Center for BRIGHT BEAMS

A National Science Foundation Science & Technology Center

Senior Investigator Reports 2023

Gaining the fundamental understanding needed to transform the brightness of electron beams available to science, medicine, and industry.

CBB Institutions

Partner	Project Leaders	Area of Expertise
Arizona State	Karkare	Photocathode materials
BYU	Transtrum	Theoretical condensed matter physics
UCLA	Musumeci, Rosenzweig	Laser applications for particle beams, beam self-interaction
U Chicago	Kim, Sibener	Particle physics, surface chemistry
Cornell	Arias, Bazarov, Hines, Liepe, Maxson, Muller, Sethna, Shen	Electronic structure calculation, photoemission & transport, surface characterization, accelerating cavities, photoemission & transport, electron microscopy, condensed matter theory, ARPES
U Florida	Hennig	Materials design
U New Mexico	Biedron	Machine learning, FELs
Northern Illinois U	Piot, Chubenko	Beam dynamics, Physical processes in photocathodes.
FNAL	Posen	Accelerating cavities

Table 1. Senior Researchers and Areas of Expertise.

The CBB Team

Arizona State University

PIs: Siddharth Karkare (Physics)
Postdoc: Mansoure Moeini Rizi
Graduate Students: Alimohammad Kachwala, Chris Knill, Pallavi Saha, Gevork Gevorkyan

Brigham Young University

PI: Mark Transtrum (Physics & Astronomy)
Postdoc: Benjamin Francis
Graduate Student: Aiden Harbick

University of California at Los Angeles (UCLA)

PIs: Pietro Musumeci (Physics), James Rosenzweig (Physics)
Postdoc: Nathan Majernik
Graduate Students: Eric Cropp, David Garcia, Gerard Lawler, Joshua Mann
Undergraduate Students: V. Guo, A. Kulkarni

University of Chicago

PIs: Young-Kee Kim (Physics), Steve Sibener (Chemistry)
Postdocs: Rachel Farber, Chris Pierce
Graduate Students: Juan Pablo Gonzalez Aguilera, Caleb Thompson, Sarah Willson, Van Do, Michael Van Duinen
Undergraduate Students: Helena Lew-Kiedrowska

Cornell University (lead institution)

Director: J. Ritchie Patterson (Physics)
PIs: Tomás Arias (Physics), Ivan Bazarov (Physics), Melissa Hines (Chemistry and Chemical Biology), Georg Hoffstaetter (Physics), Matthias Liepe (Physics), Jared Maxson (Physics), David Muller (Applied Physics), Kyle Shen (Physics)
Postdocs: Zeming Sun, Elena Echeverria
Graduate Students: Kevin Nangoi, Nathan Sitaraman, Michelle Kelly, Samuel Levenson, Annabel Selino, Mariam Hasany, Amy Zhu, Azriel Finsterer, Lucy Lin, Gabriel Gaitan, Liana Shpani, Chad Pennington, Zhaslan Baraissov, Desheng Ma, Vivek Anil, Charles Zhang, Tyler Wu
Undergraduate Students: Michelle Kwok (CBB REU student), Brandon Li

University of Florida

PI: Richard Hennig (Materials Science and Engineering)
Graduate students: Sam Dong, Jason Gibson, Ajinkya Hire

University of New Mexico

PI: Sandra Biedron (Electrical and Computer Engineering)

Graduate students: Aasma Aslam

Northern Illinois University

PI: Philippe Piot (Physics), Oksana Chubenko (Physics)
Postdoc: Afnan Al Marzouk
Graduate Students: Austin (AJ) Dick, Daniel Franklin, Tariqul Hasan

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Papers

C. T. Parzyck, A. Galdi, J. K. Nangoi, W. J. I. DeBenedetti, J. Balajka, B. D. Faeth, H. Paik, C. Hu, T. A. Arias, M. A. Hines, D. G. Schlom, K. M. Shen, and J. M. Maxson, “Single-Crystal Alkali Antimonide Photocathodes: High Efficiency in the Ultrathin Limit,” *Phys. Rev. Lett.*, vol. 128, no. 11, p. 114801, Mar. 2022, doi: 10.1103/PhysRevLett.128.114801.

A. A. McMillan, C. J. Thompson, M. M. Kelley, J. D. Graham, T. A. Arias, and S. J. Sibener, “A combined helium atom scattering and density-functional theory study of the Nb(100) surface oxide reconstruction: Phonon band structures and vibrational dynamics,” *J. Chem. Phys.*, vol. 156, no. 12, p. 124702, Mar. 2022, doi: 10.1063/5.0085653.

S. Deyo, M. Kelley, N. Sitaraman, T. Oseroff, D. B. Liarte, T. Arias, M. Liepe, and J. P. Sethna, “Dissipation by surface states in superconducting RF cavities,” *arXiv:2201.07747 [cond-mat, physics:physics]*, Jan. 2022, doi: 10.48550/arXiv.2201.07747. [Online]. Available: http://arxiv.org/abs/2201.07747

P. Saha, O. Chubenko, J. Kevin Nangoi, T. Arias, E. Montgomery, S. Poddar, H. A. Padmore, and S. Karkare, “Theory of photoemission from cathodes with disordered surfaces,” *Journal of Applied Physics*, vol. 133, no. 5, p. 053102, Feb. 2023, doi: 10.1063/5.0135629.

Talks

"Ab initio theory of coherent electron-phonon-photon processes in photoemission from single-crystal materials," by Johannes Kevin Nangoi, Siddharth Karkare, Ravishankar Sundararaman, Howard A Padmore, and Tomás A Arias, 14-18 March 2022, APS March Meeting. [Oral Presentation]

"Ab initio prediction of mode-selected surface electron-phonon coupling strengths probed by Helium atom scattering (HAS) from the 3x1-reconstructed Nb(100) surface oxide," by Michelle M. Kelley, Caleb Thompson, Alison McMillan, Steven Sibener, Tomás Arias, 14-18 March 2022, APS March Meeting; Chicago, Illinois. [Oral presentation]

"Density-Functional Theory Results on Niobium-Zirconium Alloys for SRF Cavities," by Nathan Sitaraman, Michelle Kelley, Tomás Arias, Zeming Sun, Matthias Liepe, Zhaslan Baraissov, and David Muller, 23-25 May 2022, MRS Spring Meeting. [Oral presentation]

"A model of superconducting rf dissipation from Bogoliubov quasiparticles in a modified two-fluid framework," by Michelle Kelley, Sean Deyo, Nathan S Sitaraman, Thomas Oseroff, Danilo B Liarte, Matthias U Liepe, James P Sethna, Tomás A Arias, 5-10 March 2023, APS March Meeting. [Oral Presentation]

"Dissipation by Surface States in Superconducting Radio-Frequency Cavities," by Michelle Kelley, Sean Deyo, Nathan S Sitaraman, Thomas Oseroff, Danilo B Liarte, Matthias U Liepe, James P Sethna, Tomás A Arias, 25-30 June 2023, 21st International Conference on Radio-Frequency Superconductivity (SRF2023). [Invited Oral Presentation]

Posters

"Novel ab initio theory of inelastic intensities from helium atom scattering (HAS) events demonstrated on the 3x1-reconstructed Nb(100) surface oxide," by Michelle M. Kelley, , Alison McMillan, Caleb Thompson, Steven Sibener, Ravishankar Sundararaman, Tomás Arias, 22-25 August 2022, Psi-k Conference; Lausanne, Switzerlandmarh. [Poster presentation]

