SPUTTER GROWTH OF ALKALI ANTIMONIDE PHOTOCATHODES: AN IN OPERANDO MATERIALS ANALYSIS*

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Abstract

Alkali antimonide photocathodes are a strong contender for the cathode of choice for next-generation photon sources such as LCLS II or the XFEL. These materials have already found extensive use in photodetectors and image intensifiers. However, only recently have modern synchrotron techniques enabled a systematic study of the formation chemistry of these materials. Such analysis has led to the understanding that these materials are inherently rough when grown through traditional sequential deposition; this roughness has a detrimental impact on the intrinsic emittance of the emitted beam.

Sputter deposition may provide a path to achieving a far smoother photocathode, while maintaining reasonable quantum efficiency. We report on the creation and vacuum transport of a K_2CsSb sputter target, and its use to create an ultra-smooth (sub nm roughness) cathode with a 2% quantum efficiency at 532 nm.

ROUGHNESS AND EMITTANCE

The Alkali Antimonides are a class of materials (I_3V semiconductors) often used as photocathodes, both in detectors [1] and accelerators [2]. Many of these compounds are capable of achieving a quantum efficiency (QE) of several percent for green light, making them attractive for high average current accelerator applications. The typical sequential deposition process used to form these materials results in a cathode which is very rough (25 nm RMS for a 50 nm thick cathode, with a 100 nm spatial period) [3]. This results in an undesirable field dependence of the intrinsic emittance [4,5]; an emission field of 6 MV/m is sufficient to double the intrinsic emittance of a K_2CsSb cathode illuminated with green light (the expected value should be 0.5 µm/mm [6]). Assuming a simple sinusoidal modulation of the surface height, the field-induced emittance growth can be shown to be given by Eq. (1)

$$\varepsilon_{rough} = \sigma_{x,y}\sqrt{\frac{\pi^2 a^2}{2m_0 c^2 \lambda}Ee} \quad (1)$$

where a is the amplitude, λ is the wavelength of the surface height modulation, σ is the rms beam size and E is the field gradient. Figure 1 shows the effect of roughness using this model for an amplitude of 25 nm and a surface wavelength of 100 nm. These values are typical for a sequential thermal deposition process. We can see that at fields that are relevant to high gradient photo-guns, there would be a drastic increase in emittance. We can also use this same model to detemine the amplitude necessary so that we can achieve room temperature kT defined emittance (0.225 microns / mm rms). We assume that the wavelength of the sinusoidal modulation is always 4 times the amplitude, as often observed in thin film growth. For a field of 20 MV/m, we find that a roughness amplitude of less than 1 nm is required. This is close to the roughness of super-polished etched Silicon,

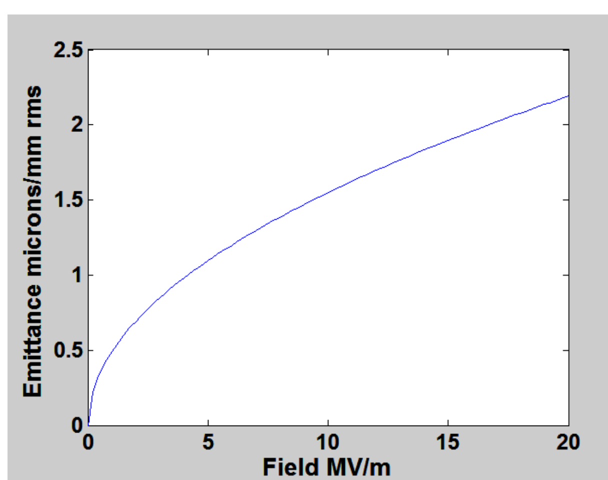

Figure 1: Dependence of emittance on field, for a sinusoidal modulation of the surface with an amplitude of 25 nm and a period of 100 nm.

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and so essentially no roughening from growth of the film can be tolerated.

It is interesting to note that this roughness is intrinsic to the growth process; it occurs even on atomically smooth silicon substrates. The source of this roughening has been identified [7] through the use of *in operando* x-ray analysis during growth. By simultaneously observing the material crystalline structure via x-ray diffraction (XRD) and the roughness via x-ray reflectivity (XRR), it was determined that the conversion of crystalline antimony into an amorphous potassium-antimony compound and then into crystalline potassium antimonide results in a dramatic increase in the roughness of the surface. Indeed, prior to the deposition of potassium, the antimony layer has a roughness comparable to that of the silicon substrate; after the potassium deposition is complete, the cathode roughness is >10 nm (and no reflected spot is observed). Using the same *in operando* tools, several cathode growth methods have been investigated to modify this recrystallization process, with some success in reducing the roughness. The most successful [8] was to use antimony layers which are too thin to be crystalline, growing the cathode in multiple sequential steps (Sb-K-Cs-Sb-K-Cs). In this work, we investigate another approach – the use of sputter deposition to deposit an alkali antimonide film directly from a stoichiometric bulk K_2CsSb sputter target.

SPUTTER TARGET

The K_2CsSb target was fabricated at Radiation Monitoring Devices (RMD) Inc. The K_2CsSb compound was chemically synthesized in bulk using the elemental alkalis (K and Cs) and antimony. A sputtering target measuring 2 inches in diameter was fabricated from the as-synthesized bulk compound. The target was bonded to a metal backing plate and loaded into a Meivac RF sputtering gun. The target was prepared in a controlled environment and was loaded into sputter gun without exposure to ambient moisture or oxygen. The sputtering gun with the target was shipped under vacuum (10nTorr)

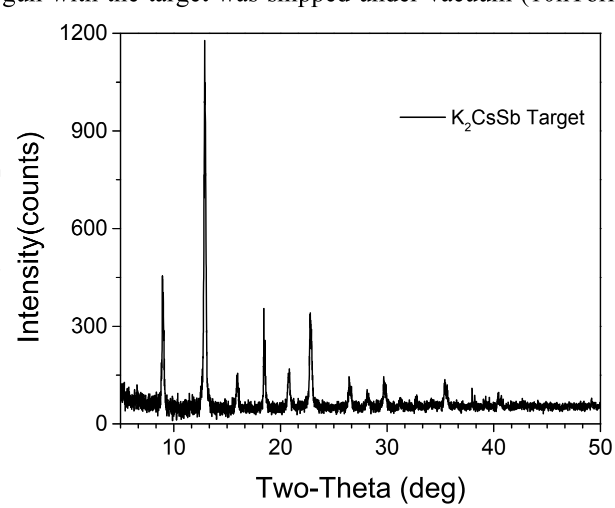

Figure 2: Powder XRD pattern acquired for the target material exhibits cubic phase K_2CsSb.

to BNL/CHESS, where it was attached to the *in operando* photocathode growth chamber. The details of the target preparation and process will be discussed elsewhere.

Figure 2 shows the powder x-ray diffraction (XRD) pattern of the target material. The powder specimen was held in a glass capillary and the background from the glass was subtracted from the diffraction pattern. The diffraction was performed using a Bruker D5000 x-ray diffractometer with Mo Kα radiation source (λ=0.7093Å) and silicon drift detector. The x-ray source was operated at 40 kV and 50 mA. The XRD pattern for the sample shows K_2CsSb crystalline phase, as confirmed by ICDD PDF#03-065-4162 in the JCPDS and NIST Crystal database. The crystallite size was estimated to be 53 nm by the Scherrer model.

SPUTTER GROWN CATHODES

The *in operando* growth chamber described in [7] was modified to mount the sputter source described above. Figure 3 shows the source mounted on the chamber, while Figure 4 shows the sputter target as produced at RMD. High purity argon was bled into the chamber through a dry-ice cold trap, with a static pressure of 20 mTorr. The sputter source was operated at 10 W power to achieve a sputter deposition rate of ~0.2 Å/s, measured both by film thickness monitor and confirmed by XRR. Films were grown on both silicon and MgO substrates. Details of the growth will be presented in a full publication.

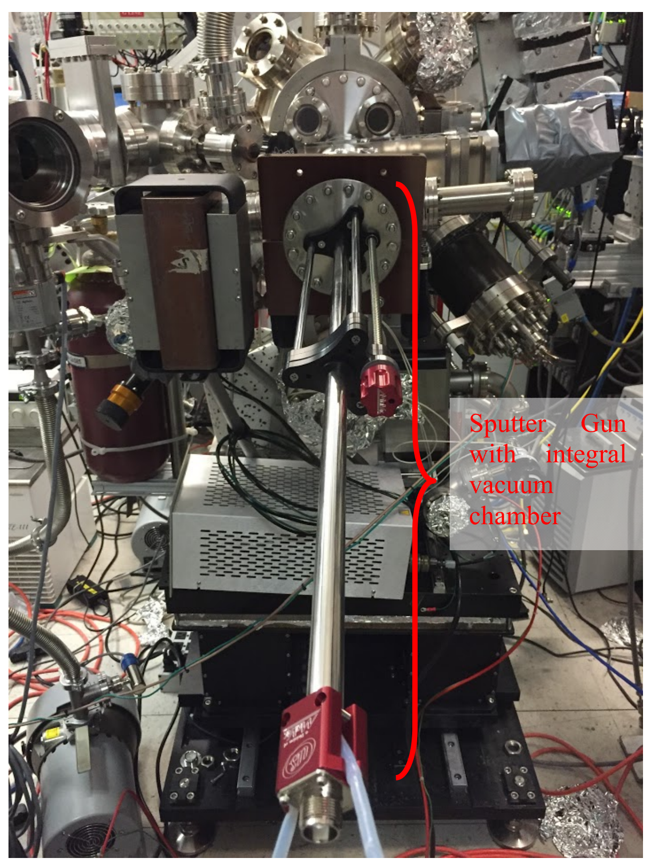

Figure 3: Sputter target mounted on cathode growth and analysis chamber, in G3 line x-ray hutch.

