Characterization of the Intrinsic Emittance Properties of Ordered Crystalline Photocathode Materials \( XY_2 \) and \( XY_2Z_3 \)

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**Scientific Opportunity, Approach, and Significance:**

The ultimate capabilities of many advanced instruments, including high-brightness photon sources (e.g. synchrotrons and free electron lasers), ultra-fast electron probes (e.g. electron microscopes), and particle accelerators depend crucially on the properties of the source from which the electrons are initially emitted. An ideal electron source will not only have high efficiency, but its photoelectrons should also be emitted in a highly collimated, laser-like beam, minimizing the spread in the direction perpendicular to beam propagation. Minimizing this transverse spread (the so-called “mean transverse energy”, or MTE) is especially crucial because it cannot be removed using any electrostatic or magnetic lenses, as can be shown by Liouville’s theorem. Therefore, the initial momentum spread of photoelectrons emitted from a cathode places limits on the characteristics of the accelerated beam further downstream, and thus on the ultimate performance of sophisticated, multi-million dollar instruments such as synchrotrons, electron microscopes, and particle accelerators.

To date, most photocathode materials are traditional materials, such as elemental metals and III-V semiconductors such as GaAs. More recently, there has been a push to find new photocathode materials with optimized properties (such as minimized MTE). One class of new possible photocathode materials are alkali antimonides, particularly \( XY_2 \) and \( XY_2Z_3 \), which demonstrate quantum efficiencies exceeding 20% [4], potentially low mean transverse energies [5] and resilience to the demanding environment of photoinjectors [1]. However, only polycrystalline samples [6], often with high surface roughness [7], have been synthesized to date. This non-uniformity and roughness of the cathode surface causes photoelectrons to be emitted in randomized directions, obscuring the intrinsic (and best-possible) photoemission characteristics of these materials. Indeed, our recent many-body calculations of the photoemission process suggest that, at threshold, the MTE of photoelectrons from single-crystal (001) \( XY_2 \) is vanishingly small, and thus ideal for a photocathode. Therefore, the synthesis of large-area, single crystalline thin films of alkali antimonides are essential to test this prediction and characterize the ultimate performance characteristics of this potential new class of photocathodes.

We propose to utilize the PARADIM thin film facility to grow atomically flat, single crystal alkali antimonides, namely \( XY_2 \) and \( XY_2Z_3 \), epitaxially on lattice matched substrates, such as \( MgF_2 \). We aim to both clarify the growth process of this material using the rapid feedback provided by in situ reflection high-energy electron diffraction (RHEED) and to characterize the electronic structure using ARPES. Structural and electronic measurements of this type are possible only in the unique integrated growth/characterization system offered by the PARADIM facility, as the extreme air sensitivity of alkali antimonides prohibits ex situ studies. In addition to the tools available at PARADIM, we will utilize a recently constructed UHV transfer system or “suitcase” to shuttle our MBE grown films to several other diagnostic tools, including an STM for characterizing the surface morphology and roughness of these films, as well as a chamber for measuring the MTE under “real-world” settings, in a realistic high field photoinjector setting.

**Expertise in scientific domain of proposed research (Expertise in equipment and software operation will be supplied by PARADIM staff as necessary); Also, results of prior PARADIM work (if any):**
James Smith and Jane Brown have extensive MBE experience using Veeco GEN10 systems (the same as in PARADIM) synthesizing materials such as SrCuO$_3$ and BaSnO$_3$. Jane Brown has also previously successfully synthesized polycrystalline XY$_2$ and XY$_2$Z$_3$ on various non-crystalline substrates in the Cornell photocathode lab, which have had their quantum efficiency and emittance characterized in a realistic photoinjector environment and the surface morphology has been studied by in situ STM. Additionally, James Smith has experience in ultraviolet angle resolved photoemission spectroscopy and Jane Brown in low energy photoemission measurements.

**Appropriateness for PARADIM Facilities:**

The PARADIM thin facility is uniquely suited to the task of synthesizing crystalline XY$_2$ and XY$_2$Z$_3$, as it is able to utilize alkali metal sources, unlike nearly all other sophisticated molecular beam epitaxy systems (this is due to the high vapor pressure of alkali metals, which can typically contaminate the chamber). Furthermore, its in situ structural diagnostics, namely RHEED, are also crucial, as photocathode materials are typically grown without the aid of structural characterization tools, and thus without any real knowledge of the structural or electronic properties of the materials synthesized. Furthermore, despite the importance of the band structure in determining the properties of photocathodes such as the MTE, the band structure of XY$_2$ and other alkali antimonides remain experimentally unknown; as this material is extremely air sensitive an in situ ARPES system, such as the one on PARADIM, provides the only viable way to characterize the electronic structure.

We request to use the PARADIM MBE system for a total of 18 days distributed over two or three blocks. The first block would be used to optimize growth conditions on different substrates and establish structural, epitaxial, and phase purity of our samples. The other (or other two) blocks would be used to characterize our films using ARPES and LEED when that capability is available, as well as transfer them (in vacuum) to other Cornell facilities for further characterization. Using our UHV transfer system to shuttle samples we aim to study the surface morphology using the Lopez group STM and the low energy, high field photoemission properties using diagnostics at the Cornell photocathode lab.

**Alignment of the Science with PARADIM’s Technical Focus** (Materials for the next generation of electronics):

Alkali antimonides, and ordered crystalline photocathodes more generally, will play a key role in the next generation of bright electron sources for synchrotrons, accelerators, microscopes, and electron diffraction systems. In contrast to disordered cathodes, single crystal films offer a path to reduce beam emittances beyond the limitations imposed by surface scattering & thermalization and utilize directly the electronic structure of the material to control the properties of the electron beam. In this way we hope to create and characterize materials that can be used to improve the efficiency and efficacy of a wide array of tools and facilities which make use of accelerated electron beams.

**Methods and Metrics for Assessing Material Quality and Implementation of Materials-by-Design methodology:**

We are currently collaborating with theory collaborators to use computational methods to both select substrate materials desirable for epitaxial growth and to predict the photoemission properties (MTEs) of the resulting films. High throughput screening of materials databases, in conjunction with density functional theory calculations have been used to identify substrates providing different strain states for alkali antimonide films and then predict how these different strain states influence the electronic structure of the photocathodes, and ultimately the properties of the electron beams they will produce.
References: