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PULSED LASER DEPOSITION ASSISTED GROWTH OF ALKALI-BASED PHOTOCATHODES*

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Abstract

Alkali-based semiconductor photocathodes are widely used as electron sources and photon detectors. The properties of alkali-based semiconductor materials such as crystallinity and surface roughness fundamentally determine the performance merits such as quantum efficiency and thermal emittance. At Brookhaven National Laboratory, pulsed laser deposition (PLD) was utilized to assist the growth of alkali-based photocathode materials, providing precise control of material growth and improving film quality. In the presented work, films prepared with thermal and PLD sources are compared. The film quality of K_2CsSb, Cs_3Sb and Cs_2Te grown with PLD-assisted technique is reported.

INTRODUCTION

Alkali-based semiconductor photocathodes are crucial components in various applications such as photodetectors, solar cells, and electron sources in devices like free-electron lasers (FELs) and electron microscopes [1, 2]. The properties of alkali-based photocathodes, particularly crystallinity and surface roughness, play fundamental roles in the performance merits such as quantum efficiency and thermal emittance. In recent years, advancements in material science, thin-film deposition techniques, and surface engineering have driven the development of high-performance photocathodes. Epitaxial growth of alkali-based photocathodes is achieved on lattices-matching substrates with controlled growth techniques like molecular-beam epitaxy (MBE) [3]. Atomically smooth cathode material like Cs_3Sb is achieved on substrates like $SrTiO_3$ [4].

Pulsed laser deposition (PLD) involves focusing a high intensity pulsed laser beam on the source material and converting it into plasma, usually in specific inert gas environment or under high vacuum conditions. The plasma plume ejects from the target, and deposits to a substrate as thin films [5]. PLD provides high purity deposition with controlled growth parameters, as well as uniform thickness for thin film growth. These advantages in material synthesis make it a versatile technique widely used in materials research and the semiconductor industry [6].

At Brookhaven National Laboratory (BNL), PLD was utilized to assist the growth of alkali-based photocathode materials, providing precise control of film thickness, improving film surface morphology and crystallinity. In this work, we discuss the merits of the growth technique with experimental support. The film quality of K_2CsSb, Cs_3Sb and Cs_2Te grown with PLD-assisted technique are presented.

EXPERIMENTAL

The ultra-high vacuum (UHV) cathode synthesis and x-ray analysis chamber is installed at the integrated *in situ* and resonant hard x-ray studies (ISR) beamline located at NSLS-II, BNL, where the thin film deposition is monitored *in situ* and *operando* by various x-ray characterization techniques. The x-ray geometry and experimental set-up are detailed elsewhere [7, 8]. The schematic of the PLD set up at the beamline is shown in Fig. 1. A KrF laser at 248 nm is brought into the chamber through a set of focusing optics and a fused silica viewport. Alignment laser along with a CCD camera are installed to align and focus the laser to the PLD target. The average pulse energy is 180 mJ/pulse measured at the laser output. The pulsing of the laser is controlled by a digital delay generator (Stanford Research Systems DG535). During the experiment, a frequency of 0.5 ~ 1 Hz is used for deposition. The Sb and Te metal targets are commercially purchased and mounted approximated 10 cm from the substrate. The targets are installed on a rotary manipulator which rotates at a constant speed in order to provide fresh ablation surface and constant rate during film deposition. The alkali metals (K/Cs) are supplied with an effusion cell assembly (thermal source). The incident x-ray beam is 11.48 keV (λ = 1.08 Å) with a nominal beam diameter of 50 μm. Reflectivity scans in Fig. 2 and diffraction scans in Fig. 3 are collected with two Dectris Eiger R 1M cameras mounted on-axis (75 cm away from the sample) and 30° off-axis (27 cm away from the sample) with respect to the sample plane, respectively.

