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EPITAXIAL ALKALI-ANTIMONIDE PHOTOCATHODES ON LATTICE-MATCHED SUSBTRATES

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Abstract

Alkali-antimonides photocathodes, characterized by high quantum efficiency (QE) and low mean transverse energy (MTE) in the visible range of spectrum, are excellent candidates for electron sources to drive X-ray Free Electron Lasers (XFEL) and Ultrafast Electron Diffraction (UED). A key figure of merit for these applications is the electron beam brightness, which is inversely proportional to MTE. MTE can be limited by nanoscale surface roughness. Recently, we have demonstrated physically and chemically smooth Cs_3Sb cathodes on Strontium Titanate (STO) substrates grown via co-deposition technique. Such flat cathodes could result from a more ordered growth. In this paper, we present RHEED data of co-deposited Cs_3Sb cathodes on STO. Efforts to achieve epitaxial growth of Cs_3Sb on STO are then demonstrated via RHEED. We find that films grown via "deposition-recrystallization" method on substrates like STO and SiC (previously used to achieve single crystalline Cs_3Sb) exhibit QE higher than the polycrystalline Cs_3Sb cathodes, by an order of magnitude below photoemission threshold. Given the larger QE, lower laser fluence could be used to extract high charge densities, thereby leading to enhanced beam brightness.

INTRODUCTION

Alkali-antimonide photocathodes like Cs_3Sb, Na_2KSb, K_2CsSb have emerged as efficient electron sources, capable of driving a wide spectrum of linear accelerator based applications ranging from Energy Recovery Linacs (ERL), electron cooling of hadron beams to X-ray Free Electron Lasers (XFEL), Ultrafast Electron diffraction (UED) experiments. They satisfy to quite a good extent the various conflicting requirements for aforementioned applications, which include high Quantum Efficiency (QE) and low Mean Transverse Energy (MTE)/intrinsic emittance in the visible wavelengths , in addition to exhibiting relatively long lifetimes and sub-picosecond response times [1].

Electron beam brightness B which is a key figure of merit for such applications, scales inversely with the MTE as follows [1]:

$$B \propto \frac{1}{\mathrm{MTE}}. \quad (1)$$

The MTE is not just a material intrinsic property, but is also characteristic of the surface geometry. It can get limited by the nanoscale surface inhomogeneties, both topographical and chemical variations, arising due to the nature of the growth process of alkali-antimonide photocathodes.

Traditionally alkali-antimonides were grown as polycrystalline films with 100 nm tall features using sequential deposition. [2]. Co-deposition of alkali-antimonides has resulted in flatter cathodes, with reduction in roughness by almost an order of magnitude [3]. Efforts to grow atomically flat cathodes alkali antimonides have been pursued by growing on lattice-matched substrates such as silicon carbide 3C-SiC [4] and strontium titanate (STO) substrates [5] instead of the standard Si or polycrystalline metal substrates. It has been shown that high QE Cs_3Sb cathodes grown on STO were atomically flat with rms roughness $\sim$ 0.3 nm and chemically homogeneous with $\sim$ 2.5 mV rms roughness. These ultra smooth cathodes could result from a more ordered and possibly, epitaxial growth.

In this paper, we first report on the growth and Reflection High Energy Electron Diffraction (RHEED) data of co-deposited Cs_3Sb cathodes on STO. We then demonstrate our preliminary efforts to grow Cs_3Sb cathodes on STO via deposition-recrystallization technique, which has been used to achieve single crystalline Cs_3Sb cathode on SiC [6]. We compare the spectral response of QE data from Cs_3Sb cathodes grown via co-deposition and deposition-recrystallization technique, and attempt to explain the difference in QE profiles from such cathodes below photoemission threshold $\sim$ 2.1 eV.

GROWTH

Several Cs_3Sb cathodes were grown on 10 mm by 10 mm Nb-doped STO substrates purchased from MTI Corp. The substrates were mounted on stainless steel omicron paddles compatible with the sample holder of the MBE chamber at Cornell University. A thin indium foil was used between the substrate and omicron paddle to provide better thermal and electrical contact . Prior to growth, the substrates were annealed at 600 °C for two hours. The pristine, single crystalline nature of the STO substrate was confirmed by observing the diffraction spots shown in Fig. 1, by RHEED.