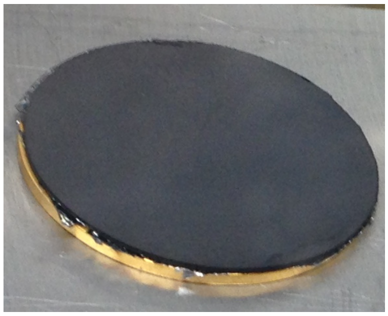

Figure 4: Sputter target bonded to a backing plate. The target was fabricated at RMD Inc.

Figure 5 shows the spectral response of a 50 nm thick film on an MgO substrate. An Energetiq EQ-1500 laser driven light source coupled with an Oriel Cornerstone 130 monochromator provided the optical source for the spectral response. The power as a function of wavelength was measured with a Thorlabs PM100D power meter. The optical system produced a monochromatic 5 mm diameter spot on the sample with a peak power of 100 uW. Glass optical components filtered out UV light below 330 nm (this is the reason the spectral response drops off at this point). Photoelectrons were collected with a biased anode, connected to a Keithley 6517 electrometer.

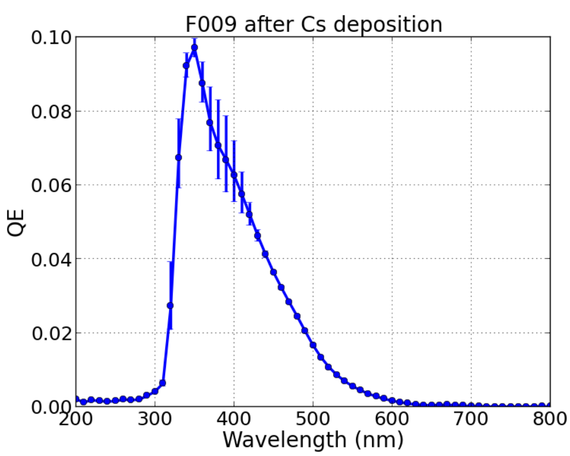

Figure 5: Spectral response of sputtered K_2CsSb cathode after final layer. Cut-off at 330 nm caused by optical source.

The growth was done in 3 steps, with XRR performed between each to measure the roughness evolution as a function of thickness. The XRR data was obtained with 11.3 keV photons, with 2θ angles from grazing to 8 degrees. Data was fitted and refined by GenX[9], which uses the Parratt algorithm as described in [10]. Figure 6 shows the XRR fit of the film. x-ray fluorescence (XRF) from the cathode was measured during the sputter deposition; this showed that the cathode was potassium-poor as compared to a sequentially grown cathode. The actual measured stoichometry was used to calculate the electron density used to fit the XRR data; this was found to be critical for achieving an adequate fit. Table 1 shows the best fit thickness and roughness values; as a point of calibration, the substrate roughness was also measured by AFM (in air) and found to be 2.67Å, in reasonable agreement with the roughness obtained via XRR.

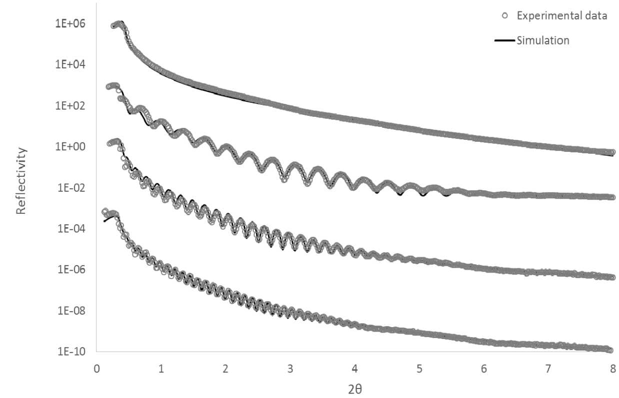

Figure 6: XRR for the substrate (top) and each growth layer (increasing thickness going down on plot), along with the theoretical fits.

Table 1: Best Fit Thickness and Roughness for Each Layer of Sputter Growth

	Thickness (Å)	Roughness (Å)
Substrate MgO		2.03
1st layer	174.9	3.63
2nd layer	334	6.25
3rd layer	512	6.25

CONCLUSION

K_2CsSb has been successfully grown via sputter deposition, on both silicon and MgO substrates. While these cathodes achieved only modest QE (peak QE of 10%, green QE of ~1-2%), the XRR fit to the cathodes suggests a roughness of under 1 nm. A cathode this smooth should have an intrinsic emittance consistent with that expected from its mean transverse energy, even in a 20 MV/m applied field. While significant room remains to optimize the growth processes of these cathodes, sputter growth of alkali antimonides appears to be a path toward much smoother high-QE cathodes.

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