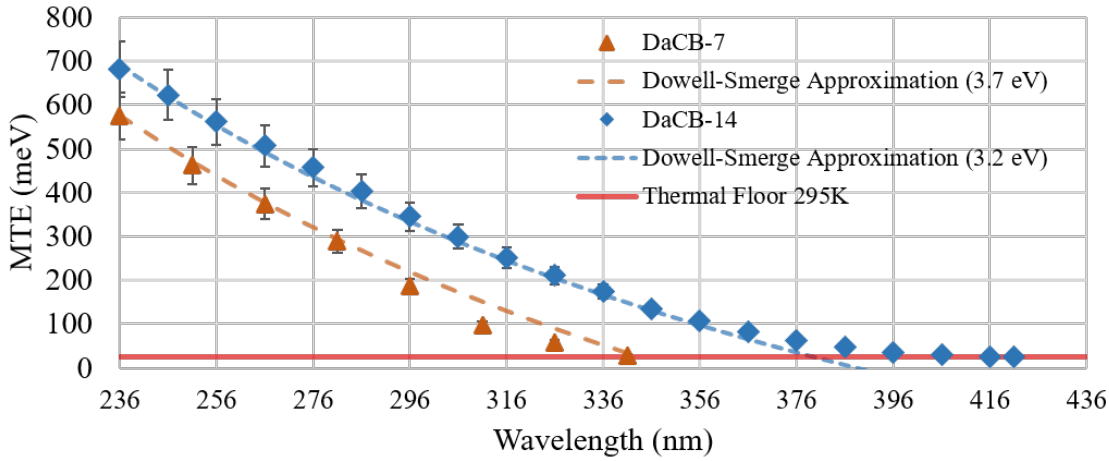

Figure 2: MTE measurements for DaCB–7 and DaCB–14 photocathodes. Dashed lines show the MTE predicted by Eq. (1). The MTE thermal floor is defined by k_bT at room temperature is shown by the solid red line.

greatly at the photoemission threshold with a difference of 80 nm, reflecting the measured difference in WF.

Fig. 3a shows surface topography over a 10 μm × 10 μm AFM scan. The distinctive step terrace array in the surface topography indicates film growth. We measured a RMS roughness of 4.1 nm over the entire surface, less than the quoted surface roughness at the time of the substrate manufacture by SPL. Figures. 3b and 3c, show the forward and backward lateral force measurements (LFM). Faint lines can be observed at the steps terraces, as well as a distinct 'snakelike' contrast.

DISCUSSION

Relative to the Cu substrates, the deposition of the ultra-thin MgO film delivers a substantial improvement in QE at 266 nm. This can be related to the reduction in the surface WF by -1.2 eV and -1.8 eV for DaCB–7 and DaCB–14 respectively. The MTE agrees strongly with the MTE approximation discussed above [2] showing a direct dependence on the excess energy defined by Eq. (1) in the photoemitted electrons.

Variations between the photocathodes in their MTE and WFs can be attributed to the thickness and quality of the film. The thinner film, DaCB–7, has a smaller relative shift in the work function when compared to the thicker DaCB–14. With reference to to Fig. 1, the XPS suggests Cu_2O surface contamination on DaCB–7 which would greatly affect the observed surface WF. Conversely, DaCB–7 exhibits a factor of 70 increase in QE relative to the bare substrate surface, compared to only 56 for DaCB–14. This difference in QE may be ascribed to the increased thickness of the MgO film on DaCB–14 attenuating the photoemitted electron yield.

As discussed above, at their respective threshold energies the photocathodes both approached the thermal limit of 25 meV. Based on the surface roughness measured for DaCB–7 of 4.1 nm RMS, we conclude that the surface roughness is not a contributing factor for the MTE in DaCB–7.

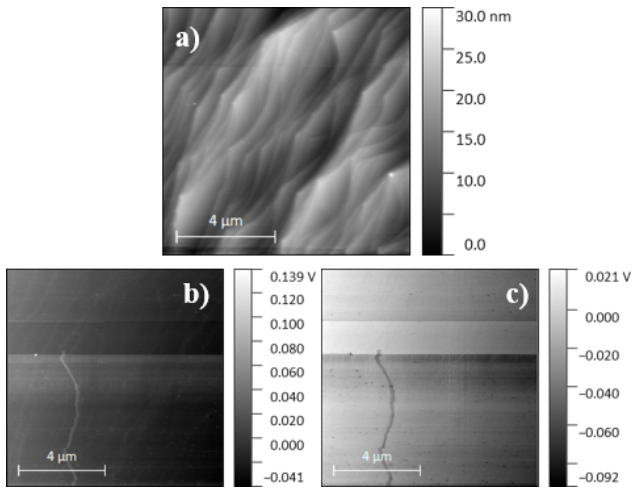

Figure 3: 10 μm × 10 μm AFM and LFM scans of DaCB–7 photocathode: a) AFM height showing step terrace features; b) LFM image of forward scan; c) LFM image of backward scan showing minimal friction contrast.