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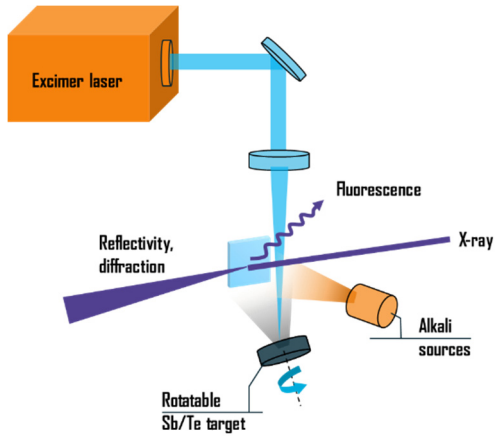

Figure 1: Experimental set-up with PLD at ISR, NSLS-II. Schematic of the experimental geometry and characterization tools are detailed in W.G. Stam et al. and C. Pennington et al. [7, 8].

RESULTS AND DISCUSSION

The intensity of the reflected x-ray from the sample is recorded during film growth, which typically exhibits oscillating behaviour as film thickness increases. The change in intensity generally attributes to the change in the film surface roughness [7]. Fig. 2 compares the growth oscillation plots from the Cs_3Sb films deposited using thermal and PLD Sb sources, respectively. Other growth parameters including substrate temperature and Cs rate are kept the same. In Fig. 2a, 22 nm of Cs_3Sb film is deposited on a 4H-SiC (0001) substrate using co-evaporation of thermal Sb and Cs sources. An initial drop of intensity from the reflected x-ray beam is observed before the growth oscillation starts, which could suggest unstable deposition during source thermalization that roughens the sample surface. Measures including adding a sample/source shutter as well as thermal regulation in the configuration of the Sb source could mitigate such effects. Fig. 2b shows the reflectivity curves from two Cs_3Sb layers grown successively on a

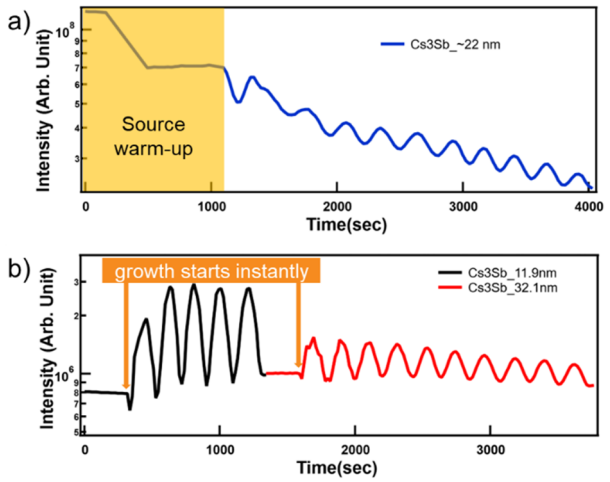

Figure 2: Growth oscillations of Cs_3Sb (top) on 4H-SiC with Sb thermal source and (bottom) on Gr/4H-SiC with Sb PLD target.

