

Final Technical Report

Proposal Title:

“Investigation of fundamental limits to beam brightness available from photoinjectors”

Period of Report:

4/15/2010 to 4/15/2015

Principle Investigator:

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Institution:

Cornell University

DOE Award Number:

DE-SC0003965

Award Amount:

\$750,000

Work-scope overview:

The goal of this project was investigation of fundamental limits to beam brightness available from photoinjectors. This basic research in accelerator physics spanned over 5 years aiming to extend the fundamental understanding of high average current low emittance sources of relativistic electrons based on photoemission guns, a necessary prerequisite for a new generation of coherent X-ray synchrotron radiation facilities based on continuous duty superconducting linacs. The program focused on two areas critical to making advances in the electron source performance: 1) the physics of photocathodes for the production of low emittance electrons and 2) control of space charge forces in the immediate vicinity to the cathode via 3D laser pulse shaping.

Major goals of the project along with the accomplishments:

1) Physics of photocathodes for production of low emittance electrons

The goals for advancing the photocathode research included activities in two research areas: developing a theoretical model of photoemission physics with its subsequent experimental verification; and demonstration of superior photocathode performance in the actual accelerator environment of a high-voltage DC photoemission gun. Below, we provide highlights from all the major accomplishments.

Monte-Carlo modeling for photoemission.

Photoemission-based electron sources can produce extremely bright beams with subpicosecond time resolution, which find uses in many applications including modern light sources based on free-electron lasers and energy recovery linacs. The ultimate brightness of an electron beam obtained from a photoinjector is limited by the mean transverse energy (MTE) of the electrons emitted from the photocathode (or similarly, intrinsic emittance of the photocathode, where the two quantities are related by an expression:

$$\epsilon_{n,x} = \sigma_{l,x} \sqrt{\frac{\text{MTE}}{m_e c^2}},$$

with $\epsilon_{n,x}$ being normalized transverse rms emittance, $\sigma_{l,x}$ being rms laser spot size at the photocathode, and $m_e c^2$ being the electron rest mass energy). An ideal photocathode for use in a photoinjector must have a high quantum efficiency (QE), low MTE, a quick response time, and a good operational lifetime. High quantum efficiency photocathodes are limited to semiconducting materials, such as cesiated direct band gap III-V crystals and alkali-antimonides or -tellurides. Despite its extreme sensitivity to vacuum conditions, GaAs activated to negative electron affinity (NEA) with Cs/O or Cs/NF₃ remains an important photocathode due to its high QE in the visible/near infrared range

and low MTE. Though GaAs photocathodes were studied for several decades, much of the physics of photoemission still remains unclear. Specifically, the energy distribution and response time, both of which are important for the operation of photoinjectors, predicted by a simple diffusion theory tend to be quite different from the observed values. Resolution of this problem calls for a more intricate modeling of electron transport during photoemission. In addition, a better theory and modeling of photoemission would provide a much-needed direction for further improving photocathodes.

To this end, we have developed a Monte-Carlo charge transport and photoemission code for negative electron affinity GaAs photocathodes. The simulation code has to meet a number of criteria, among which are: a) be able to predict QE, MTE, and response time from photocathodes without *ad hoc* fit parameters; b) to undergo experimental benchmarking to establish its validity; c) to be flexible so that new materials and layered structures can be simulated. The resulting code is based on the expanded 3-step model of photoemission (excitation, transport, emission), with the detailed account of various scattering processes in the bulk of the material (with impurities, phonons, and carriers) and quantum probabilities for the bulk-vacuum interface. Additionally, the code is interfaced to a Poisson-Schrodinger solver to obtain details of the electric field acting on the carriers due to band bending of hetero- and homo-junctions as well as the surface interface. Although the code was written primarily with III-V semiconductors in mind, its structure is flexible so that other materials can be modeled after some work. For III-V's, we use a 3-valley model as shown in Figure 1.

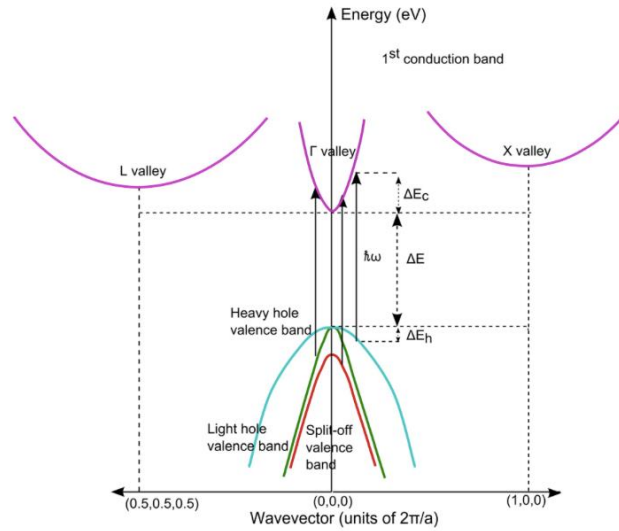


Figure 1. Electron excitation from top 3 valence bands to the lowest conduction band. Only direct (vertical) transitions are shown. Indirect, inter- valence band and multi-photon transitions have been ignored.

The code, which was developed in collaboration with Dr. Dimitrov (Tech-X), has undergone extensive benchmarking using both previously published and newly collected data. With only one “free parameter”, namely the magnitude of electron affinity, we were

able to obtain good agreement for cesiated bulk GaAs for QE and MTE vs. laser wavelength, see Figure 2. Details of this work have been published in [J. Appl. Phys. 113, 104904 \(2013\)](#). The good agreement between experiment and modeling later enabled us to perform the first proof-of-principle experiment with “engineered” photocathode for low emittance, where the results of the cathode simulations were used to predict improved emittance of a specially designed GaAs structure (see below a section on the engineered photocathode for more details).

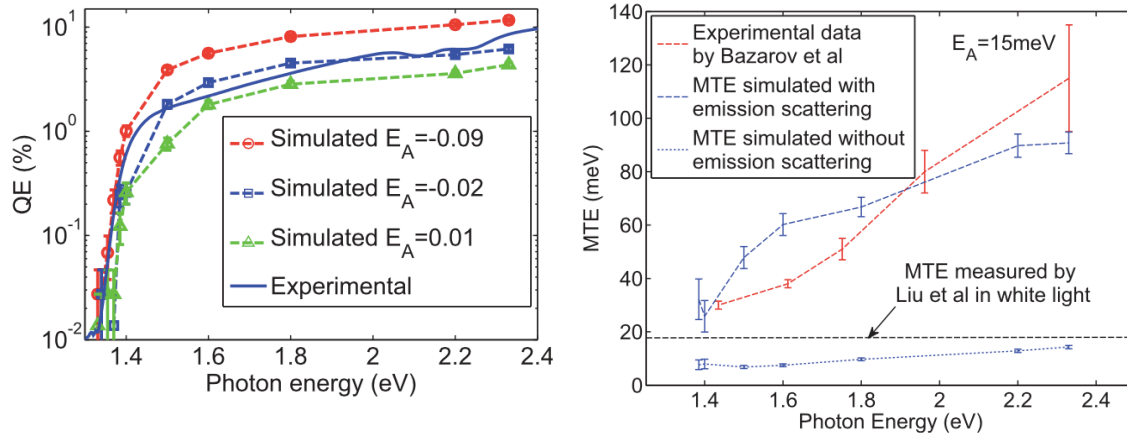


Figure 2. Comparison of measured and simulated spectral response (left) and mean transverse energy (right) from cesiated bulk GaAs. Electron affinity (E_A) values used for simulations are indicated in the figure.

Photocathode Research Laboratory at Cornell University.