Awards and Honors

Michelle Kelley, Poster Commendation (Psi-k conference), Psi-k conference fellowship NCCR MARVEL, Invited Oral Talk by ISPC for SRF2023

Collaborating Groups

1. Arizona State University: Siddharth Karkare
2. Brigham Young University: Mark Transtrum

3. University of California at Los Angeles: James Rosenzweig
4. University of Chicago: Steven Sibener
5. Cornell University: Melissa Hines, Matthias Liepe, Jared Maxson, David Muller, James Sethna, Kyle Shen
6. University of Florida: Richard Hennig

PROJECT UPDATE: **Ab initio theory of many-body photoemission and of photomaterials (PHC)**

Arias student: Kevin Nangoi (Graduate Student), Tyler Wu (Graduate Student), Brandon Li (undergraduate student)

(A) Photoemission theory including many-body electron-phonon-photon interactions
We published and presented orally our detailed photoemission theory explaining the unexpectedly large MTE from Cs_3Sb and the importance of coherent three-body, electron-photon-phononn scattering processes. **CBB Collaborators:** Publication includes detailed comparison with experiments from the Karkare and Padmore groups. **References:**

- "Theory of photoemission from cathodes with disordered surfaces," by Pallavi Saha, Oksana Chubenko, J. Kevin Nangoi, Tomás Arias, Eric Montgomery, Shashi Poddar, Howard A. Padmore, and Siddharth Karkare, Journal of Applied Physics 133, 053102 (2023). DOI: 10.1063/5.0135629
- "Ab initio theory of coherent electron-phonon-photon processes in photoemission from single-crystal materials," by Johannes Kevin Nangoi, Siddharth Karkare, Ravishankar Sundararaman, Howard A Padmore, and Tomás A Arias, 14-18 March 2022, APS March Meeting. [Oral Presentation]

(B) Advances in understanding of the crystal and electronic structure of alkali antimonides
We submitted a manuscript (still in review) describing the first comprehensive study of the crystal and electronic structure of alkali antimonides, demonstrating that the commonly assumed structure is actually unstable for many of these materials and establishing the criteria for stability. **CBB Collaborators:** This work included information from Maxson and Galdi's CBB experiences with these materials and confirmation with x-ray data from collaborators of the Karkare group. **References:**

- "Ab initio study of the crystal and electronic structure of mono- and bi-alkali antimonides: Stability, Goldschmidt-like tolerance factors, and optical properties," by J. K. Nangoi, M. Gaowei, A. Galdi, J. M. Maxson, S. Karkare, J. Smedley, and T. A. Arias, Physical Review Materials (in review).

(C) PhD thesis
At the end of Summer 2022, we produced one new PhD (Kevin Nangoi, now a postdoc at UCSB) and in Fall 2022 took on a new student (Tyler Wu, a first generation student), who learned extremely rapidly and has already produced important results. **CBB Collaborators:** The thesis includes comparisons with results from the Maxson, Karare and Shen groups. **References:**

- "Ab initio study of photoemission processes and photocathode materials for next-generation high-brightness electron emitters," Kevin J. Nangoi, Doctor of Philosophy in Physics, Cornell University, August 2022.

(D) *Ab initio* Lippmann-Schwinger equation results for electron scattering from graphene

Our new student. Tyler Wu, has made incredible progress, developing the techniques and proof-of-principle Lippmann-Schwinger calculations from Nangoi's thesis above into a practical tool that has addressed issues of direct interest to the Center. For example, Wu has predicted the scattering effects on quantum efficiency and MTE that can be expected from coating photocathodes with protective graphene overlayers. Figure 1 (under "Imagery" below) shows the first *ab initio* predictions of these effects, showing that, above 10 meV excess energy, electrons experience nearly perfect transmission and that. below this energy, transmission drops off rapidly (Figure 1a). We also find that, for angles up to 30°, transmission is nearly constant (Figure 1b). Taken together, these results indicate that such coatings will have little impact on quantum efficiency, unless cathodes are operated within a few meV of threshold, and that a graphene coating will have little effect on MTE for photocathode materials with already low MTE. Work is currently underway to explore the corresponding impacts from other 2D materials of potential interest to CBB, including BN and transition metal dichalcogenides. **CBB Collaborators:** Hines provides samples and Karakare provides experimental measurements of quantum efficiency and MTE.

(E) *Ab initio* Lippmann-Schwinger predictions of photoemission transverse momentum distributions in graphene.

One unresolved weakness in Nangoi's thesis and the publication above on photoemission theory was lack of access to the outgoing propagating electronic wave functions, making *ab initio* computation of direct excitations into vacuum impossible and requiring uncontrolled approximations for the transmission coefficients of bulk-excited electrons into vacuum. Our new Lippmann-Schwinger capability gives direct access to the vacuum propagating states and removes these limitations. Our new student has already demonstrated the new capability in predicting the transverse momentum distribution (TMD) to be expected for graphene photoemission, which is always a direct excitation into vacuum and could not be calculated using Nangoi's original techniques. Figure 2 (under "Imagery" below) shows our preliminary predictions for the TMD from the highest occupied graphene band due to photons incoming at normal incidence with an energy of 7.0 eV and with linear polarization along x. We will continue to refine these calculations and also explore other 2D materials of interest to the center as they arise. **CBB Collaborators:** Karakare and Shen to provide distributions of photoemitted electrons.

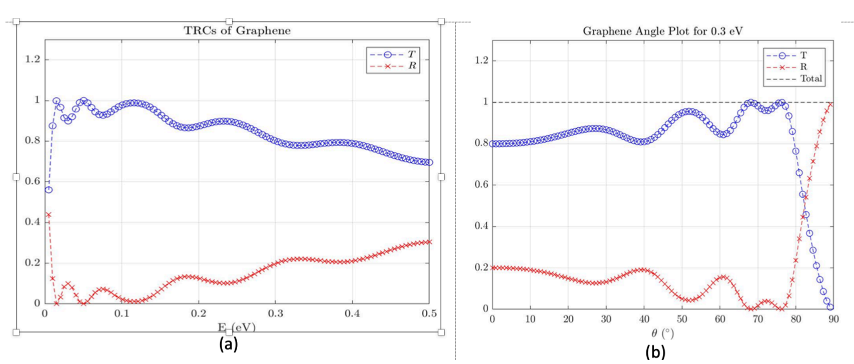


Figure 1. *Ab initio* prediction of transmission (blue) and reflection (red) probabilities for electrons impinging on graphene for (a) normal incidence as a function of kinetic energy and (b) 0.3 eV kinetic energy as a function of angle of incidence.

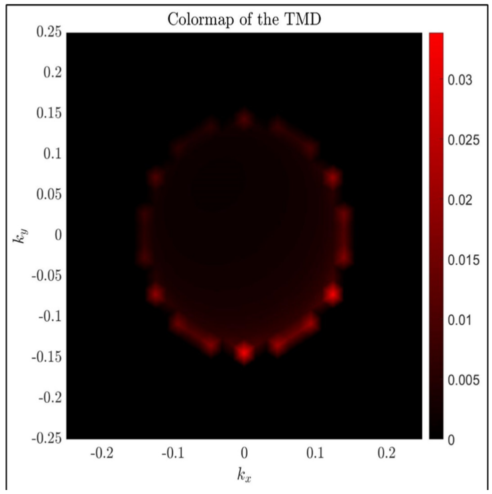


Figure 2. Preliminary results for *ab initio* Lippmann-Schwinger predictions of transverse momentum distribution of photoelectrons from graphene resulting from from the highest occupied band due to photons incoming at normal incidence with an energy of 7.0 eV and with linear polarization along x.

PROJECT UPDATE: *Ab initio* studies of lattice and electron excitations relevant to SRF performance and inverse-Q behavior

Arias students: Michelle Kelley (Graduate Student), Michelle Kwok (REU student)

(A) *Ab initio* prediction of Helium atom scattering (HAS)

Using density-functional theory (DFT), we calculated surface lattice dynamics on both the bare Nb(001) niobium and the 3x1-O/NbO(001) niobium monolayer-oxide surfaces for direct comparison with helium atom scattering (HAS) of the Sibener group. (See Figure 1, under "Imagery" below.) We have developed a new and fully *ab initio* framework for inelastic–atom surface scattering with Sundararaman (RPI) and are preparing a manuscript for publication. **CBB collaborators:** Caleb Thompson and Michael Van Duinen of the Sibener group provided experimental data. **References:**

- "A combined helium atom scattering and density-functional theory study of the Nb(100) surface oxide reconstruction: phonon band structures and vibrational dynamics," by Alison A. McMillan, Caleb J. Thompson, Michelle M. Kelley, Jacob D. Graham, Tomás A. Arias, and S.J. Sibener, The Journal of Chemical Physics 156, 124702 (March 2022), DOI: 10.1063/5.0085653
- "Ab initio prediction of mode-selected surface electron-phonon coupling strengths probed by Helium atom scattering (HAS) from the 3x1-reconstructed Nb(100) surface oxide," by Michelle M. Kelley, Caleb Thompson, Alison McMillan, Steven Sibener, Tomás Arias, 14-18 March 2022, APS March Meeting; Chicago, Illinois. [Oral presentation]
- "Novel ab initio theory of inelastic intensities from helium atom scattering (HAS) events demonstrated on the 3x1-reconstructed Nb(100) surface oxide," by Michelle M. Kelley, , Alison McMillan, Caleb Thompson, Steven Sibener, Ravishankar Sundararaman, Tomás Arias, 22-25 August 2022, Psi-k Conference; Lausanne, Switzerlandmarh. [Award winning poster presentation]

(B) Theoretical studies of anti-Q effects in Nb

Typically the quality factor of SRF cavities degrades with accelerating field, but some Nb SRF cavities exhibit quality factors that increase with field. Optimizing SRF cavity performance requires understanding all of the competing effects at play that can produce such a so-called 'anti-Q slope'. Solving the Bogolioubov-de Gennes (BdG) self-consistent field equations at the surface of a superconductor in an AC magnetic field, we have found quasiparticle surface states with energies below the superconducting gap that emerge and disappear with the RF field. Using *ab initio* calculations, we then obtained estimates of the associated loss mechanisms and demonstrated how such bound surface states contribute to anti-Q slopes through a new, "three-fluid" model. Properly distinguishing the surface-localized bound normal fluid states from the bulk normal continuum states, we are able to reproduce the observed anti-Q slope behavior when the localization of the surface-bound normal fluid leads to scattering rates exceeding those of the continuum states by roughly a factor of 5. (See Figure 2, under "Imagery" below.) **CBB collaborators:** Thomas Oseroff of the Liepe Group and Liepe himself provided deep insight, experience, and knowledge regarding the experimental conditions producing an anti-Q slope, and Sean Deyo of the Sethna provided initial theory and code which required a number of corrections, and Sethna guided the project. **References:**

- "Dissipation by surface states in superconducting radio-frequency cavities," by Sean Deyo, Michelle Kelley, Nathan Sitaraman, Thomas Oseroff, Danilo Liarte, Tomás Arias, Matthias Liepe, and James Sethna, Physical Review B106, 104502 (September 2022), DOI: 10.1103/PhysRevB.106.104502.
- "A model of superconducting rf dissipation from Bogoliubov quasiparticles in a modified two-fluid framework," by Michelle Kelley, Sean Deyo, Nathan S Sitaraman, Thomas Oseroff, Danilo B Liarte, Matthias U Liepe, James P Sethna, Tomás A Arias, 5-10 March 2023, APS March Meeting. [Oral Presentation]
- "Dissipation by Surface States in Superconducting Radio-Frequency Cavities," by Michelle Kelley, Sean Deyo, Nathan S Sitaraman, Thomas Oseroff, Danilo B Liarte, Matthias U Liepe, James P Sethna, Tomás A Arias, 25-30 June 2023, 21st International Conference on Radio-Frequency Superconductivity (SRF2023). [Invited Oral Presentation]

(C) Core-level shifts on SRF materials

Michelle Kelley mentored an REU student (Michelle Kwok) this past summer who performed DFT calculations on Nb, NbO_2, Nb_2O_5, Nb_3Sn, Nb-Zr, and various defects within these materials to identify the core-electron energy levels. This work characterized, for example, how the energy levels of the 4s shell in niobium shifts based on its chemical environment, and the results were used to help interpret Sun's x-ray photoelectron spectroscopy measurements. **CBB collaborators:** Data from Zeming Sun of the Liepe group.

(D) PhD Thesis

The graduating graduate student (Michelle Kelley) has been working on her thesis over the last several months. It will be defended on April 20, 2023, and we anticipate her graduation this spring. Michelle will then be continuing development of novel *ab initio* methods under Ravishankar Sundararaman at RPI. **References:**

- "Effective quantum many-body theories: studies of electronic structure and electron-phonon couples properties near interfaces in superconductors," Michelle Marie Kelley, April 2023.

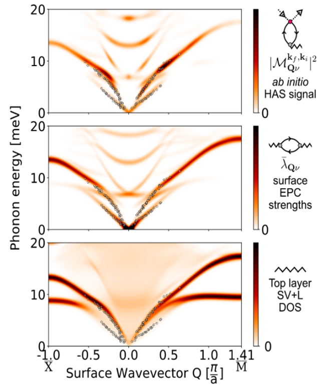


Figure 1. Colormaps showing predicted inelastic helium atom scattering (HAS) signals from the Nb(100) surface at three levels of theory. Colorbar labels show the Feynman diagram elements considered at each level. The bottom panel considers only phonons to show the top-layer phonon density-of-states for shear vertical (SV) and longitudinal (L) polarizations. The middle panel incorporates the electron--phonon vertex to give the inelastic HAS predictions estimated from surface electron-phonon coupling (EPC) strengths. The top panel includes both the electron-phonon and helium-electron vertices, giving the final prediction of the fully *ab initio* HAS calculation. Black circles in each panel correspond to measured helium-scattering data from Hulpke et al. (1992) Phys. Rev. B 45, 1820, which are best represented at the deepest level of theory.

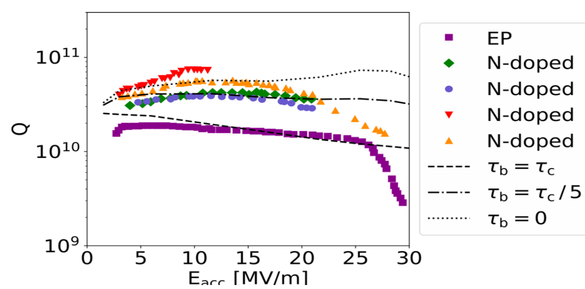


Figure 2. Quality factor vs. field amplitude for a 1.3 GHz niobium cavity. Markers show experimental data from Grassellino *et al.* for a selection of electropolished (EP) or nitrogen-doped cavities. The black lines give the results of the three-fluid model for two limiting cases and one intermediate case, all with continuum scattering-lifetime τ_c=0.2 ps. The dashed line shows the quality factor assuming all states scatter at the same rate, the dotted line shows the quality factor assuming bound states scatter much faster than continuum states, and the dashed-dotted line shows the intermediate case of bound states scattering about a factor of 5 faster than continuum states.

PROJECT UPDATE: *Ab initio* exploration of beyond-Nb SRF materials for low cooling power and high field performance

Arias students: Nathan Sitaraman (Graduate Student)

(A) Microscopic hydride model for prediction of high-field Q-slope of Nb

We developed a kinetic Monte Carlo model of the formation of hydride clusters in Nb as a function of doping profile (Figure 1a, under "Imagery" below). We also computed superconducting critical temperatures and normal-state resistivities of hydrides and passed all of this information to the Transtrum group. The Transtrum group then predicted the resulting high-field Q-slope (HFQS) behavior through time-dependent Ginzburg-Landau calculations, finding very promising agreement with experiment based upon our *ab initio* inputs (Figures 1b and 1c, under "Imagery" below). This work is described in Sitaraman's PhD thesis, and we anticipate preparation of a manuscript in the near future. **CBB Collaborators:** Aiden Harbick of Transtrum group performed calculations based upon our data.

(B) Nb-Zr alloys

Our work regarding Nb-Zr progressed significantly over the reporting period as we continued to refine our theoretical and experimental understanding. We helped to identify the rocksalt phases in the Muller group's diffraction data of electrochemically prepared Nb-Zr samples, and we also demonstrated that the vast over prediction of T_c from the virtual crystal approximation (VCA) calculations of the Hennig group was due to the lack of Jahn-Teller-Peierls effects within that theory, and we quite accurately reproduced the traditional experimental findings. We also demonstrated that novel, ordered alloy structures may exist up to a stability limit which we identified and which included the higher T_c's observed by Zeming Sun of the Liepe group. (See Figure 2 under "Imagery" below.) A manuscript describing this work is currently under review. **CBB Collaborators:** Zeming Sun of the Liepe group grew the material performed the T_c experiments, Benjamin Francis of the Transtrum group performed Ginzburg-Landau calculations of the super-heating fields, Ajinkya Hire of the Hennig group performed the VCA calculations of both T_c and coherence lengths, and Zhaslan Baraissov of the Muller group performed microscopy on the samples. **References:**

- Nathan Sitaraman, Zeming Sun, Benjamin Francis, Ajinkya Hire, Thomas Oseroff, Zhaslan Baraissov, Tomas Arias, Richard Hennig, Matthias Liepe, David Muller, and Mark Transtrum, "Enhanced surface superconductivity of niobium by zirconium doping", Phys. Rev. Applied (in review).
- "Density-Functional Theory Results on Niobium-Zirconium Alloys for SRF Cavities," by Nathan Sitaraman, Michelle Kelley, Tomás Arias, Zeming Sun, Matthias Liepe, Zhaslan Baraissov, and David Muller, 23-25 May 2022, MRS Spring Meeting. [Oral presentation]

(C) PhD thesis and additional research by Sitaraman

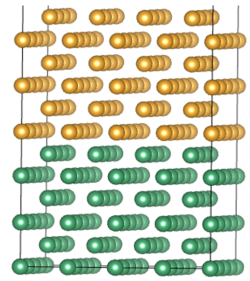
Sitaraman completed his PhD thesis, and is now pursuing a postdoc in accelerator physics, a clear success story for CBB in bringing scientists from a range of backgrounds into accelerator physics. Sitaraman also was productive in a number of other areas beyond those above and for which there is insufficient space to describe in detail here. Briefly, he continued providing theory-inspired nucleation recipes for Nb_3Sn which were then explored by Liana Shpani of the Liepe group, and, in collaboration with Femilab, he gave the first explanation for the mysterious formation of ω phase Nb in sputtered samples, showing that this harmful transition results from the presence of vacancies. **CBB Collaborators:** Liana Shpani and Ryan Porter of the Liepe group. **References:**

- Jaeyel Lee, Nathan Sitaraman, Zuhawn Sung, Akshay A. Murthy, Grigory V. Eremeev, Tomás Arias, Anna Grassellino, and Alexander Romanenko, "Stress-induced omega (ω) phase transition in Nb thin films for superconducting qubits", submitted to Phys. Rev. Applied
- "Theory work on SRF materials," Nathan S. Sitaraman, Doctor of Philosophy in Physics, Cornell University, November 2022.

(D) Au treatment of Nb cavities

After Sitaraman's graduation, I have been working with a new student, Cristóbal Méndez. Recent experiments by Thomas Oseroff (former CBB grad, current Liepe group postdoc) found that stripping the native oxide from the niobium surface and depositing approximately 1 monolayer of gold results in a surface with lower RF resistance and higher quench field. To extend our understanding of this system and explore new experimental options, in collaboration with Nathan Sitaraman, now of the Liepe group, Méndez has been exploring the growth of gold films on niobium (Au/Nb).

Niobium is a bcc metal and gold is a fcc metal. For very low gold coverages (much less than 1 monolayer) gold atoms will occupy bcc lattice sites. For very high gold coverages (hundreds of monolayers) the gold layer will revert to its fcc ground-state structure and thus develop an incoherent interface with the underlying niobium. To address the point at which this happens, the new student, Méndez has calculated the following energies from first principles: the surface energy of niobium without a gold coating (0.76 eV/surface atom), the energy of the gold-on-niobium interface (-0.23 eV/interface atom, figure to the right), the surface energy of the coherent gold film (0.37 eV/surface atom), and the excess energy of gold atoms in the epitaxial gold film (0.018 eV/atom). The result is that, up to a number of layers n = (0.23 eV + 0.76 eV - 0.37 eV) / (0.018 eV) = 34, it is more energetically favorable for gold to form the interface than to exist in its bulk form.



We stress that this result is preliminary and that additional calculations are underway to ascertain the energy of an incoherent interface between gold and niobium. Future work will include the energies controlling intermixing between the gold and niobium layers, the impact of gold overlayers on the superconducting properties, the contact resistance of the interface, and exploration of other potential materials including palladium. **CBB Collaborators:** Nathan Sitaraman of the Liepe Group.

