

AN OVERVIEW OF SPIN-POLARIZED PHOTOCATHODE RESEARCH AT CORNELL UNIVERSITY

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Abstract

The development of a robust spin-polarized electron source capable of sustaining mA-scale average beam currents for an extended period of time in a photoinjector is critical for many future accelerator facilities such as the International Linear Collider (ILC). In this proceeding, we overview the several efforts being carried out at Cornell towards this end, including: high current (> 1 mA) gun tests of robust activation recipes of GaAs at the HERACLES beamline, the development and demonstration of GaN as a robust spin polarized source and Density Functional Theory (DFT) ab initio studies of alkali-antimonide photocathodes as potential spin polarized electron sources.

INTRODUCTION

In both nuclear and high energy physics communities, there is a strong desire for a spin-polarized electron source that can operate robustly at a high average current (> 1 mA) in a photoinjector to enable experiments requiring high luminosity spin-polarized electron beams[1, 2] and spin-polarized positron production[3]. To date, state-of-the-art spin polarized electron production is performed with a Gallium Arsenide (GaAs) based photocathode.

At the material bandgap of GaAs, a degeneracy between the Light-Hole (LH) and Heavy-Hole (HH) bandstates is partially lifted when operating the photocathode with circularly polarized light with near bandgap photon energies[4, 5]. For efficient operation of the photocathode with bandgap energy photons, the GaAs surface is brought to a condition called Negative Electron Affinity (NEA) where the vacuum surface potential is lowered to below the conduction band minimum. In this state, electrons that have been excited into the conduction band in the bulk can escape and contribute to the photoemission process if they reach the material surface. While in NEA, GaAs can have a Quantum Efficiency (QE) of a few percent with 780 nm light. The QE can be further increased through material engineering such as the use of a Distributed Bragg Reflector (DBR) where QE's as high as 15% have been reported[6].

The NEA state is accomplished by depositing a monolayer of an electropositive metal such as Cesium and an oxidant, typically oxygen or NF_3 and is extremely chemically reactive. This necessitates an NEA GaAs photocathode be both prepared and operated in an Ultra High Vacuum (UHV) environment. Even in UHV, degradation in the QE due to

chemical poisoning is readily observed. During operation inside a photoinjector, ion back-bombardment and laser induced thermal degradation further reduces the operational lifetime of GaAs. To increase the robustness of GaAs, a variety of alternative NEA activation recipes have been studied at Cornell University and elsewhere[7–10].

Given the variety of expertise and resources available, Cornell is uniquely positioned to contribute to the problem of producing a more robust spin-polarized electron source. In this contribution we give an overview of spin-polarized electron source research performed at Cornell, focusing on three unique research thrusts: (i) the High Electron Average Current for Lifetime ExperimentS (HERACLES) beamline, (ii) the development of GaN as a spin-polarized electron source and (iii) the development of a Density Functional Theory (DFT)-based simulation suite for identifying alternative spin-polarized electron sources.

RESEARCH OVERVIEW AT CORNELL

HERACLES

The HERACLES beamline[11] is a dedicated facility for performing charge lifetime experiments of photocathodes at high average current at Cornell. It has been commissioned to operate with up to 10 mA average beam current. The DC electron gun is typically operated with a beam energy of 200 keV. It was originally designed and built during Cornell's Electron Recovery Linac (ERL) program and was used to set the record high average current from a photoinjector [12] before being repurposed for HERACLES. The beamline is shown in Fig. 1, for more details see [11].

One of the major research thrusts in HERACLES is testing the robustness of alternative NEA activation techniques for GaAs, such as Cs-Sb-O and Cs-Te-O based recipes. Although numerous studies report these recipes produce improved robustness when chemical poisoning[8, 9, 13] is the dominant degradation mechanism, no study has yet shown their efficacy when operating at high average current. In HERACLES we performed the first lifetime studies of Cs-Sb-O activated GaAs but found no improvement in the lifetime over Cs-O activated GaAs[14]. However, this study was limited in two ways: (i) the Cs-Sb-O activated GaAs was transported via a vacuum suitcase to HERACLES and lifetime measurements were performed more than 24 hours after the initial activation when some degradation in the photocathode QE was already observed. Meanwhile the Cs-O activations were performed in HERACLES's auxiliary vacuum system and lifetime measurements were performed