Part of the reason for limited progress in understanding the physics of photoemission is a lack of self-consistent data on photocathode characteristics relevant to the accelerator performance. For example, intrinsic emittance is a function of not only photocathode material and its stoichiometry, but also the details of its photo-activation, surface preparation, etc. All this necessitates a measurement system, which can perform as many of the desired measurements *in-situ* as possible. A significant effort of this project was dedicated to deploying such a photocathode research system, which consists of a series of interconnected vacuum chambers dedicated to growth (or activation) and characterization of photocathodes without the need to break the vacuum. Figure 3 shows a diagram and a photograph of the system developed at Cornell University. Various techniques such as Scanning Auger Spectroscopy, Low Energy Electron Diffraction (LEED), Reflected High Energy Electron Diffraction (RHEED), and work function measurements are used to characterize the surface properties of the specimens. Spectral response, photoemission uniformity, and electron energy distributions are used to characterize quality of the samples and to relate it to the measured surface properties.

One of the major goals (and expenses) of this award was to design and build a measurement system capable of simultaneously obtaining electron energy spectra with 2 different components of momentum: transverse and longitudinal kinetic energies relative

to the normal of the photocathode. The diagnostic had to be compact and capable of measuring intrinsic photocathode emittances *in-situ* without the need of radiation producing high voltage for beam acceleration.

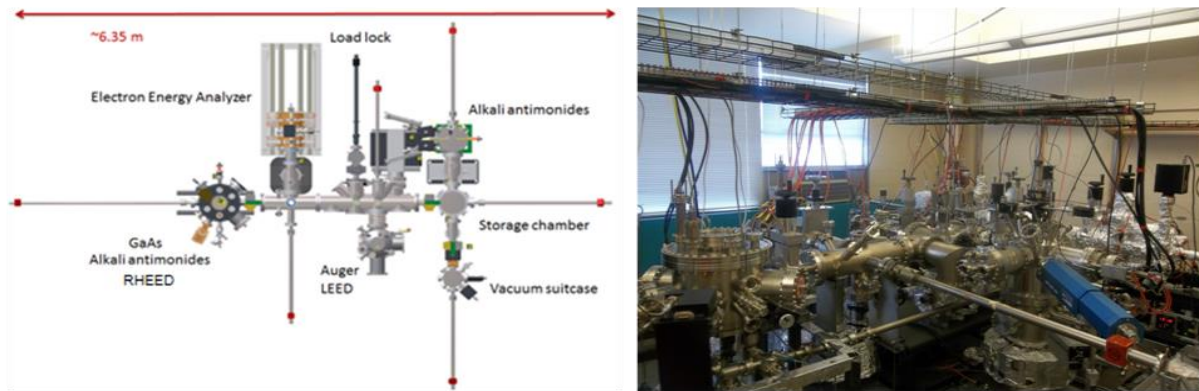


Figure 3. (Left) Schematic of the growth and analysis vacuum system in photocathode research lab at Cornell University. (Right) Photograph of the system

Following an idea demonstrated previously at Heidelberg University, a photocathode is submerged in a strong magnetic field. This magnetic field varies smoothly between the cathode and the detector, so that precessing electrons exchange their transverse and kinetic energies via an adiabatic invariant. A couple of “marking” electrons placed at different locations thus allow mapping of the energy distribution of photoelectrons with both transverse and longitudinal energies uniquely “tagged”. We have constructed such an Energy Analyzer, and have successfully improved its resolution by a factor of 3 compared to the original. Figure 4 shows a cross-section of the Analyzer and [Rev. Sci. Instrum. 86, 033301 \(2015\)](#) contains further details on the implementation. The data obtained using the Energy Analyzer were successfully crosschecked against other independent methods of measuring transverse emittance ensuring our confidence in this method.

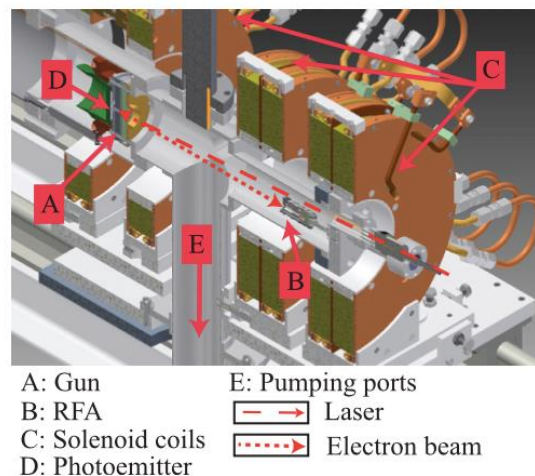


Figure 4. Cross-section of the model of the energy analyzer built at Cornell University.

Photocathode “engineering” proof-of-principle experiment.

One of the longer-term goals for the photocathode community as a whole is to be able to synthesize novel photocathodes to meet ever-increasing demands on emittance and other parameters relevant to accelerator applications. Using the newly developed simulation capability (see the section on Monte-Carlo simulations above) and ability to perform semiconductor growth to specs on campus, we have performed a proof-of-principle experiment demonstrating improved emittance from a layered GaAs-based photocathode. In a nutshell, the band-bending region was engineered to reduce access kinetic energy of photoexcited electrons in the bulk, so that the final MTE after the emission into vacuum was $\times 2$ lower as compared to the bulk GaAs. Figure 5 gives a summary of the results. Refer to [Phys. Rev. Lett. 112, 097601](#) for details.

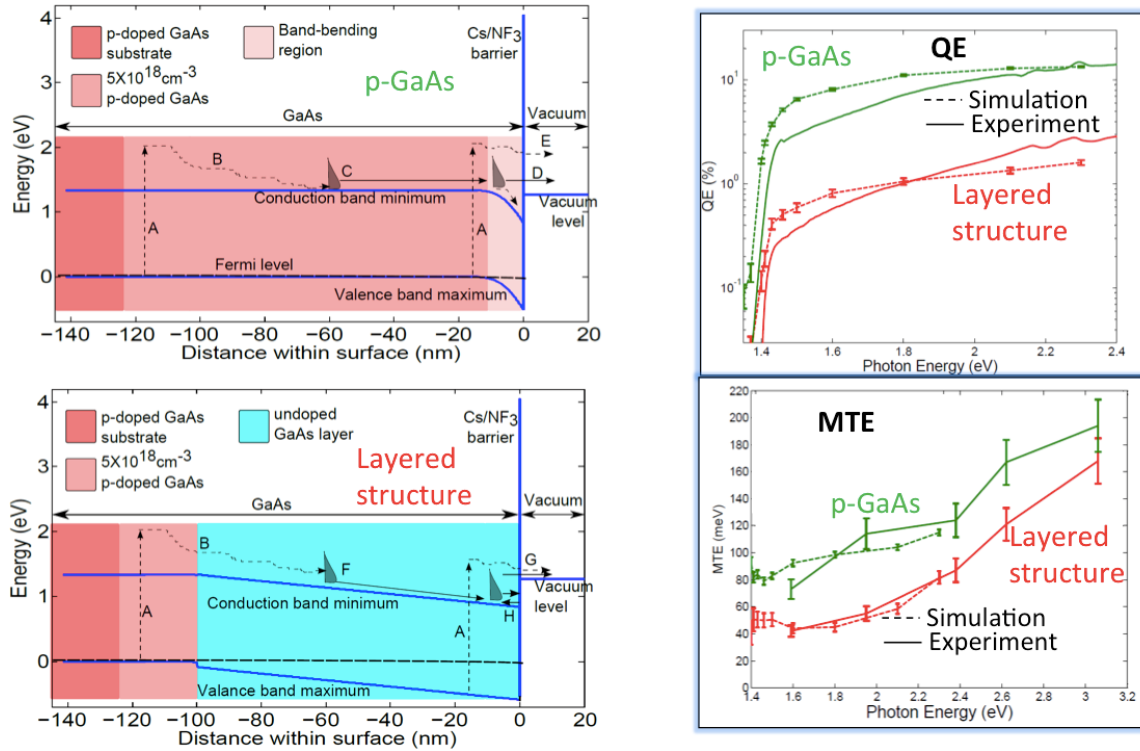


Figure 5. Proof-of-principle experiment showing reduced emittance from a layered GaAs photocathode. The left column shows energy diagram of a control bulk GaAs sample (top) and a layered structure specifically grown for a lower intrinsic emittance. The right column shows the simulated and measured values for QE and MTE for both samples.

