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MOLECULAR BEAM EPITAXIAL GROWTH OF SODIUM ANTIMONIDE PHOTOCATHODES *

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Abstract

Cornell University has been working on developing techniques to grow single-crystal photocathodes for electron sources using the molecular beam epitaxy (MBE) technique. As a result, the first single-crystal Cs_3Sb photocathode was produced, which has shown high quantum efficiency and is expected to have a low mean transverse energy (MTE). Currently, other alkali materials are being explored. In this work, we report the epitaxial growth of Na-Sb photocathodes at the PHOtocathode Epitaxy Beam Experiments (PHOEBE) laboratory at Cornell University.The photocathodes were characterized through quantum efficiency (QE) measurements and reflection high-energy electron diffraction (RHEED) patterns collected during growth. The RHEED streaky pattern shows angle dependence, confirming their epitaxial formation. Notably, these Na-Sb photocathodes exhibited a QE exceeding 1% at 400 nm, which is much higher than previous reports on this compound. The possible reasons for this discrepancy are discussed.

INTRODUCTION

Multi-alkali antimonide photocathodes are good candidates for high-brightness electron sources due to their high quantum efficiency (QE) and low thermal emittance [1, 2]. Among these, potassium-based compounds like K-Cs-Sb have historically received the most attention. At Cornell University, our recent work has focused on the molecular beam epitaxy (MBE) growth of single-crystal alkali-antimonide photocathodes as a way to achieve thin, highly ordered films with high performance, instead of the sequential method typically used to grow these films. Since sequential deposition normally leads to high QEs, the original aim of this work was to establish the Na-Sb photocathode as a structural base layer for the subsequent deposition of potassium, thus enabling the fabrication of high-performance K-Na-Sb photocathodes.

Surprisingly, during this process, we discovered that Na-Sb photocathodes alone exhibited unexpectedly high QE, surpassing previously reported values for this material. This finding varies from our initial expectations, as Na-Sb was primarily intended to play a structural role rather than contribute significantly to photoemission. In this paper, we report the epitaxial growth of Na-Sb photocathodes using both single-temperature and two-step deposition methods. We analyze the resulting film quality using reflection high-energy electron diffraction (RHEED) and present QE measurements that highlight the unique properties of these films. The implications of these findings are discussed in the context of ongoing efforts to develop high-performance photocathode materials.

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EXPERIMENTAL DETAILS

All photocathodes were grown and characterized at the PHOtocathode Epitaxy Beam Experiments (PHOEBE) laboratory at Cornell University [3]. The detailed procedure for fabricating alkali-antimonide photocathodes via MBE is described in [3, 4]. Briefly, thermal evaporation of alkali metals and antimony is achieved using effusion cells under ultra-high vacuum conditions. QE measurements and RHEED imaging were performed in situ during growth to assess film quality and crystallinity.

This study compares two growth approaches. In the single-temperature growth method, the substrate is maintained at a constant temperature throughout deposition. In contrast, the two-step deposition method begins with film growth at a lower temperature (90 C), followed by annealing to 135 C to promote crystallization. After annealing, the film is cooled, and the cycle is repeated as needed to achieve the desired film structure. Upon completion, samples are transferred to a storage chamber for subsequent spectral response characterization.

RESULTS

Single Temperature Growth

The source fluxes were adjusted to achieve a Na:Sb ratio of 3:1. Deposition was carried out on Si(100) substrates, which impose a -2% lattice strain on Na3Sb films. Figure 1 shows the spectral response of the initial samples (001 and 002), which were grown by MBE at a constant temperature. The peak values at 400 nm exceed those previously reported by Sommer [5].

The RHEED patterns, shown in the insets of Fig. 1, display spotty rings characteristic of rough, polycrystalline films. To optimize growth conditions, the substrate temperature was varied during deposition. It was observed that lower temperatures enhance QE, while higher temperatures result in thinner films with less pronounced spotty features. These findings suggest that a two-step growth process may provide a more effective route for fabricating high-performance Na-Sb photocathodes, as will be discussed in the following section.

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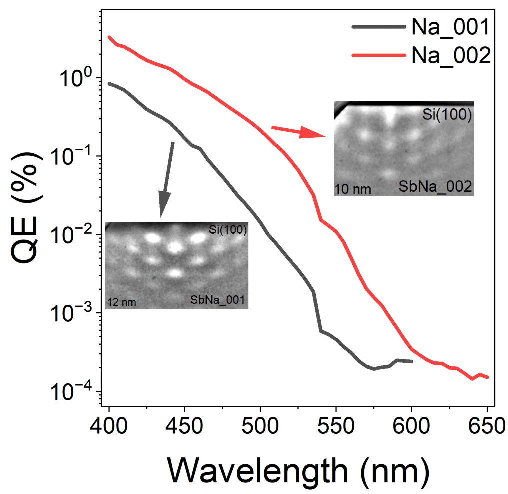

Figure 1: Spectral response for samples 001 and 002 using a single temperature deposition. RHEED Images of each sample are shown in the insets.

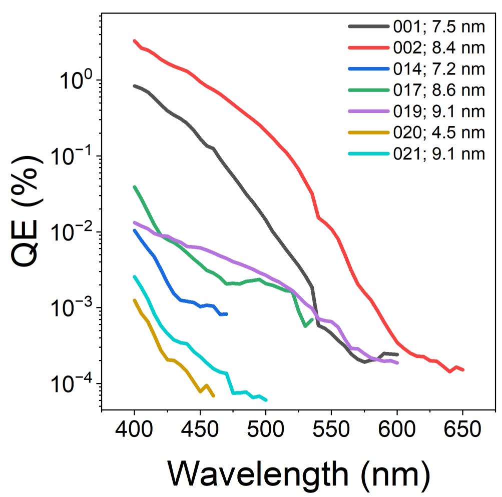

Figure 2: Spectral response for other photocathodes grown at a single temperature. The thickness of each cathode is shown in the legend.

To explore the high QEs presented in these cathodes, more single-temperature growths were performed. The spectral response of these new cathodes is shown in Fig. 2. Although the results show cathodes with different photoemission thresholds, all the QE values are above those reported by Sommer [5].

Partial pressures of other alkali elements were monitored right before growth. Cesium levels range from 3E-12 Torr to 3E-9 Torr, while potassium levels were between 1E-11 Torr and 5E-10 Torr. However, no clear correlation was observed between these background levels and the variations in photoemission threshold. Further research is ongoing to investigate the mechanisms behind these unexpectedly high QE values, although this lies beyond the scope of the present proceedings.

Two-Step Deposition Method

To promote improved crystallization of the Na-Sb films, a two-step deposition process was employed (Fig. 3), as detailed in the Experimental section. The initial deposition was performed at 90 C for 20 minutes, followed by a gradual increase in substrate temperature up to 135 C, during which the source shutters remained open. Upon reaching 135 C, the shutters were closed, and the sample was annealed at this temperature for 10 minutes. The temperature was then slowly reduced back to the initial deposition temperature. This cycle was repeated twice to produce a film approximately 11.6 nm thick.

The RHEED patterns of sample 010 at each stage of the process are shown in Fig. 3. After the first deposition step, faint streaks were observed, which became progressively brighter as the deposition proceeded. Other samples (not shown here) exhibited significant narrowing of the streaks and a notable reduction in surface roughness. However, this improvement was less evident in film 010, as it already exhibited a well-defined RHEED pattern from the beggining of the deposition.