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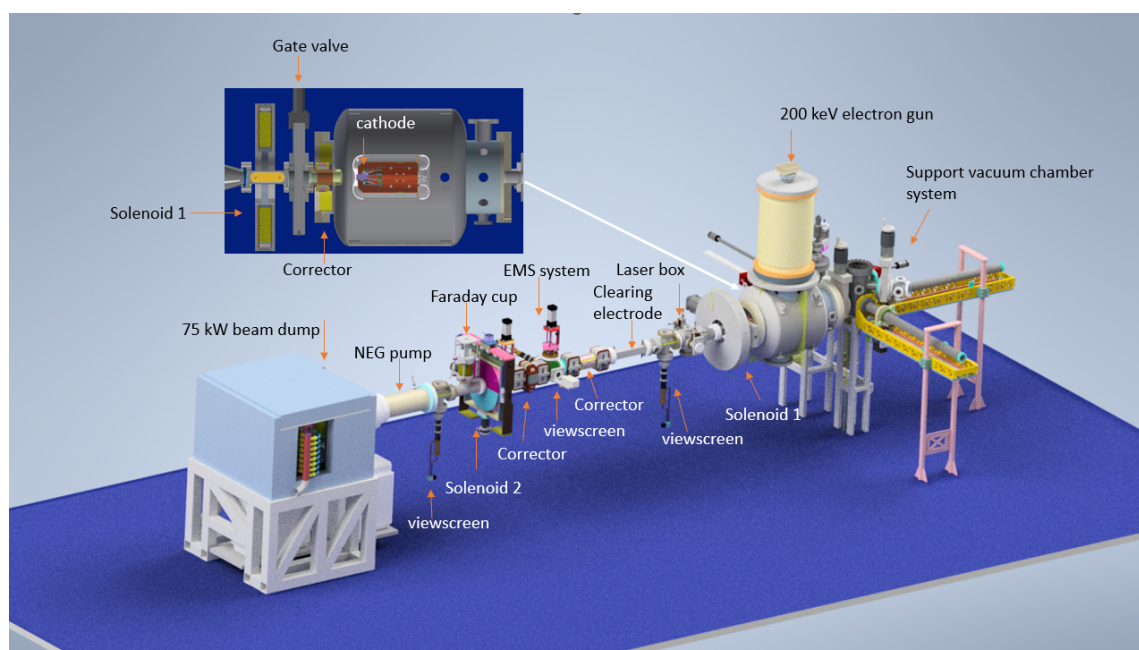


Figure 1: The HERACLES beamline. Lead shielding and SF_6 tank are not shown. The floor corresponds to the room dimensions. The inset shows a cross-section of the gun viewed from above.

immediately after growth (ii) only two samples each of Cs-Sb-O and Cs-O were compared which did not allow for optimization of the Cs-Sb-O growth recipe.

In an ongoing project, we have upgraded the growth system in HERACLES to allow for Cs-Sb-O and Cs-Te-O activations to enable lifetime measurements of these recipes without needing to transport the photocathode with a vacuum suitcase. For more details see MOPR80 of this proceeding.

Development of GaN as a Spin-Polarized Electron Source

Like GaAs, GaN can also be operated as a NEA photocathode. GaN can be grown in two phases, hexagonal or cubic and has several interesting characteristics that point toward it being a possible robust spin polarized electron source. For example, QE's higher than 50% have been reported[15] and Cs free NEA has been achieved with N-Polar GaN[16]. The thermal conductivity of GaN is larger by a factor of 2 over GaAs[17, 18] which may help mitigate thermal degradation in the photocathode QE due to laser induced heating during high average current operation. Interestingly, when comparing bandgap energy dark lifetimes between hexagonal GaN and GaAs, it was found that hexagonal GaN is 20 times more robust[19]. Measurements performed at Cornell indicated the dark lifetime for cubic GaN is 12 times longer than GaAs.

Recently we have demonstrated for the first time ever that NEA GaN-based photocathodes can generate spin polarized photoemission from both cubic and hexagonal phases. Both samples were grown epitaxially at Cornell. For cubic GaN 3C-SiC sample purchased from NOVASiC was used for the substrate while for the hexagonal sample single crystal GaN

substrate was purchased from Ammono. To set the atomic structure, a 100 nm unintentionally doped GaN layer was grown for each sample followed by a 1 μm Mg-doped GaN layer to serve as the photoemitter material. The doping concentration on both samples was $3 \times 10^{19} \text{ cm}^{-3}$ based on work which reported an optimum QE at that doping concentration[20]. A Mott scattering polarimeter with a tungsten target biased at 20 keV was used to measure the photoemission electron spin polarization (ESP). A peak ESP value of 29% was measured for cubic GaN. This value is comparable to the 35% that is typically reported for bulk GaAs. The peak ESP value of hexagonal GaN was measured to be 17%. Figure 2 shows the spectral ESP response obtained using a home built tunable UV source with typical bandwidth of 1.4 nm. For more details and characterization of spin-polarized photoemission from GaN see [21].

With spin-polarized photoemission from GaN now established, there are several opportunities for improvement. One avenue is better crystal quality which may increase both QE and lifetime, particularly for cubic GaN which presently suffers from high Anti-Phase Boundaries (APB) and a high surface roughness. For example, the use of a AlN buffer layer between the 3C-SiC substrate and cubic GaN has been used to improve surface roughness[22].

Similar to the development of GaAs as a photocathode, the introduction of a strained lattice can be used increase the ESP of cubic GaN while DBR's can be used to enhance the QE in both hexagonal and cubic GaN.