New photocathode measurements (emittance and lifetime).

While GaAs and other III-V's provide a flexible platform for systematic investigation of the photoemission physics, these materials have a major practical drawback. Their superior photoemissive properties arise largely due to a thin activation dipole layer consisting of Cs and typically some oxidant. The activation layer is only a monolayer or

so thick, and thus, is extremely delicate. As a result, this puts a rather stringent requirement on the base vacuum and limits operational lifetime of this photocathode family for high current applications.

Much attention in the framework of this award has been given to promising alkali-antimonide photocathodes, such as Cs-K-Sb, Na-K-Sb, and Cs-Sb. These materials are “bulk-active” rather than “surface-active”, meaning that much thicker films of the material (tens of nanometers as opposed to several angstroms) are responsible for photoemission. These materials have been studied in a systematic manner for their intrinsic emittance resulting in the much-needed data for the accelerator community.

In addition to the emittance characterization, these materials have allowed Cornell Photoinjector Group to push average current from a photoinjector to new records, both in terms of average current and photocathode lifetime. In particular, Na-K-Sb photocathode has allowed sustained operation at 65 mA of average current for over 8 hours (Figure 6) with measured $1/e$ lifetime of 2.6 days (Figure 7).

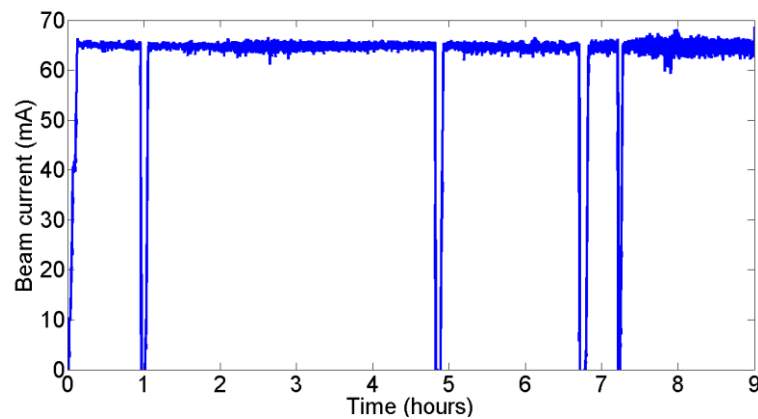


Figure 6. High current operation using Na-K-Sb photocathode from Cornell High Current Photoinjector.

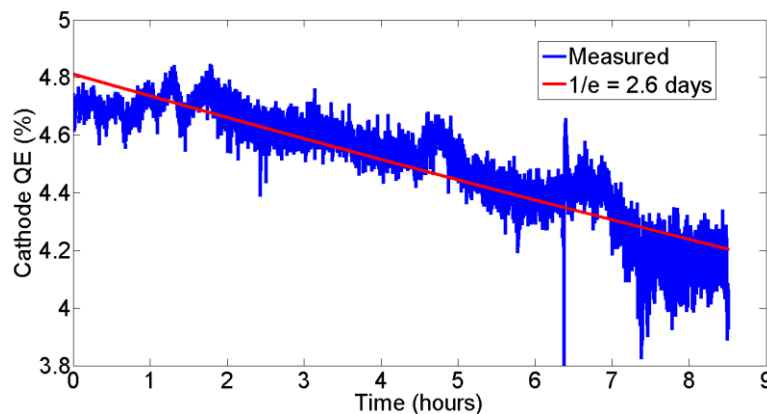


Figure 7. QE degradation during the 65 mA run.

Details of this work are published in [Appl. Phys. Lett. 103, 103504 \(2013\)](#).

2) Physics of space charge near the photocathode and 3D laser shaping

The main goals here included understanding of beam brightness limits that arise due to both controlled and uncontrolled space charge interactions inside electron bunches shortly after they are generated from the photocathode.

Disorder-induced heating and its limit on lowest achievable emittance.

Space charge forces inside electron bunches can be categorized as controllable and uncontrollable depending on whether they arise from causes that can be directly affected or not. For example, controlling initial 3D laser distribution impinging on the photocathode can largely compensate both linear and non-linear space charge forces inside the electron bunch. On the other hand, any electron interactions within a Debye sphere, can potentially lead to “heating up” of the electron distribution and set a limit to beam brightness which cannot be compensated by affecting electron bunch as a whole.

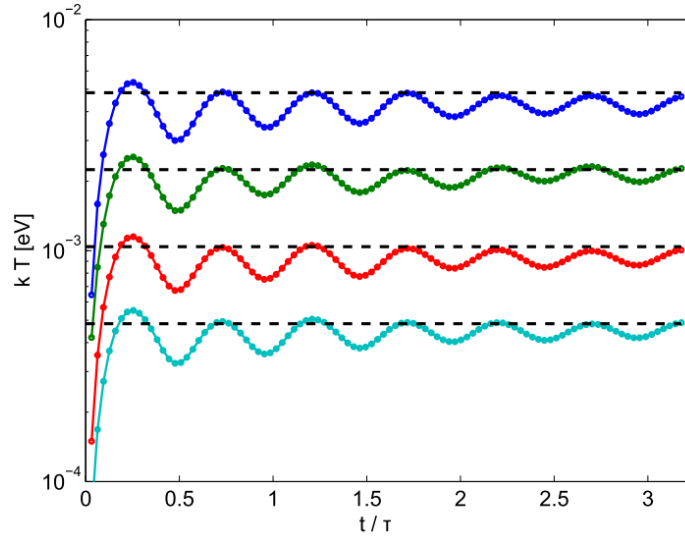


Figure 8. Electron temperature versus number of plasma periods after emission from treecode simulation using 10^5 contained electrons, and $n_0 = 10^{20}$ (top curve), 10^{19} , 10^{18} , 10^{17} m^{-3} (bottom curve), with zero initial temperature. The predicted equilibrium temperature is shown in dotted lines.

As a part of this award, we set out to address the new limit to beam brightness that arises from unshielded close electron encounters within intense electron bunches shortly after they are generated from the cathode. Together with Dr. Padmore (LBNL), we performed treecode space charge simulations to understand the scaling of emittance limit from such disorder-induced self-heating of the electrons. We were able to formulate a new fundamental limit ([New J. Phys. 15 103024](#)), which computes the lower limit to the effective temperature or energy spread from strongly coupled Coulomb collisions in the immediate vicinity of the photocathode depending on the electron bunch density. This limit applies to the emerging high brightness photoemission electron sources used for many scientific applications, and motivates the photocathode research by setting the

lowest intrinsic cathode emittance which still can translate to a higher brightness beam after initial acceleration. Figure 8 illustrates how initially perfectly cold beam develops a temperature increase (and a corresponding emittance growth) due to short range Coulomb interactions.

Adaptive laser shaping.

The initial laser distribution plays an important role in determining the lowest emittance achievable from a photoinjector. An ideal laser shape is a function of many parameters including bunch charge, gun voltage and accelerating gradient, compression scheme being used, etc. There exists a substantial difficulty in “dialing” a desired transverse laser shape on the photocathode without being overly demanding on the laser pointing stability and loosing exorbitant amounts of laser power in the process. Furthermore, it is known from the previous operational experience that QE of a photocathode does not stay constant over its lifetime, and therefore, an adaptive method to shape laser pulses is ultimately called for.