The same conclusion may also be drawn for DaCB–14 due to the comparative sample preparation and surface treatment.

CONCLUSION

In this paper we show the potential, predicted by Chang [4], of ultra-thin MgO film metal photocathodes. The photocathodes presented exhibit a WF reduction of more than 1 eV relative to the substrate and an increase in QE of more than an order of magnitude. Furthermore, their MTE precisely follows that predicted by Dowell and Schmerge [1]. This significant and measurable improvement in photocathode performance and the potential for enhanced robustness and operational lifetime in an accelerator environment, highlights the use of MgO films as a candidate for future photocathode applications.

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REFERENCES

[1] D. Dowell *et al.*, "Cathode r&d for future light sources," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 622, pp. 685–697, 2010, doi:10.1016/j.nima.2010.03.104

[2] D. H. Dowell and J. F. Schmerge, "Quantum efficiency and thermal emittance of metal photocathodes," *Phys. Rev. ST Accel. Beams*, vol. 12, p. 074 201, 2009, doi:10.1103/PhysRevSTAB.12.074201

[3] S. Prada, U. Martinez, and G. Pacchioni, "Work function changes induced by deposition of ultrathin dielectric films on metals: A theoretical analysis," *Physical Review B*, vol. 78, p. 235 423, 2008, doi:10.1103/PhysRevB.78.235423

[4] V. Chang, T. C. Q. Noakes, and N. M. Harrison, "Work function and quantum efficiency study of metal oxide thin films on ag(100)," *Physical Review B*, vol. 97, p. 155 436, 2018, doi:10.1103/PhysRevB.97.155436

[5] T. Jaouen, G. Jézéquel, G. Delhaye, B. Lépine, P. Turban, and P. Schieffer, "Work function shifts, schottky barrier height, and ionization potential determination of thin mgo films on ag(001)," *Applied Physics Letters*, vol. 97, p. 232 104, 2010, doi:10.1063/1.3525159

[6] P. Ruffieux *et al.*, "Experimental determination of the transmission factor for the omicron ea125 electron analyzer," *Review of Scientific Instruments*, vol. 71, p. 3634, 2000, doi:10.1063/1.1313798

[7] N. Fairley *et al.*, "Systematic and collaborative approach to problem solving using x-ray photoelectron spectroscopy," *Applied Surface Science Advances*, vol. 5, p. 100 112, 2021, doi:10.1016/j.apsadv.2021.100112

[8] L. B. Jones *et al.*, "The Commissioning of Tess: An Experimental Facility for Measuring the Electron Energy Distribution From Photocathodes," in *Proc. FEL'13*, New York, NY, USA, Aug. 2013, pp. 290–293, https://jacow.org/FEL2013/papers/TUPSO33.pdf

[9] L. B. Jones, H. E. Scheibler, D. V. Gorshkov, A. S. Terekhov, B. L. Militsyn, and T. C. Q. Noakes, "Evolution of the transverse and longitudinal energy distributions of electrons emitted from a gaasp photocathode as a function of its degradation state," *Journal of Applied Physics*, vol. 121, p. 225 703, 2017, doi:10.1063/1.4984603

[10] H. Kawano, "Effective work functions for ionic and electronic emissions from mono- and polycrystalline surfaces," *Progress in Surface Science*, vol. 83, pp. 1–165, 2008, doi:10.1016/j.progsurf.2007.11.001

[11] P. J. Cumpson, "The thickogram: A method for easy film thickness measurement in xps," *Surface and Interface Analysis*, vol. 29, pp. 403–406, 2000, doi:10.1002/1096-9918(200006)29:6<403::AID-SIA884>3.0.CO;2-8

[12] L. Jones, T. Beaver, B. Militsyn, S. Mistry, T. Noakes, and R. Valizadeh, "Energy Distribution and Work Function Measurements for Metal Photocathodes with Measured Levels of Surface Roughness," in *Proc. IPAC'17*, Copenhagen, Denmark, May 2017, pp. 1580–1583, doi:10.18429/JACoW-IPAC2017-TUPAB111