Cs_3Sb cathodes were grown by standard co-deposition technique using pure metallic Sb pellets and Cs-In alloy as sources, placed in effusion cells equipped with pneumatic shutters. The sources were pre-calibrated in terms of flux rates with a quartz crystal microbalance at the growth position of the substrate.

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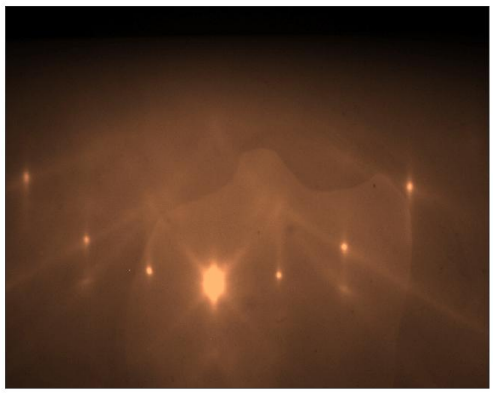

Figure 1: RHEED pattern observed from a clean, annealed STO substrate.

During growth, the Cs and Sb sources were heated at 264°C and 359°C respectively, which corresponds to 2 x 10^{13} atoms/cm^2/s and 3 x 10^{12} atoms/cm^2/s, while the substrate temperature was maintained at 75°C.

The photocurrent emitted by the cathode under illumination of a 532 nm diode laser, was measured to monitor the evolution of QE during growth. A negative bias of 30 V was applied to the sample holder w.r.t the walls of the chamber and the photocurrent was measured using a lock-in amplifier. When the QE began to plateau/drop, growth was terminated by cooling down both the sources simultaneously as well as the substrate heater. The cathodes had a final QE of 3.5% in green after 1.5 hours of co-deposition.

Deposition-recrystallization technique which was developed to achieve single crystal Cs_3Sb cathodes [6], was also attempted to grow Cs_3Sb cathodes epitaxially on STO substrates. Thin films were grown on STO substrates via MBE, using a sequence of shuttered growth of three unit cells (u.c.) at a substrate temperature of 55°C and annealing at 100°C for recrystallization to occur. During the growth phase, Sb was deposited in twelve doses of 1/4 nominal monolayers per STO unit cell, separated by a 20 s pause, while Cs was continuously provided. The sample was then heated to the recrystallization temperature in vacuum, annealed for 15 mins in Cs flux at 100°C and cooled back to the growth temperature in Cs flux. Each such cycle produces 3 u.c. of Cs_3Sb, within the 15% uncertainty of the quartz-crystal microbalance measurement of the Sb flux. This growth process is structure oriented in nature and the sample was continuously studied under the RHEED to monitor the quality of the film.

RHEED MEASUREMENTS

RHEED measurements were performed a couple of times during the growth of Cs_3Sb cathode via co-deposition as well as after the cathode was cooled down to the room temperature. Lack of any RHEED pattern suggests that the cathode was not ordered and was probably still amorphous at the top few layers. This does not eliminate the possibility of the bulk being well-ordered and crystalline. However information on that cannot be obtained via RHEED measurement, which is limited to only a few monolayers at the cathode surface.

During the growth process via deposition-recrystallization technique, RHEED meausurements were continuously performed. During the first iteration of 12 cycles of quarter monolayer Sb deposition in the presence of Cs flux, the diffraction pattern corresponding to bare STO substrate became very dim until it completely disappeared. After the second iteration of Sb deposition, a very fuzzy blob started to appear (see Fig. 2), which may indicate the onset of some order in the lattice at the top few monolayers. The faint pattern persisted during subsequent annealing at higher temperature and Sb deposition at growth temperature. However, the pattern was too dim and fuzzy for us to extract any real information on the crystal structure of the cathode.

Figure 2: Fuzzy RHEED pattern observed from Cs_3Sb cathodes grown on STO substrate via deposition-recrystallization method. The fuzzy features have been marked out using rectangular textboxes.

SPECTRAL RESPONSE MEASUREMENTS

Post growth, Cs_3Sb cathodes were transported in vacuum from the growth chamber to the storage chamber, where the spectral response measurement of QE was performed. A Newport monochromator was used as the light source to measure the QE from the Cs_3Sb cathodes over wavelengths ranging between 400-700 nm.