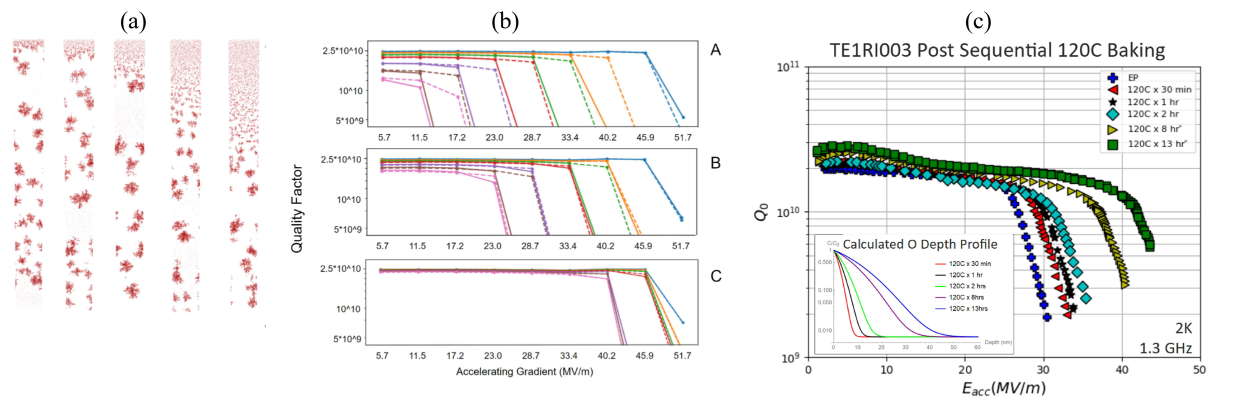


Figure 1. Prediction of high-field Q-slope (HFQS) phenomenon: (a) kinetic Monte-Carlo model of hydride formation, (b) Transtrum group predictions based on our *ab initio* T_c and normal-phase resistivity calculations for hydrides of different radii (colored lines) at depths of 0.5, 1.0, and 1.5 penetration depths (A, B, C, respectively), (c) experimental data showing comparable behaviors for reasonable parameter values.

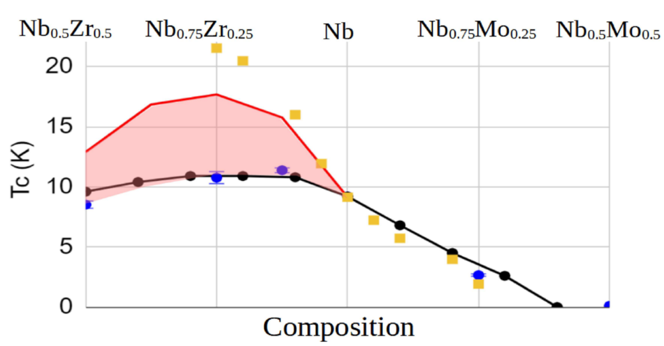


Figure 2. Superconducting transition temperature versus composition for Zr-Nb-Mo alloy system: experiment (black circles), simple virtual crystal approximation (yellow squares), our *ab initio* random supercell theory (blue circles), our identification of the stability limit (red curve). Recent experiments of electrochemically grown Nb-Zr films showed T_c values significantly above expectations from past experiments but within our expected stability range.

Bazarov, Ivan (Cornell)

Publications

Papers

J. K. Bae, M. Andorf, A. Bartnik, A. Galdi, L. Cultrera, J. Maxson, and I. Bazarov, “Operation of Cs-Sb-O activated GaAs in a high voltage DC electron gun at high average current,” May 2022, doi: 10.48550/arXiv.2205.13632. [Online]. Available: http://arxiv.org/abs/2205.13632

M. Gordon, W. H. Li, M. B. Andorf, A. C. Bartnik, C. J. R. Duncan, M. Kaemingk, C. A. Pennington, I. V. Bazarov, Y.-K. Kim, and J. M. Maxson, “Four-dimensional emittance measurements of ultrafast electron diffraction optics corrected up to sextupole order,” *Phys. Rev. Accel. Beams*, vol. 25, no. 8, p. 084001, Aug. 2022, doi: 10.1103/PhysRevAccelBeams.25.084001.

M. Andorf, J. K. Bae, A. Bartnik, L. Cultrera, J. Maxson, and I. Bazarov, “HERACLES: A high-voltage DC test beamline for high average current photocathodes,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 1052, p. 168240, Jul. 2023, doi: 10.1016/j.nima.2023.168240.

C. M. Pierce, D. B. Durham, F. Riminucci, S. Dhuey, I. Bazarov, J. Maxson, A. M. Minor, and D. Filippetto, “Experimental Characterization of Photoemission from Plasmonic Nanogroove Arrays,” *Phys. Rev. Appl.*, vol. 19, no. 3, p. 034034, Mar. 2023, doi: 10.1103/PhysRevApplied.19.034034.

L. Cultrera, E. Rocco, F. Shahedipour-Sandvik, L. D. Bell, J. K. Bae, I. V. Bazarov, P. Saha, S. Karkare, and A. Arjunan, “Photoemission characterization of N-polar III-nitride photocathodes as candidate bright electron beam sources for accelerator applications,” *Journal of Applied Physics*, vol. 131, no. 12, p. 124902, Mar. 2022, doi: 10.1063/5.0076488.

Posters and Talks

W.H. Li, M. Gordon, M.B. Andorf, A.C. Bartnik, C.J.R. Duncan, M. Kaemingk, S.J. Levenson, C. A. Pennington, I.V. Bazarov, Y.-K. Kim, J.M. Maxson, "Four-Dimensional Emittance Measurements and Correction of UED Optics up to Sextupole Order", Proceedings of the 13th International Particle Accelerator Conference, Bangkok, Thailand (2022) paper MOPOTK026.

S.J. Levenson, M.B. Andorf, I.V. Bazarov, J. Encomendero, D. Jena, J.M. Maxson, V.V. Protasenko, and H.G. Xing, "Characterization of Various GaN Samples for Photoinjectors", Proceedings of the 13th International Particle Accelerator Conference, Bangkok, Thailand (2022) paper MOPOTK027.

J. Bae, M. Andorf, I. Bazarov, J. Maxson, L. Cultrera, A. Galdi, "Optimizing Activation Recipe with Cs, Te, O for GaAs-Based Photocathodes", Proceedings of the 13th International Particle Accelerator Conference, Bangkok, Thailand (2022) paper MOPOMS004.

S.J. Levenson, M.B. Andorf, I.V. Bazarov, D.C. Burke, J.M. Maxson, D.L. Rubin, S.T. Wang, "A Path-Length Stability Experiment for Optical Stochastic Cooling at the Cornell Electron Storage Ring", Proceedings of the 13th International Particle Accelerator Conference, Bangkok, Thailand (2022) paper WEPOMS030.

S.J. Levenson, M.B. Andorf, I.V. Bazarov, D.C. Burke, J.M. Maxson, D.L. Rubin, S.T. Wang, "Light Path Construction for an Optical Stochastic Cooling Stability Test at the Cornell Electron Storage Ring", Proceedings of the 13th International Particle Accelerator Conference, Bangkok, Thailand (2022) paper WEPOMS031.

V.A. Khachatryan, M.B. Andorf, I.V. Bazarov, W.F. Bergan, J.A. Crittenden, S.J. Levenson, J.M. Maxson, D.L. Rubin, J.P. Shanks, S.T. Wang, "Helical Wiggler Design for Optical Stochastic Cooling at CESR", Proceedings of the 13th International Particle Accelerator Conference, Bangkok, Thailand (2022) paper THPOPT066.

M.B. Andorf, J.K. Bae, A.C. Bartnik, L. Cultrera, J.M. Maxson, I.V. Bazarov, "HERACLES: A High Average Current Electron Beamline for Lifetime Testing of Novel Photocathodes", Proceedings of the 13th International Particle Accelerator Conference, Bangkok, Thailand (2022) paper THPOMS036.

C.M. Pierce, I.V. Bazarov, J.M. Maxson, S. Cabrini, D.B. Durham, D. Filippetto, A. Kachwala, S.S. Karkare, F. Riminucci, A. Minor, Towards High Brightness from Plasmon-Enhanced Photoemitters, North American Particle Accelerator Conference, Albuquerque, NM (2022) TUYD4

Collaborating Groups

1. Cornell University: Jared Maxson, Melissa Hines

PROJECT UPDATE: Improvement of Photocathode Robustness with Alkali-Halides

Bazarov student: Chris Pierce (Graduate Student)

According to the CBB strategic plan, one of the goals is “developing new robust work function reducing layers for GaAs or other III-V materials.“ In order to meet our CBB deliverables, a more robust and efficient photocathode is required. Cesium Iodide (CsI) photocathodes are very robust and not susceptible to chemical poisoning like many other photocathode materials.

