IMPROVING THE SMOOTHNESS OF MULTIALKALI ANTIMONIDE PHOTOCATHODES: AN IN-SITU X-RAY REFLECTIVITY STUDY

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Abstract

Multialkali antimonide photocathodes have been shown to be excellent electron sources for a wide range of applications because of high quantum efficiency, low emittance, good lifetime, and fast response. In recent years, synchrotron X-ray methods have been used to study the growth mechanism of K_2CsSb photocathodes. The traditional sequential growth of K_2CsSb has been shown to result in rough surface, which will have an adverse impact on the emittance of the electron beam. However, co-evaporation of alkali metals on the evaporated Sb layer and sputter deposition may offer a route to solving the roughness problem. Recent studies on K_2CsSb grown by these methods are presented and surface roughness is determined by X-ray reflectivity (XRR) and results are compared.

INTRODUCTION

During recent years, numerous studies have been carried out on the K_2CsSb photocathode as a promising candidate for high quality electron sources [1, 2]. The traditional growth method is to sequentially evaporate Sb, K and Cs onto the substrate. In-situ X-ray analysis on photocathodes grown by this traditional recipe indicates that the surface roughness to be ~ 25 nm rms for a 50 nm thick cathode [2], however a photocathode with this roughness would impact the intrinsic emittance in the high gradient fields used in most applications [3]. Studies have shown that for an emission field of 20 MV/m a roughness of less than 1 nm is required [4].

Previous studies have revealed the growth mechanism of the traditionally grown K_2CsSb photocathode [5, 6]. The evolution of crystalline Sb layer to a K-Sb compound and then to a crystalline K_3Sb significantly increases the roughness from the sub nm scale to more than 10 nm. In order to improve this process, we have developed a method in which multiple thin layers of the antimonide are grown. Sb is deposited in thickness that prevents the formation of crystalline antimony, followed by sequential evaporation of K and Cs [6].

In this work, X-ray reflectivity analysis on photocathodes grown by this method as well as two new approaches, co-evaporation of K and Cs and sputter deposition, will be discussed in details.

EXPERIMENTAL

In-situ synchrotron X-ray growth studies of K_2CsSb photocathodes were carried out at beam line G3, on the Cornell High Energy Synchrotron Source (CHESS), using a photon energy of 11.26 keV. Two Pilatus 100K pixel detectors were installed on the growth system and a Vortex detector was placed about 18 cm away from the sample, so that X-ray reflectivity (XRR), wide angle X-ray diffraction (WAXD), X-ray diffraction (XRD) and X-ray fluorescence (XRF) can be measured during the growth. The base pressure of the chamber during cathode growth was kept in the low 10^{-10} Torr range and partial pressure of water and oxygen were in the low 10^{-11} Torr range. A quartz crystal microbalance (QCM) was positioned alongside the sample holder, and the growth rate of all materials set controlled to be 0.2 Å/s. The temperature of the sample was kept at approximately 110 ℃.

The XRR data is fitted by the refinement program GenX [7], which uses Parratt's recursion and adopts the Nevot-Croce model for the calculation of roughness [8].

Sequential Evaporation

For sequential evaporation experiments, a first layer of 3nm Sb was deposited on a Si substrate, followed by K deposition until the photocurrent maximized. Cs was deposited finally to maximize the photocurrent. A second layer of 5nm Sb was deposited afterwards and the deposition of K and Cs was repeated. X-ray reflectivity analysis was done on the substrate and after each evaporation. Further details can be found in [6].

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Co-evaporation

For co-evaporation experiments, a first layer of 1.5nm Sb, according to QCM, was deposited on the MgO substrate, K and Cs were then co-evaporated onto the Sb layer until the photocurrent reached a maximum. A second layer of Sb followed by co-evaporation of K and Cs was repeated. A final photocathode of 6 layers in total was grown.

Sputter Deposition

A radio-frequency (RF) sputter system was installed in the growth chamber. The K_2CsSb target was synthesized at Radiation Monitoring Devices (RMD), Inc. The detailed description of the sputter experiment can be found in [4]. Sputter deposition was carried out on MgO substrates, grown in 3 layers for a total thickness of 30 nm (sample F009) and on a Si substrate, where the film was grown in one layer of 30 nm thick (sample F010), with final Cs deposition on both samples.

RESULTS

Sequential Evaporation

XRR simulation result for the sample grown by sequential evaporation is shown in Fig. 1 (left). The Si substrate has a roughness determined by fitting the simulation model of 3.1 Å. The first evaporation of Sb has a thickness of 35 Å and a roughness of 2.9 Å. The next step of K evaporation results in a 141 Å thick layer which has 10.5 Å roughness due to the recrystallization process. The cesiation process produces a photocathode that is 5 times as thick as the Sb layer. The fitted roughness for the first layer of cathode is 13.2 Å.

XRR analysis on the final photocathode reveals that the thickness is 469 Å and the roughness is 32 Å, which is approximately an order of magnitude lower than that of the traditionally grown sequential photocathodes. The quantum efficiency at the 532 nm wavelength is measured to be 4.9%.

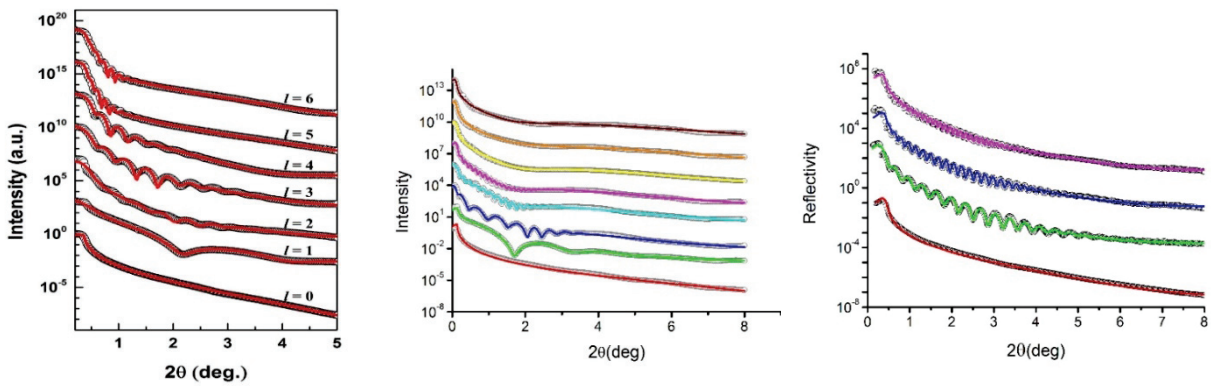

Figure 1: XRR fitting results, simulations are fitted to experimental data. Open circles stand for measured data; Colored curves stand for simulation. Left: Sequential evaporation; Middle: Co-evaporation; Right: Sputter deposition (F009).

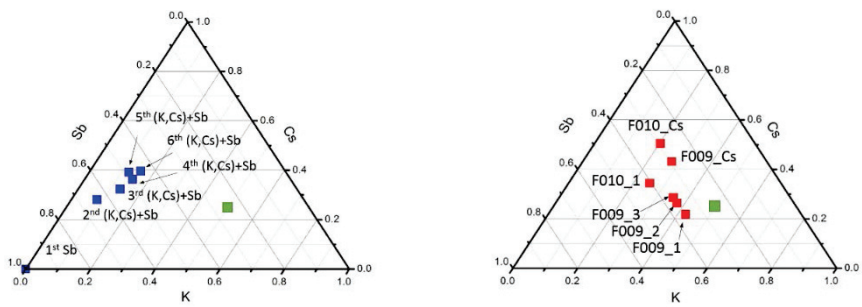

Figure 2: Calculated stoichiometry of each layer of the photocathodes. Left: Co-evaporation sample; Right: Sputtered sample.

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Co-evaporation

With the real-time stoichiometry taken into account, the XRR data can be precisely fitted and the result is shown in Fig. 1 (middle). Figure 2 (left) shows the calculated stoichiometry for each layer of the co-evaporation cathode. For the final cathode, more than enough Sb has been deposited while the amount of K remains deficient while compared to the standard K_2CsSb marked green in the figure.