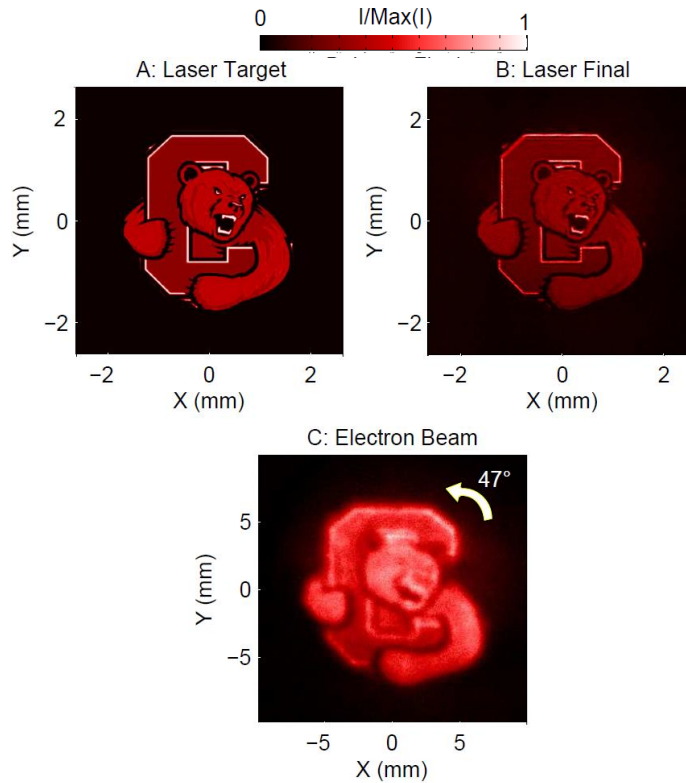


Figure 9. A: Desired distribution for the photoemission drive laser transverse profile. B: Actual laser distribution obtained with SLM shaping. C: 300kV electron beam profile using B and a DC gun (without space charge). The cathode plane is imaged via a solenoid lens on to a viewscreen.

We were able to implement a high accuracy laser shaping method using a spatial light modulator (SLM) as a polarization-encoder. An active feedback algorithm, imaging only

the electron beam, has been used to change the laser profile to accommodate for photocathode degradation or electron aberrations. High shaped fidelity electron beam profiles were successfully demonstrated using a high voltage photoemission gun, see Figure 9. This method, along with a previously developed temporal shaping technique via pulse stacking with birefringent crystals, enables flexible and efficient 3D laser pulse shaping for photoinjectors. Refer to [Phys. Rev. ST Accel. Beams 18, 023401](#) for details.

Publications resulting from this award:

- S. Karkare and I.V. Bazarov, "Effect of nano-scale surface roughness on transverse energy spread from GaAs photocathodes", Applied Physics Letters 98 (2011) 094104
- I.V. Bazarov, L. Cultrera, A. Bartnik, B. Dunham, S. Karkare, Y. Li, X. Xianghong, J. Maxson, W. Roussel, "Thermal emittance measurements of a cesium potassium antimonide photocathode", Applied Physics Letters 98 (2011) 224101
- S.S. Karkare, I.V. Bazarov, L. Cultrera, A. Iyer, X. Liu, W.J. Schaff, "Effect of Surface Roughness on the Emittance from GaAs Photocathode", Proceedings of the 2011 Particle Accelerator Conference, New York, NY, (2011) 2480-2
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- L. Cultrera, I. Bazarov, A. Bartnik, B. Dunham, S. Karkare, R. Merluzzi, M. Nichols, "Thermal emittance and response time of a cesium antimonide photocathode", Applied Physics Letters 99 (2011) 152110
- L. Cultrera, J. Maxson, I. Bazarov, S. Belomestnykh, J. Dobbins, B. Dunham, S. Karkare, R. Kaplan, V. Kostroun, Y. Li, X. Liu, F. Loehl, K. Smolenski, Z. Zhao, D. Rice, P. Quigley, M. Tigner, V. Veshcherevich, K. Finkelstein, D. Dale, B. Pichler, "Photocathode Behavior During High Current Running in the Cornell ERL Photoinjector", Physical Review Special Topics - Accelerators and Beams 14 (2011) 120101
- J.M. Maxson, S. Karkare, I.V. Bazarov, S. Belomestnykh, L. Cultrera, D. Dale, J. Dobbins, B. Dunham, K. Finkelstein, R. Kaplan, V.O. Kostroun, Y. Li, X. Liu, F. Loehl, B.K. Pichler, P. Quigley, D. Rice, K.W. Smolenski, M. Tigner, V. Veshcherevich, Z. Zhao, "Characterization of Photocathode Damage during High Current Operation of the Cornell ERL Photoinjector", Proceedings of the 2012 International Particle Accelerator Conference, New Orleans, Louisiana (2012) 2717-9