Although the work function of CsI is far too large for practical use in a photoinjector, it has been used as an efficient and protective coating layer for other photocathodes [1]. GaAs photocathodes, while widely used (specifically for spin-polarized electron beam production), are very sensitive to chemical poisoning and often have short operational lifetimes when activated to Negative Electron Affinity (NEA). with Cs or Cs/O2. While other materials have been used to protect the NEA activation layer on GaAs photocathodes [2-3], Samuel Levenson's CBB project has demonstrated that a CsI layer can be used to enhance the dark lifetime of a GaAs photocathode activated with Cs to NEA. After tuning the growth parameters of a CsI film in our dedicated CsI growth chamber, studies commenced with GaAs photocathodes.

Other works have demonstrated the improvement in the quality of CsI films upon annealing [4-5]. As shown in Fig. 1 below, a GaAs photocathode activated to NEA with Cs only will have its QE decay with only a few Langmuirs of residual gas exposure. Once a 1 nm CsI layer is grown on the surface (shown in the left AFM scan in Fig. 2), a Cs NEA activation produces a low-QE and short-lived NEA layer. However, annealing the CsI layer at 600 degrees C will smooth out the surface (as shown in the right AFM scan in Fig. 2) and produce a much better NEA layer when activated with Cs. After two such annealing cycles, we see a factor of almost 13 improvement in lifetime. Current efforts are underway to apply a Cs/O2 activation layer to an annealed CsI binder, as well as measure the spin-polarization of such a photocathode.

This work is done in close collaboration with M. B. Andorf and J. M. Maxson and A. Galdi. XPS measurements have been done on CsI photocathodes in collaboration with the CBB members from the Hines group (Q. Zhu, D. Somaratne)

References:

1. L. Kong, et al. Chemical Physics Letters 621 (2015) 155–159
2. L. Cultrera et al. Phys. Rev. Accel. Beams 23, 023401
3. J.K. Bae et al. Applied Physice Letters 112(15):154101
4. J . Almeida et al. I Nucl. Instr. and Meth. in Phys. Res. A 367 (1995) 337-341
5. T. Jing et al, IEEE Trans. Nucl. Sci. NS-41 (1994) 903

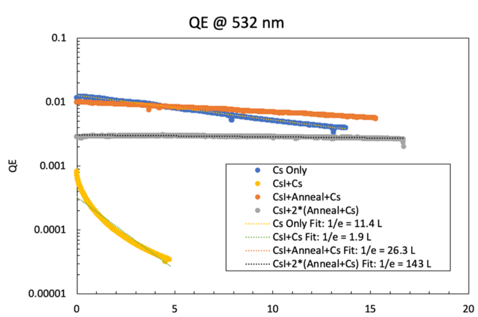


Fig. 1. The dark lifetime of a Cs:GaAs photocathode at different stages of the CsI NEA-preservation process. As shown in the plot, a 1 nm CsI layer annealed twice increases the cathode's dark lifetime by a factor of 13.

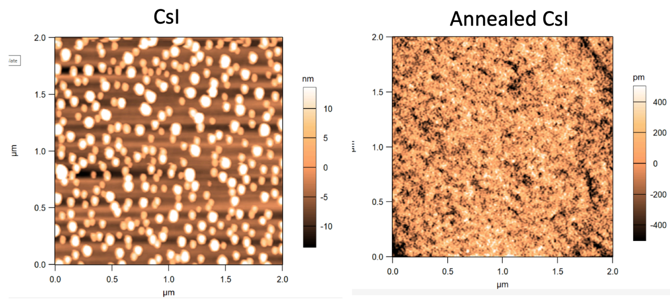


Fig. 2. AFM scans of a pre- and post- annealed CsI layer grown on a GaAs photocathode. Immediately after growth (left), the CsI clusters in "islands," that are ~10 nm tall (RMS height 7.9 nm). After annealing at 600 degrees C, we see a very smooth and continuous CsI layer, with only a 191 pm RMS height. Other AFM scans confirmed that this surface differs greatly from an ordinary GaAs surface.

Biedron, Sandra (University of New Mexico)

Publications

Papers

A. Aslam, M. Babzien, and S. Biedron, "Applications of Machine Learning in Photo-Cathode Injectors," Proceedings of the 5th North American Particle Accelerator Conference, vol. NAPAC2022, p. 2 pages, 0.300 MB, 2022, doi: 10.18429/JACOW-NAPAC2022-TUPA41. [Online]. Available: https://jacow.org/napac2022/doi/JACoW-NAPAC2022-TUPA41.html

Talks

S.G. Biedron, Invited Speaker, "Data Driven Concepts For Laser And Particle Accelerator-Based User Facility Systems: All Systems Work Together," Invited Session - Let There Be Data: Analyzing Data From Lasers And Light Sources," Conference on Data Analysis, Santa Fe, New Mexico, March 5-7 2023.

S.G. Biedron (Moderator), "Workforce Pipeline Panel: Meeting the Future Needs Today," with panelists James Hadaway (Principal Research Scientist, Center for Applied Optics, The University of Alabama in Huntsville); Justin Martin (Chief Engineer, Raytheon High Energy Laser Product Line); Laura McGill (Deputy Lab Director and Chief Technology Officer, Sandia National Labs); Martin Richardson (Director, Center for Directed Energy, Pegasus and Northrop Grumman Professor of Optics, Physics Electrical and Computer Engineering, University of Central Florida); and Frank Sage (Survivability, Vulnerability and Assessment Director, White Sands Missile Range) at the 2023 Joint Conference on Test & Evaluation (T&E) Support to Prototyping &

Experimentation, hosted by the International Test and Evaluation Association (ITEA) and the Directed Energy Professional Society (DEPS), 30 January - 2 February 2023.

S.G. Biedron, “Superconductivity in Particle Accelerators: A Bridge to Industrial, Security, and Medical Applications - Dr. Sandra Biedron,” Seminars of Applied Superconductivity, CBMM | Niobium, Araxá, Brazil, November 18, 2022.

S.G. Biedron, “Knowledge Transfer,” Center for Bright Beams, National Science Foundation Science and Technology Center, Site Visit, Ithaca, New York, 12-13 September 2022.
S. G. Biedron, Invited Speaker, “Accelerator Development for Global Security,” XXXI International Linear Accelerator Conference, Liverpool, UK, 29 August 29 - 2 September 2022.

S.G. Biedron, Invited Speaker and Panel Member, “Opportunity to Innovate: Five Opportunities for Collaborative Innovation for Decarbonized Discovery, Innovation, Design and Manufacturing,” Sustainability Summit at SEMICON WEST, San Francisco, California, 13 July 2022. (Other authors M. Peters, R. Rosner, J.L. Sarrao)

S.G. Biedron, Invited Speaker and Panel Member, Industrial Session, “Present status and opportunities for implementing disruptive technologies arising in particle accelerator R&D industrial market,” International Particle Accelerator Conference, Bangkok, Thailand, 15 June 2022.

S.G. Biedron, “Activities in Nuclear Energy and Particle Accelerators,” Japan Atomic Energy Commission, Tokyo, Japan, 10 June 2022. Cabinet Briefing.