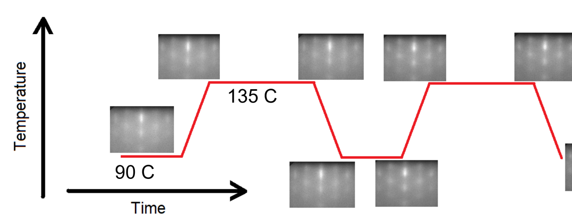

Figure 3: Graphical description of the two-step deposition. Images of the RHEED at each stage for sample 010 are also shown.

Sample 010 also demonstrated angle-dependent RHEED features. Figure 4a presents the intensity profiles at different azimuthal angles, revealing variation in streak intensity as a function of angle. This angular dependence is indicative of epitaxial growth—a finding reported here for the first time in Na-Sb photocathodes.

The spectral response of sample 010 is shown in Fig. 4b, together with the responses of samples 001 and 002 for comparison. Although the growth processes differ, all samples exhibit comparable quantum efficiency (QE), with peak values on the order of a few percent.

While the QE values for Na-Sb photocathodes grown via both single-temperature and two-step processes are consistently higher than those historically reported, the reasons for this enhancement are not yet fully understood. This unexpected outcome results in a reevaluation of the assumed limitations of Na-Sb as a photoemitter material and raises new questions about its intrinsic photoemissive properties.

One hypothesis is that the use of MBE, particularly with the implementation of in situ RHEED monitoring, enables the growth of films with superior crystallinity and fewer defects compared to earlier deposition techniques such as thermal evaporation. This structural improvement could

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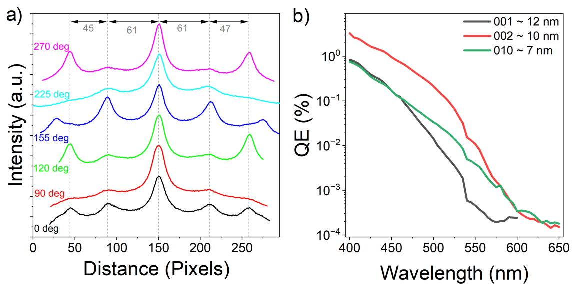

Figure 4: a) Intensity profile from RHEED Images at different angle positions for sample 010, b) Spectral response for samples 001, 002, and 010.

lead to reduced recombination at the grain boundaries and better transport of the carrier to the surface, which improves QE.

The angle-dependent RHEED streaks observed in the two-step growth of sample 010 indicate epitaxial ordering, which may be crucial to the unexpectedly high QE. This suggests that even small improvements in surface order and crystallinity can have a large impact on photocathode performance. Moreover, the variation in QE across samples with differing photoemission thresholds and morphologies implies that the observed photoemission is highly sensitive to minor growth parameter fluctuations, which may lead to different stoichiometries of the samples. This assumption needs different characterization techniques and will be studied further.

Interestingly, no strong correlation was found between QE and the residual presence of other alkali metals before deposition, which suggests that contamination or accidental compound formation is not a primary cause. Although other types of contamination due to having sources at high temperatures cannot be ruled out. This further supports the notion that pure Na-Sb, when grown under optimized MBE conditions, may possess photoemissive properties that have been previously underestimated.

CONCLUSION

This work challenges the conventional assumption that Na-Sb is not a good photoemitter in the visible spectrum. Instead, we demonstrate that Na-Sb, when epitaxially grown via MBE, exhibits QE values exceeding 1% at 400 nm—significantly higher than previously reported.

These findings open up several new avenues for photocathode research. First, Na-Sb itself may be viable as a high-performance photocathode material. Second, the high QE observed underlines the importance of growth technique and film quality, suggesting that further optimization of epitaxial methods could unlock enhanced performance in other alkali antimonides. Finally, these results emphasize the value of in situ diagnostics like RHEED in real-time film assessment and process tuning.

Future work will explore the fundamental mechanisms driving the enhanced QE, including band structure analysis, defect density characterization, and detailed surface science studies. Such understanding could ultimately lead to the rational design of next-generation photocathodes with tailored properties for demanding applications in accelerator science and beyond.

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