

# PHYSICAL AND CHEMICAL ROUGHNESS OF ALKALI-ANTIMONIDE CATHODES

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## Abstract

Over the last decade, alkali-antimonides have been investigated as high QE cathodes in green light and more recently as ultra-low intrinsic emittance cathodes in near-threshold red wavelengths at cryogenic temperatures [1]. Nano-meter scale surface non-uniformities (physical roughness and chemical roughness or work function variations) are thought to limit the smallest possible emittance from these materials at the photoemission threshold under cryogenic conditions [2]. Despite this, the surfaces of alkali-antimonides have not been well characterized in terms of the surface non-uniformities. Here, we present measurements of surface non-uniformities of alkali-antimonide thin films using several surface characterization techniques like atomic force microscopy, kelvin probe force microscopy, low energy electron microscopy and near-threshold photoemission electron microscopy. This helps show how such non-uniformities limit the intrinsic emittance.

## INTRODUCTION

Thin films of alkali-antimonide materials like K<sub>2</sub>CsSb, Cs<sub>3</sub>Sb and Na<sub>2</sub>KSb have been successfully used as efficient, robust and low-intrinsic-emittance photocathodes in photoinjectors with large beam current requirements [3, 4], making them ideal cathodes for high current applications like Energy Recovery Linacs and high repetition rate Free Electron Lasers. More recently, ultra-small intrinsic emittance as low as 0.20  $\mu\text{m}/\text{mm}$  (rms) with QE greater than  $7 \times 10^{-5}$  has been demonstrated from such cathodes when operated near the photoemission threshold at liquid nitrogen temperatures [1]. The intrinsic emittance is related to the mean transverse energy (MTE) of the emitted electrons through the relation  $\epsilon_n = \sqrt{\frac{\text{MTE}}{mc^2}}$ , where  $\epsilon_n$  is the intrinsic emittance (per unit laser spot size),  $m$  is the rest mass of an electron and  $c$  is the speed of light in vacuum. An intrinsic emittance of 0.20  $\mu\text{m}/\text{mm}$  (rms) corresponds to 17 meV of MTE. Such small intrinsic emittance (or equivalently low MTE) with a reasonable QE can make alkali-antimonides, when operated at the photoemission threshold and at cryogenic temperatures, excellent ultra-low emittance cathodes ideal for low-charge ultrafast electron diffraction (UED) applications.

According to the three step photoemission model [5], the MTE at the photoemission threshold is limited to  $k_B T$ , where  $k_B$  is the boltzmann constant and  $T$  is the electron temperature, which at low laser fluence is equal to the lattice tempera-

ture. This thermal contribution to the MTE should be as low as 8 meV at the liquid nitrogen temperature of 90 K. This is significantly lower than the measured value of 17 meV from Cs<sub>3</sub>Sb cathodes. One possible cause for this discrepancy in the nano-scale physical and chemical surface roughness on alkali-antimonide thin films.

The physical roughness on the surface of cathodes causes the electric field lines close to the surface to bend giving transverse kicks to the emitted electrons, increasing their MTE. The MTE increase due to the physical roughness is proportional to the accelerating electric field at the cathode [2]. The chemical roughness (or work function variation on the surface) causes transverse electric fields close to the cathode surface increasing the MTE. The chemical roughness MTE increase is inversely proportional to the accelerating electric field for high fields (larger than the transverse fields due to chemical roughness), and remains constant for low accelerating fields [6]. Physical and chemical roughness can be present simultaneously on a real cathode surface. The combined effect of these two can result in a more complex dependence of accelerating electric field on the MTE. Calculating this complex dependence requires estimating the electric field close to the surface due to both the physical and chemical roughness and tracking electrons in this field to obtain the MTE. It is imperative to measure the degree and nature of physical and chemical surface non-uniformities and develop alkali-antimonide growth techniques that minimize these surface non-uniformities to obtain the smallest possible MTE.