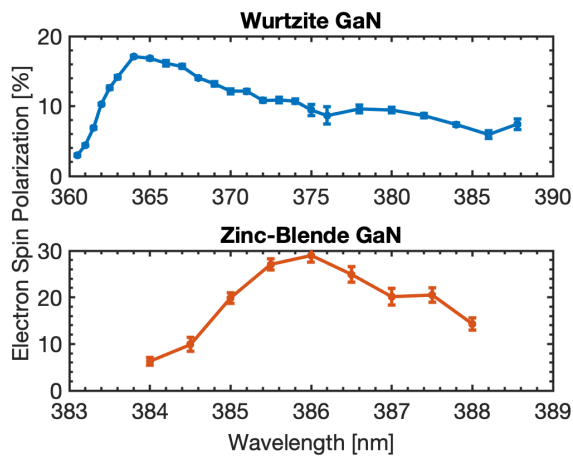


Figure 2: Measured electron spin polarization of hexagonal (top) and cubic (bottom) from GaN based photocathodes.

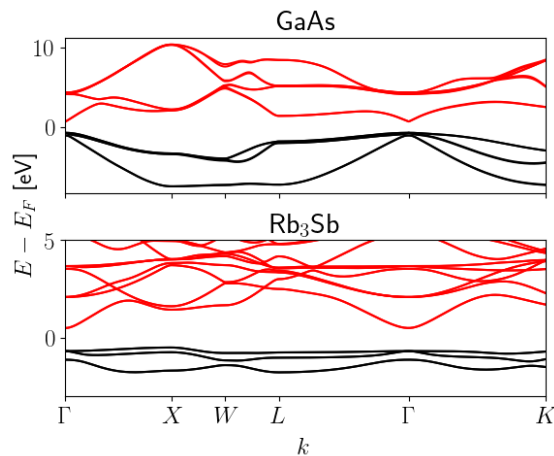


Figure 3: DFT calculated band structures for bulk GaAs (top) and Rb_3Sb (bottom). Black bands indicate filled bands and red bands indicate unfilled bands.

Ab Initio computation of alkali-antimonide photocathodes as spin-polarized electron sources

While progress in increased robustness from III-V NEA photocathodes like GaAs and GaN remains fruitful, it is extremely interesting that an indirect observation of spin-polarized photoemission from the alkali-antimonide Na_2KSb activated to NEA with Cs_3Sb was recently reported [23]. As a photoemitter, the alkali-antimonide family is far more robust than III-V NEA photocathodes and are often employed in demanding high current unpolarized electron beam applications such as Electron Recovery Linacs (ERL)[24] and hadron beam cooling[25]. Thus, the discovery of spin-polarized photoemission from an alkali-antimonide photocathode opens several avenues of research.

To better understand the possible use of alkali-antimonide photocathodes for high current spin polarized beam applications, we have developed a computational method based on Kohn-Sham Density Functional Theory to predict the

initial degree of spin-polarization excited in a photocathode material. While there exists limited studies on microscopic *ab initio* approaches to calculating the spin polarization [26], our approach to performing accurate high throughput calculations employs Wannier Functions to obtain very fine Brillouin zone sampling which is then used to perform a Monte Carlo integration to obtain a final ESP curve. One current limitation of the computation is we do not account effects that occur between the excitation of the an electron to a conducting state and its subsequent emission. Thus, any excited electron is assumed to photo-emit and depolarization effects are not included.

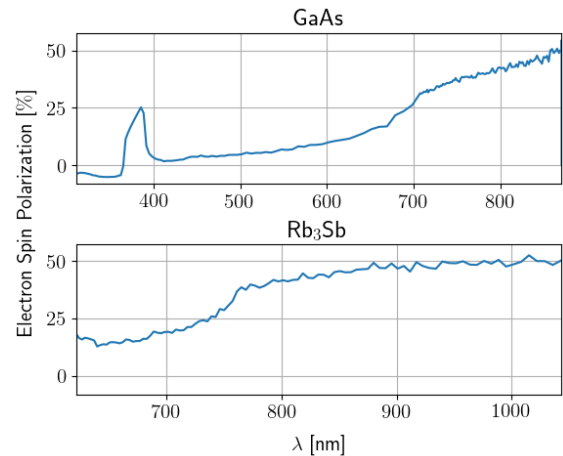


Figure 4: DFT calculated initial electron spin polarization for bulk GaAs (top) and Rb_3Sb (bottom).

As a proof-of-concept, the ESP curve for GaAs was computed as shown in Fig. 4 with the corresponding band structure in Fig.3. As expected, the ESP has a peak polarization of 50% when excited with near band gap photon energies and decreases with increasing photon energy. Note that a second peak near 400 nm as well as a spin inversion was also observed in [5].

We next consider the alkali-antimonide Rb_3Sb in its face-centered cubic phase; the obtained band-structure and ESP curve are shown in Figs. 3 and 4, respectively. Both Rb_3Sb and GaAs are FCC structures and consequently have similar band-structures as can be seen in Fig. 3, particularly around the Γ point, implying similar direct bandgap transitions with near bandgap photon energies. Is is therefore reasonable that we also compute a peak ESP value of 50% with nearband gap photon energies from this material. Thus, Rb_3Sb looks promising as a spin-polarized photo-emitter. Experimental work to measure the ESP curve of Rb_3Sb is currently underway at Cornell.

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