- L. Cultrera, I.V. Bazarov, J.V. Conway, B. Dunham, Y. Hwang, Y. Li, X. Liu, R. Merluzzi, T.P. Moore, K.W. Smolenski, S. Karkare, J.M. Maxson, W. Schaff, "Photocathode R&D at Cornell University", Proceedings of the 2012 International Particle Accelerator Conference, New Orleans, Louisiana (2012) 2137-9
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- J. Maxson, I. Bazarov, C. Coleman-Smith, W. Wan, H. Padmore, "Fundamental Photoemission Brightness Limit from Disorder Induced Heating", New Journal of Physics 15 (2013) 103024
- Colwyn Gulliford, Adam Bartnik, Ivan Bazarov, Luca Cultrera, John Dobbins, Bruce Dunham, Francisco Gonzalez, Siddharth Karkare, Hyeri Lee, Heng Li, Yulin Li, Xianghong Liu, Jared Maxson, Christian Nguyen, Karl Smolenski, Zhi Zhao, "Demonstration of Low Emittance in the Cornell Energy Recovery Linac Injector Prototype", Physics Review Special Topics - Accelerators and Beams 16 (2013) 073401
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- S. Karkare, L. Boulet, L. Cultrera, B. Dunham, X. Liu, W. Schaff, I. Bazarov, "Ultrabright and ultrafast III-V semiconductor photocathodes", Physical Review Letters 112 (2014) 097601
- S.S. Karkare, I.V. Bazarov, L. Cultrera, W.J. Schaff, X.G. Jin, Y. Takeda, "Photoemission from III-V semiconductor cathodes", Proceedings of the 5th International Particle Accelerator Conference, Dresden, Germany (2014) 736-8

- L. Cultrera, M. Brown, S. Karkare, W. Schaff, I. Bazarov, B. Dunham, "Alkali azide based growth of high quantum efficiency photocathodes", Journal of Vacuum Science and Technology 32 (2014) 031211
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- J. Maxson, H. Lee, A.C. Bartnik, J. Kiefer, I.V. Bazarov, "Adaptive electron beam shaping using a photoemission gun and spatial light modulator", Physical Review Special Topics - Accelerators and Beams 18 (2015) 023401
- S. Karkare, L. Boulet, A. Signh, H. Richard, I. Bazarov, "Ab-initio studies of Cs on GaAs (100) and (110) surfaces", Physical Review B 91 (2015) 035408
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- S. Karkare, L. Cultrera, Y-W. Hwang, R. Merluzzi, I. Bazarov, "2-D energy analyzer for low energy electrons", Review of Scientific Instruments 86 (2015) 033301

Summary and Conclusion:

The project has explored limits to beam brightness in modern photoinjectors from a number of directions, which can be broadly categorized into 2 classes: 1) physics of high performance photocathodes and 2) space charge interactions inside electron bunches in the vicinity of the cathode. The performed research has shed a new light on the mechanisms of photoemission as they relate to fundamental beam properties. A new limit to beam brightness was identified, which arises from disorder-induced heating of the photoelectrons randomly generated at the cathode. In addition, a new adaptive method for 3D laser shaping was realized and successfully demonstrated in an actual photoemission gun. The research results have stimulated the area of photocathodes and have had an impact on bright beam production field as a whole. Finally, many of the advances made within this proposal have directly contributed to new record-setting low emittance and high current demonstrations at the Cornell Photoinjector, which now represents a new state-of-the-art in the area of high brightness photoelectron sources.