The first Sb layer shows a thickness of 36.7 Å and 5.2 Å roughness. After evaporating K and Cs, the thickness increases to 159.3 Å and the roughness rises to 9.22 Å. As the growth continues, the surface roughness of this photocathode roughly stays at around 10 Å, while the thickness increases to 489 Å after three layers have been deposited. Further evaporation of Sb and co-evaporation of alkalis doesn't improve the QE substantially and results in a final photocathode with a thickness of 725 Å and 24.9 Å roughness. The final QE at a wavelength of 532 nm is 4.5 %.

Sputter Deposition

The fitted XRR data with stoichiometry taken into account is shown in Fig 1 (right). From the calculated stoichiometry for sputtered cathode shown in Fig. 2 (right) it can be concluded that the sputtered cathode is slightly deficient in K compared to the standard stoichiometry. It can be observed from the curve that the surface roughness become relatively small as demonstrated by the fact that there are intensity oscillations up to a very high angle, showing the coherence of the substrate and surface interfaces . The surface roughness for sample F009 after 3 layers of sputtering is about 6.87 Å and the thickness is fitted to be 512 Å. XRR fitting result on sample F010 after Cs deposition reveals that the surface roughness is about 7.56 Å, with a final thickness of 776 Å. The simulated electron density which is related to the model used for the best fit indicates that the sputtered layer has a uniform density. The spectral response measurement for two sputtered cathode shows the quantum efficiency of both cathodes is approximately 1% at 532 nm wavelength. Figure 3 shows the comparison of surface roughness vs. thickness between photocathodes grown by these three growth methods.

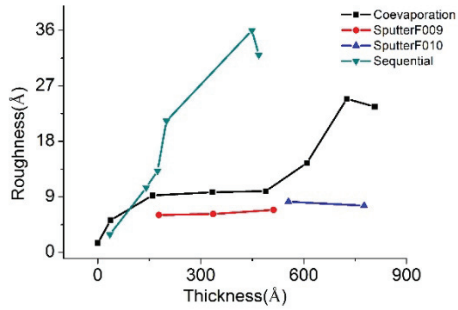

Figure 3: Comparison of surface roughness vs. thickness between photocathodes grown by sequential evaporation, co-evaporation and sputter deposition.

CONCLUSION

In order to produce a CsK_2Sb photocathode with high quantum efficiency and a smooth surface, it is essential to prevent the initial Sb layer from crystallization. The photocathode grown by alternating deposition of a thin Sb layer and alkali metals has been verified to result in a smoother surface than obtained by sequential deposition. A series of experiments on alternating deposition of Sb followed by sequential evaporation and co-evaporation of alkali metals have been carried out. XRR analysis show that surface roughness of these photocathodes is approximately an order of magnitude lower than sequentially produced photocathodes, with acceptable quantum efficiency of about 4% at 532 nm wavelength.

Sputter deposition can result in an even smoother photocathode with sub nm surface roughness according to the XRR analysis but with a lower quantum efficiency of 1% at 532 nm wavelength due to loss of K.

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REFERENCES

[1] A. H. Sommer, Photoemissive Materials: Preparation, Properties and Use (John Wiley & Sons Inc., 1969).
[2] T. Vecchione et al., APL, vol 99, 034103, 2011.
[3] S. Schubert et al., APL Materials 1, 032119 (2013).
[4] T. Vecchione et al., Proc. of IPAC12, 655 (2012).
[5] J. Smedley et al., "Sputter growth of alkali antimonide photocathodes: an in-operando materials analysis", IPAC'15, Richmond, USA (2015).
[6] M. Ruiz-Osés et al., APL Materials 2, 121101 (2014).
[7] J. Xie et al., "Synchrotron X-ray study for a low roughness and high efficiency K_2CsSb photocathode during film growth", manuscript submitted, (2015).
[8] J. Appl. Cryst. (2007). 40, 1174–1178.

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