Figure 3 shows the spectral response data of QE obtained from co-deposited Cs_3Sb cathode on STO (red curve), nearly epitaxial Cs_3Sb cathode on STO (blue curve) and epitaxial Cs_3Sb cathode on SiC (black curve) grown in the MBE chamber in the PHOtocathode Epitaxy and Beam Experiments (PHOEBE) laboratory at Cornell University. The points marked in green represent the QE measured from single crystalline Cs_3Sb cathodes reported in [6].

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All these plots show a knee around 600 nm/2.1 eV, which is typically where the photoemission threshold of such cathodes lies. There is a sharp contrast in profile between the co-deposited Cs_3Sb cathodes and the cathodes grown via deposition-recrystallization technique. In case of co-deposited Cs_3Sb cathodes, below threshold the QE drops very sharply with wavelength. On the contrary in case of Cs_3Sb cathodes grown via deposition-recrystallization technique, the drop in QE below threshold is much gradual with respect to wavelength.

The difference in QE profile between the cathodes grown via two different techniques can be attributed to impurity/defect states which arise due to the growth process itself.

The deposition-recrystallization growth technique forces the film to take up the lattice structure of the substrate, leading to build up of strain at the substrate-film interface. In case of large lattice mismatch, interface dislocations are often formed to relieve the lattice strain between a film and substrate, resulting in impurity/defect states in the bulk. Films grown via deposition-recrystallization method on nearly lattice matched substrates may end up having more defect states within the band gap energy than polycrystalline Cs_3Sb cathodes. The gradual decay of QE below threshold in that case is due to emission of electrons from these large number of defect states, with enough energy to be photo-excited above threshold.

There is a possibility of other phenomena for example work function modulation due to interface/surface effects, or changes in band structure induced by strain which could also explain the higher QE observed beyond threshold.

The QE spectral response obtained from Cs_3Sb cathodes grown via deposition-recrystallization method on STO, resembles the QE profile measured from single crystalline Cs_3Sb cathodes on SiC grown at PARADIM facility, very closely. Identical QE spectral response profiles suggest that the Cs_3Sb cathodes grown on STO via "deposition-recrystallization" might be nearly as ordered in the bulk as was the first single crystalline Cs_3Sb cathode ever grown and reported in [6].

The prospect of alkali-antimonide photocathodes grown via deposition-recrystallization technique yielding higher QE than polycrystalline cathodes near threshold by almost an order of magnitude is highly promising. In case of epitaxial Cs_3Sb cathodes because of higher QE, lower laser fluence can be used to extract charge densities required for high charge applications. Otherwise, in order to extract the same charge density, larger laser fluence would have to be used which often causes MTE degrading effects like laser-induced heating and multi-photon photoemission and brightness gets limited/compromised.

CONCLUSION AND FUTURE WORK

We have presented our preliminary data on efforts towards epitaxial growth of alkali-antimonide photocathodes on STO substrate in this paper. Future work is underway to: (a) Optimize the growth parameters to achieve epitaxial growth of single crystalline Cs_3Sb cathodes on STO substrate, (b) To measure MTE from such cathodes, and (c) To eventually use these cathodes to grow heterostructures of alkali-antimonides.

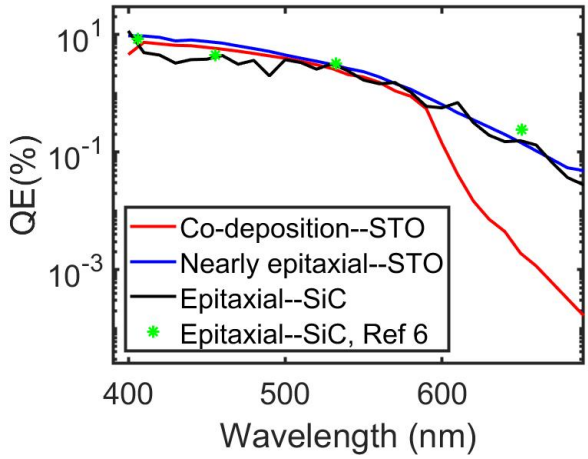

Figure 3: Spectral response of QE from Cs_3Sb cathodes grown via (a)co-deposition on STO, (b) deposition-recrystallization technique on STO (c) deposition-recrystallization technique on SiC substrate, and (d) deposition-recrystallization technique on SiC reported in reference [6].

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