Physical roughness on alkali-antimonides has been successfully characterized using an ultra high vacuum Atomic Force Microscope (AFM) connected in vacuum to an alkali-antimonide growth chamber [2]. It was shown that the traditional, sequential deposition technique of alkali-antimonide growth produces physical roughness larger than 2.6 nm rms, whereas a co-deposition technique produces a nearly atomically flat film with rms roughness less than 0.6 nm. Calculations of effects of physical roughness on MTE show that for a moderate accelerating electric field of 20 MV/m, the nearly atomically smooth surface can result in an MTE increase of up to 8 meV, comparable to  $k_B T$  at liquid nitrogen temperatures. Hence, growth procedures to obtain even smoother, atomically-flat surfaces of alkali-antimonide need to be developed in order to obtain MTE smaller than a few meV to minimize the cathode emittance.

Measurement and characterization of chemical roughness has proven much more challenging. In principle, a Kelvin Probe Force Microscopy (KPFM) measurement can be per-

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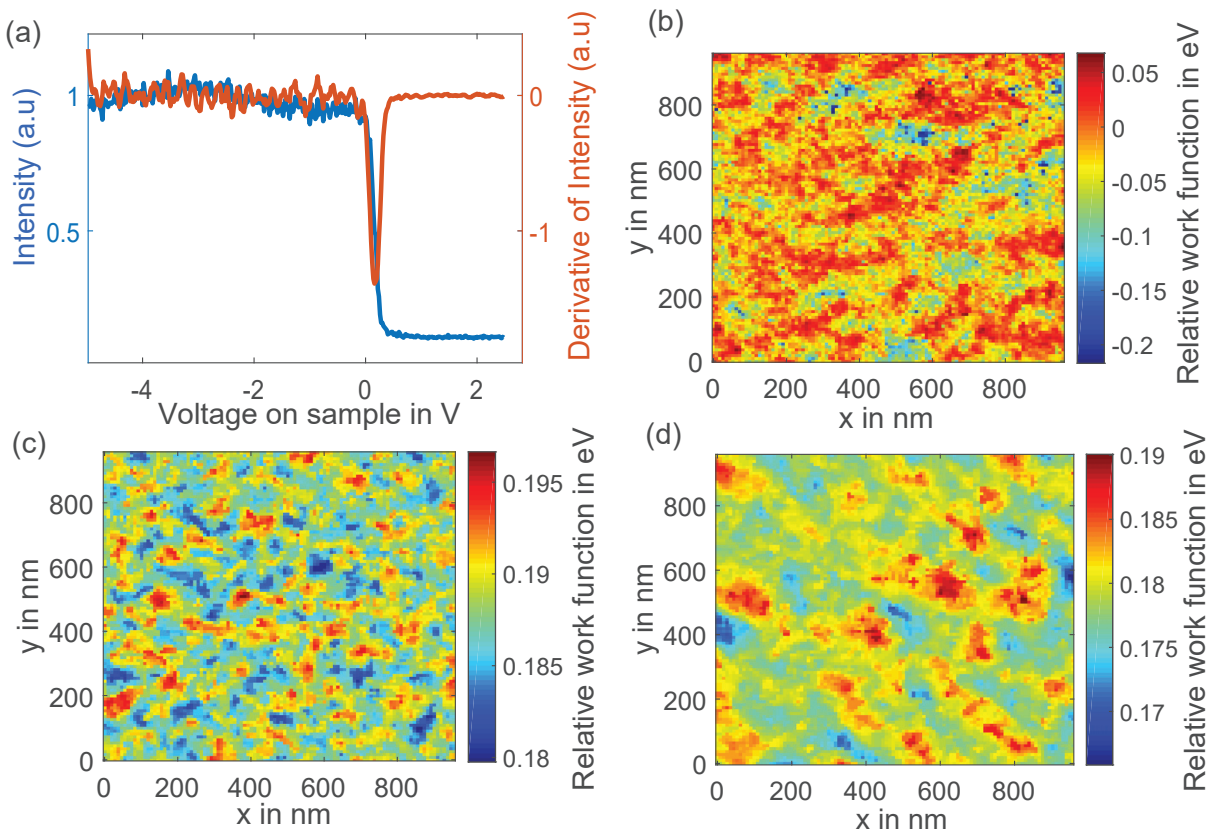


Figure 1: (a) Intensity vs sample voltage (blue) and derivative of intensity vs sample voltage (red) for one pixel in the set of LEEM images recorded at various sample voltages. The minima in the derivative is taken as the relative work function at that pixel. (b) Relative work function map of a Cs<sub>3</sub>Sb cathode grown using sequential growth technique. (c) Relative work function map obtained of a Cs<sub>3</sub>Sb cathode grown using the co-deposition technique with substrate held at 90°C and (d) at 70°C during growth.

formed with an AFM to obtain the topographical and work-function variations on a surface with sub-nm resolutions simultaneously [7]. However, in practice, measurements of work-function variations (or chemical roughness) on alkali-antimonide cathodes have so far proven unreliable due to the volatile nature of these compounds.

In this proceedings, we demonstrate alternative ways to measure the work-function variations on alkali-antimonide thin films using Low Energy Electron Microscopy (LEEM) and PhotoEmission Electron Microscopy (PEEM) and show how different growth recipes can result in dramatically different surfaces of alkali-antimonide films.

## EXPERIMENTAL SETUP AND GROWTH CONDITIONS

The LEEM and PEEM measurements were performed at the spin-polarized low electron microscope (SPLEEM) facility at the National Center for Electron Microscopy, Lawrence Berkeley Lab. The spin-polarized capability of the microscope was not used during this study. Cs<sub>3</sub>Sb samples were grown on Si substrates in a growth chamber connected in vacuum to the microscope. The base pressure in the micro-

scope and the growth chamber was less than 10<sup>-10</sup> torr. Si substrates were heated to 600°C for 1 hour prior to growth for cleaning. For co-deposition based growth, the Sb deposition rate was set to 0.1 Å/s. Cs was evaporated from getter sources manufactured by SAES by passing a current of 5-5.5 A through 5 sources connected in a serial fashion about 5 cm away from the substrate. Deposition was continued until the QE in green light (532 nm) was maximized. For the sequential deposition growth, first a 10-15 nm thick Sb film was deposited followed by evaporation of Cs until the QE maximized. the maximum QE during both co-deposition and sequential growth procedures reached ~2%. Following growth, the samples were transferred into the SPLEEM microscope.

## RESULTS

### Work Function Variation Using LEEM

LEEM measurements were performed on the samples to obtain the work function variation map over the sample surface. Images of the electron beam reflected from the sample are recorded as the bias on the sample is varied from -5 V to 2.5 V in steps of 20 mV. All electrons incident on the

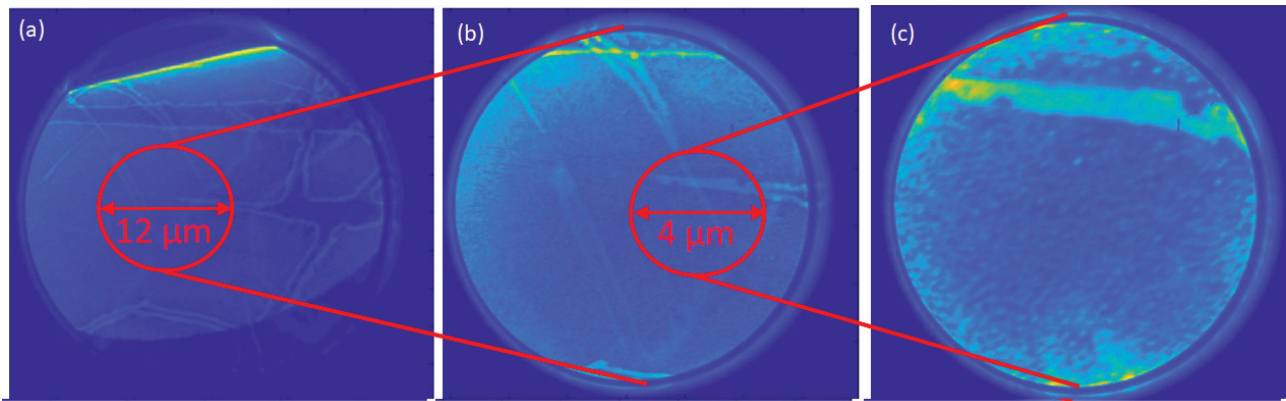


Figure 2: PEEM image (obtained using 690 nm incident light wavelength) of a  $\text{Cs}_3\text{Sb}$  cathode grown using the sequential deposition technique at various length scales. Few 10s of nm scale surface non-uniformities, similar in spatial scale to those observed using an AFM [2], can be seen in the image with the largest zoom. Other images show presence of few micron sized ‘cracks’ on the surface.

sample are reflected at low sample bias voltages, creating a uniform LEEM image. When the voltage applied to the sample is equal to the difference between the work function of the sample and the work function of the electron source (in this case the electron source is a GaAs cathode activated to negative electron affinity), the electrons incident on the sample stop getting reflected and are absorbed into the sample. Due to the work function variation on the sample surface, the LEEM image recorded at this voltage is non-uniform. The work function map of the surface can be calculated by from the non-uniform LEEM images recorded around this voltage. The work function at each pixel (relative to the electron source work function) is given by the point of minimum derivative of intensity vs sample voltage curve of each pixel. This is shown for one pixel in Figure 1a. The pixel wise work function maps of three samples obtained using the above procedure are shown in Figures 1b, c and d. The spatial resolution of the microscope is known to be  $\sim 20$  nm.

Figure 1b shows the work function map of a cathode grown using the sequential deposition process. Figures 1c and d show the work function maps of a cathode grown using the co-deposition process with the substrate temperature held at  $90^\circ\text{C}$  and  $70^\circ\text{C}$  respectively.

The sequential deposition technique produces chemical roughness nearly an order of magnitude larger than the co-deposition technique. This is in-line with the larger physical roughness observed on the sequentially deposited alkali-antimonide films. For cathodes grown using the co-deposition technique, the work function variation shows a strong dependence with substrate temperature during growth. Although the amplitude of variation is comparable for both substrate temperatures ( $90^\circ\text{C}$  and  $70^\circ\text{C}$ ), the periodicity of the variation is much larger for the  $70^\circ\text{C}$  case. A larger periodicity implies lower transverse electric fields reducing the MTE increase due to work function variations.

### Surface Non-uniformities Using PEEM

Work function maps can, in principle, also be obtained using the PEEM technique. Pixel wise Fowler curves can be extracted from the PEEM images taken at several wavelengths close to the photoemission threshold. This, however, requires a bright tunable wavelength monochromatic light source. Detailed work function maps using this technique could not be obtained due to lack of such a source.

Figure 2 shows PEEM images of a sequentially deposited cathode at various length scales using a 690 nm diode laser. The smallest field of view (Figure 2c) of  $4\ \mu\text{m}$  shows roughness at similar spatial scales as obtained from the LEEM measurements. The larger field of views (Figures 2 a, b) reveal a presence of micron scale cracks on the surface. Such larger scale features on cathode surfaces need better characterization in order to determine their origin and their impact on MTE.

In conclusion, we have discussed how AFM, KPFM, LEEM and PEEM can be effectively used to characterized surface non-uniformities on alkali-antimonide cathodes and how different growth procedures and recipes can result in dramatically different non-uniformities. Growth procedures to minimize these non-uniformities need to be developed in order to minimize the emittance from these cathodes.

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