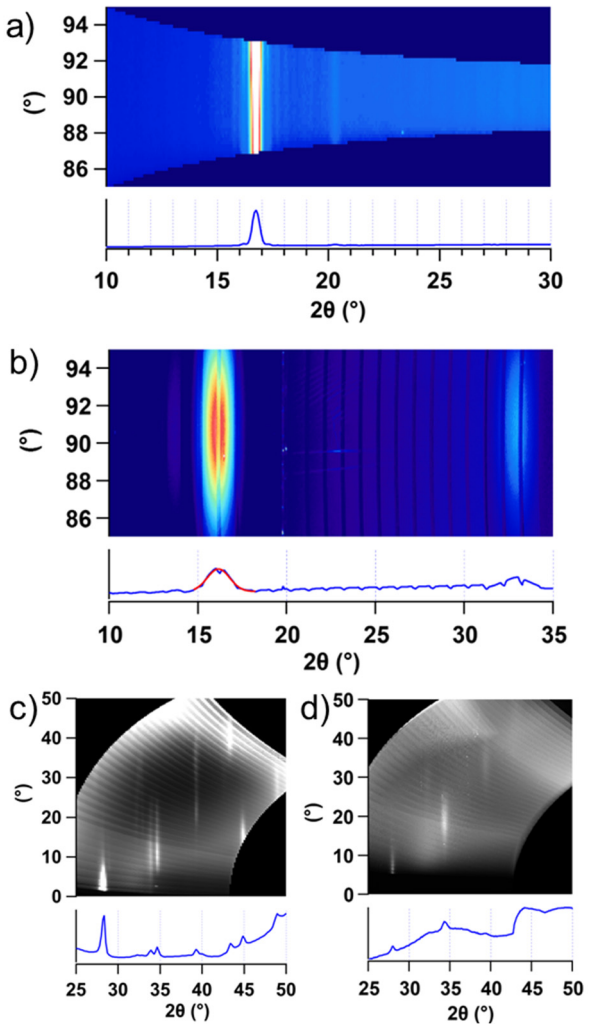

Figure 3: XRD 2θ scans of Sb film deposited on Si (100) substrates from a) c) 20 nm Sb film deposited with thermal Sb source and b) d) 4.4 nm Sb film deposited with PLD Sb target.

Gr/4H-SiC substrate. Growth oscillations are found to start and turn off instantly with the PLD laser switching on and off, suggesting that PLD-assisted growth can be a precisely controlled process for film growth. No degradation is observed for the reflectivity intensity during each film growth in Fig. 2b, indicating less surface roughening is introduced as the film thickness increases.

On substrates with no lattice-matching conditions, films grown by PLD could be more textured than films from thermal sources. The XRD *2θ* scans of Sb films deposited on Si (100) substrates with thermal and PLD Sb sources are compared in Fig. 3. The Sb film deposited with the thermal source is the first step of a sequential process in a cathode growth. The target thickness is 20 nm, with a substrate temperature held at ~ 100°C. The film deposited using the PLD Sb target is prepared at a substrate temperature of 80°C, with a film thickness of 4.4 nm (estimated from XRR). Lowered growth temperature and less film thickness are typically considered less favorable for the crystallization of a Sb film [11]. However, the diffraction arcs in Fig. 3b and 3d, although broadened due to lower thickness, appear to

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be more discontinued than the thicker films in Fig. 3a and 3c, indicating that the Sb film prepared using PLD is more structurally aligned.

Table 1: Comparison of Roughness for PLD-assisted Growth of Cathode Materials vs Literature

Cathode Material	Preparation Method	Thickness (nm)	Roughness (Å)
Cs_3Sb [7]	PLD+Cs	11.3	3.0 (XRR)
K_2CsSb [9]	PLD+K/Cs	11	6.0 (XRR)
Cs_2Te [10]	PLD+Cs	21	8.0 (XRR)
Cs_3Sb [4]	Thermal co-dep	20~60	4~6 (AFM)
K_2CsSb [12]	Thermal co-dep	30~40	6 (AFM)
Cs_2Te [13]	Thermal co-dep	24	10 (XRR)

We tested several popular alkali-based photocathode materials in the current accelerator applications with PLD assisted growth technique. The details of preparation are described elsewhere [7-10]. It is found that atomically smooth cathode films are obtained in our study. Table. 1 lists the roughness obtained for the three types of cathode material in our experiments, along with the roughness values for the same cathode material found in literature. It is worth noting that the roughness obtained in our experiments are estimated from the XRR measurements, which averages over the entire length of the sample film, with an estimated sampling area of mm in size. From the comparison in Table. 1, we can see that for all the cathode materials listed, the films prepared with the PLD-assisted growth technique matches the roughness record in literature, with the Cs_2Te films improved from the polycrystalline film with similar thickness prepared with thermal Sb sources. The improvement in the photocathode film roughness is crucial in the development of high-brightness electron sources for electron accelerator applications like x-ray free electron laser and the electron microscope.

CONCLUSION

In conclusion, PLD is utilized to assist the growth of alkali-based photocathode materials, providing precise control of film thickness and improving film in crystallinity and surface roughness. It is found that on substrates with no lattice-matching conditions, films grown by PLD could be more textured than the film from thermal sources. PLD assisted growth of K_2CsSb, Cs_3Sb and Cs_2Te are found to yield atomically smooth cathode films that either match or improve from the roughness record in literature, rendering such technique a promising development in photocathode material engineering.

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