S.G. Biedron, “Applications of Machine Learning in Photoinjectors,” Center for Bright Beams Annual Meeting, UCLA, Los Angeles, California, 1 June 2022.

A,Aslam, “Machine learning method for the control of ultrafast lasers,” Twenty-fourth Annual Directed Energy Science & Technology Symposium, 26 April 2022, Mobile, Alabama.

S.G. Biedron, “Analytical Research Tools (and Applications Derived from these Tools) helping us understand and better our universe,” Trinity Christian College Colloquium, 14 April 2022.

S.G. Biedron, “Knowledge Transfer Strategic Planning,” National Science Foundation's Center for Bright Beams, External Advisory Board, April 5, 2022.

Posters

M. Caskey, J. Diaz Cruz, S. Biedron, A. Hagberg, “Gradient Descent Optimization and Resonance Control of Superconducting RF Cavities,” presented at NAPAC’22, Albuquerque, New Mexico, USA, Aug. 2022, paper MOPA11.

A. Aslam, M. Babzien, and S. Biedron, “Applications of Machine Learning in Photo-Cathode Injectors,” presented at NAPAC’22, Albuquerque, NM, USA, Aug 2022, paper TUPA41

A. Aslam, M. Babzien, and S. Biedron, “Applications of Machine Learning in Photo-Cathode Injectors,” presented at CODA March 2023.

Collaborating Groups

1. Arizona State University: Siddharth Karkare
2. Cornell University: Matthias Liepe

PROJECT UPDATE: Applications of Machine Learning in Compact Photoinjectors

Biedron student: Aasma Aslam (graduate student)

Through funding provided by the NSF Center for Bright Beams (CBB), Beam Dynamics and Control (BDC) focus area, I am performing a project that can significantly benefit from Machine Learning (ML) and Neural Network (NN) based modeling. Neural network-based controllers have been shown to improve several complex systems and can be applied to beam dynamics to help provide optimal control. These techniques can be applied to several beam-based commercial devices, such as electron microscopes, to improve the measurement outcomes.

For this project, my advisor, Sandra Biedron, connected me with the Accelerator Test Facility which is a Department of Energy’s user facility. My main point of contact is one of the facility’s laser physicists, Marcus Babzien. Our experimental goal is to improve the facility’s electron beam through laser pulse shaping of the photocathode-RF gun’s drive laser system.

We will operate the laser and photocathode gun systems and their diagnostics for data collection. The data will be used to implement a controls approach to improve brightness of electron beam. The laser system for this main electron source is located at main ATF beamline in building 820.

Our goal is to align with the objectives presented in the CBB's strategic plan. Neural network-based controllers aid in achieving optimal outcomes in beam dynamics and control. The knowledge and techniques we develop here can also be applied to MeV-class ultrafast electron diffraction systems and microscopes, such as at the other ATF beamline and at SLAC as well as free-electron lasers and other coherent radiators.
Moreover, outside of Marcus and Sandra, I am also collaborating with Salvador Sosa (UNM and LNL) and Trudy Bolin (UNM and Element Aero), who are members of our team, on simulating the ATF electron gun using VSIM [1][2][3].

The goal is to develop a comprehensive model of electron beam generation through the gun including the laser pulse shape model, which can be compared to subsequent emittance measurements. In 2023, I plan to work more closely with the Cornell team and will start by visiting them during the CBB meeting at Cornell.

Dr. Biedron's main idea is to leverage machine learning skills on experimental data to improve the electron beam by reducing the emittance.

Chubenko, Oksana (Northern Illinois University)

Publications

Papers

P. Saha, O. Chubenko, J. K. Nangoi, T. Arias, E. Montgomery, S. Poddar, H. A. Padmore, and S. Karkare, Theory of photoemission from cathodes with disordered surfaces, J. Appl. Phys. 133, 053102 (2023).

A. Kachwala, O. Chubenko, D. Kim, E. I. Simakov, and S. Karkare, Quantum efficiency, photoemission energy spectra, and mean transverse energy of ultrananocrystalline diamond photocathode, J. Appl. Phys. 132, 224901 (2022).

P. Saha, O. Chubenko, G. S. Gevorkyan, A. Kachwala, C. J. Knill, C. Sarabia-Cardenas, E. Montgomery, S. Poddar, J. T. Paul, R. G. Hennig, H. A. Padmore, and S. Karkare, Physically and chemically smooth cesium-antimonide photocathodes on single crystal strontium titanate substrates, Appl. Phys. Lett. 120, 194102 (2022).

Talks

A.H. Kachwala, O. Chubenko, S.S. Karkare, R. Ahsan, H.U. Chae, R. Kapadia, On-Chip Photonics Integrated Photocathodes, North American Particle Accelerator Conference, Albuquerque, NM (2022) WEPA65

Collaborating Groups

Arizona State University: Siddharth Karkare
Cornell University: Jared Maxson
Northern Illinois University: Philippe Piot

PROJECT UPDATE: Monte Carlo modeling of photoemission from semiconductors

Chubenko students: Daniel Franklin (Graduate Student)

Cs-Sb compound photocathodes demonstrate superior photoemission properties. Depending on the growth conditions and the choice of a substrate, these materials can be grown in a form of CsSb, Cs3Sb, etc. Therefore, one may suggest that these photocathodes demonstrate different optical, electrical, and other material properties resulting in different photoemission response.

The primary goal of this project is to gain a deeper understanding of the mechanisms that govern MTE and QE by modeling the optical properties of Cs-Sb-compound materials. To achieve this goal, Daniel franklin, who had joined CBB in January 2023, have been conducting research to identify the relevant literature and develop methods for modeling the optical properties of semiconductors.

Additionally, Daniel have developed Mathematica code based on Adachi's model of the dielectric function to test various possible model parameters and compare the calculated values of absorption coefficient and reflectivity against experimental data available in literature.

PROJECT UPDATE: Photocathodes under realistic accelerator conditions

Chubenko students: Tariqul Hasan (Graduate Student)

The NIU is conveniently located in close proximity to two national labs, Fermi National Accelerator Laboratory and Argonne National Laboratory. Both these labs have capabilities of testing CBB-developed photocathodes under realistic photoinjectror conditions and expresses their intention to collaborate with on this subject. The current focus of the project is on the development of a photocathode growth chamber that will be compatible with Argonne's or/and FermiLab's beamline. The chamber will be used to grow high-quality photocathodes, which then will be tested at these facilities. Close proximity of the National Labs can significantly reduce the likelihood of degradation in photocathode quality, which typically occurs during transportation over long distances.

Tariqul Hasan has recently joined CBB (Januarry 2023) and has mostly conducted literature reviews to gain a good understanding of the photoemission process and the requirements to produce bright electron beams for different accelerator applications. Tariqul has also familiarized himself with the basics of working with UHV components. Specifically, he started modifying the growth chamber previously used for growing CsTe photocathodes.

Additionally, Tariqul has read papers on measuring drift velocity in semiconductors, which can provide clues on how to develop an experimental setup for measuring drift velocity and other transport characteristics in Cs-Sb films.

Hennig, Richard (University of Florida)

Publications

Papers

S. A. Willson, R. G. Farber, A. C. Hire, R. G. Hennig, and S. J. Sibener, "Submonolayer and Monolayer Sn Adsorption and Diffusion Behavior on Oxidized Nb(100)," *J. Phys. Chem. C*, Jan. 2023, doi: 10.1021/acs.jpcc.2c08458. [Online]. Available: https://doi.org/10.1021/acs.jpcc.2c08458

J. Lim, A. C. Hire, Y. Quan, J. S. Kim, S. R. Xie, R. S. Kumar, D. Popov, C. Park, R. J. Hemley, J. J. Hamlin, R. G. Hennig, P. J. Hirschfeld, and G. R. Stewart, "Creating superconductivity in WB2 through pressure-induced metastable planar defects," *arXiv:2109.11521 [cond-mat]*, Sep. 2021, doi: 10.48550/arXiv.2109.11521. [Online]. Available: http://arxiv.org/abs/2109.11521

A. C. Hire, S. Sinha, J. Lim, J. S. Kim, P. M. Dee, L. Fanfarillo, J. J. Hamlin, R. G. Hennig, P. J. Hirschfeld, and G. R. Stewart, "High critical field superconductivity at ambient pressure in MoB2 stabilized in the P6/mmm structure via Nb substitution," *Phys. Rev. B*, vol. 106, no. 17, p. 174515, Nov. 2022, doi: 10.1103/PhysRevB.106.174515.

J. Gibson, A. Hire, and R. G. Hennig, "Data-augmentation for graph neural network learning of the relaxed energies of unrelaxed structures," *npj Comput Mater*, vol. 8, no. 1, pp. 1–7, Sep. 2022, doi: 10.1038/s41524-022-00891-8.

P. Saha, O. Chubenko, G. S. Gevorkyan, A. Kachwala, C. J. Knill, C. Sarabia-Cardenas, E. Montgomery, S. Poddar, J. T. Paul, R. G. Hennig, H. A. Padmore, and S. Karkare, "Physically and chemically smooth cesium-antimonide photocathodes on single crystal strontium titanate substrates," *Appl. Phys. Lett.*, vol. 120, no. 19, p. 194102, May 2022, doi: 10.1063/5.0088306.

N. S. Sitaraman, Z. Sun, B. Francis, A. C. Hire, T. Oseroff, Z. Baraissov, T. A. Arias, R. Hennig, M. U. Liepe, D. A. Muller, and M. K. Transtrum, "Theory of Nb-Zr Alloy Superconductivity and First Experimental Demonstration for Superconducting Radio-Frequency Cavity Applications," Aug. 2022, doi: 10.48550/arXiv.2208.10678. [Online]. Available: http://arxiv.org/abs/2208.10678

J. Lim, S. Sinha, A. C. Hire, J. S. Kim, P. M. Dee, R. S. Kumar, D. Popov, R. J. Hemley, R. G. Hennig, P. J. Hirschfeld, G. R. Stewart, and J. J. Hamlin, "Suppression of superconductivity in Nb-substituted MoB2 at high pressure." arXiv, Feb. 27, 2023 [Online]. Available: http://arxiv.org/abs/2302.13936

Talks

J. B. Gibson, A. Hire and R. G. Hennig, Data-Augmentation for Graph Neural Network Learning of the Relaxed Energy of Unrelaxed Structures, MRS Spring Meeting, Honolulu, HI, May 8-13, 2022 (contributed talk).

R. G. Hennig, Ultra-Fast Interpretable Machine-Learning Potentials for Metals and Semiconductors, MRS Spring Meeting, Honolulu, HI, May 8-13, 2022 (contributed talk).

A. C. Hire, S. Sinha, J. Lim, J. S. Kim, P. M. Dee, L. Fanfarillo, J. J. Hamlin, R. G. Hennig, P. J. Hirschfeld, G. R. Stewart, Ambient pressure stabilization of high-pressure superconducting phase of MoB2 by Nb doping, APS March Meeting, Las Vegas, NV, March 5-10, 2023 (contributed talk).

R. G. Hennig, S. R. Xie, P. Prakash, A. C. Hire, R. Schmid, H. Kraß, M. Rupp, Ultra-fast Interpretable Machine-learning Accelerated Structure Prediction of Materials, TMS Meeting, San Diego, CA, March 19-23, 2023 (invited talk).

Posters

A. Hire, R. G. Hennig, Surface Phase Diagram of Nb3Sn(100) surface. Sanibel Symposium, St. Augustine, FL, February 12-17, 2023 (Poster).

A. Hire, J. Lim, Y. Quan, J. S. Kim, S. R. Xie, R. S. Kumar, D. Popov, C. Park, R. J. Hemley, J. J. Hamlin, R. G. Hennig, P. J. Hirschfeld, and G. R. Stewart, Superconductivity Induced in WB2 by the Formation of Metastable Planar Defects, MRS Spring Meeting, Honolulu, HI, May 8-13, 2022 (contributed poster).

PROJECT UPDATE: Thermodynamics and Superconducting Properties of Novel SRF Superconductors

Hennig students: Ajinkya Hire (graduate student), Sam Dong (graduate student)

Ab-initio and AI-driven predictions of surface reconstructions, phase diagrams, and defects.
Optimizing the growth conditions of Nb_3Sn on Nb-SRF cavities is essential for improving the performance of SRF cavities. Surface reconstructions, defects, and adsorbed impurities play a crucial role in determining the superconducting properties of the surface. Towards this end, we have been studying the atomic scale surface reconstructions of Nb_3Sn(100) surface as a function of chemical potentials and experimental conditions using density functional theory (DFT) calculations and genetic algorithm structure search. From about 4500 surface structures generated by the genetic algorithm package GASP, we constructed the surface phase diagram of the Nb_3Sn(100) surface that shows surface reconstructions due to excess Nb and Sn.
From our surface phase diagram, we discovered that with increasing partial vapor pressure of Sn (equivalently at low deposition temperatures)-

1. The thickness of the reconstructed surface layer increases.

2. The density of Sn_{Nb} antisite defects, which are detrimental to superconductivity, formed on the $Nb_3Sn(100)$ surface also increases.

We also calculated Sn_{Nb} antisite defect formation energies as a function of depth for a clean $Nb_3Sn(100)$ surface. These calculations showed that Sn_{Nb} defects are stable on the surface compared to the bulk (Figure 1(a)). From our density of states (DOS) calculations, we found that for clean $Nb_3Sn(100)$, near the surface, the DOS at the Fermi level is suppressed and achieves its bulk value at a depth of 16Å, comparable to the coherence length of 40Å of bulk Nb_3Sn (Figure 1b). The trends we uncovered will provide valuable insights for optimizing the Nb_3Sn deposition recipes and for understanding the performance of the deposited Nb_3Sn layer.

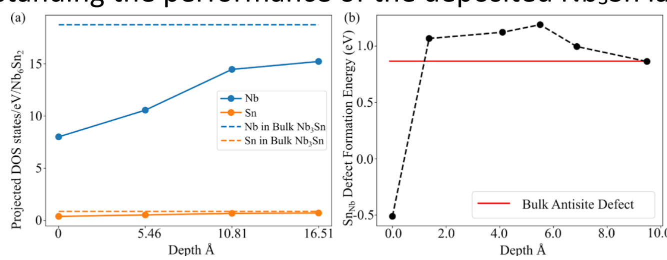


Figure 1(a) Atom projected DOS as a function of depth from the surface. Nb projected DOS is significantly suppressed near the surface compared to the bulk. (b) Sn_{Nb} antisite defect formation energy as a function of depth. Sn_{Nb} antisite defect is much more stable at the surface.

We have started incorporating oxygen in the genetic algorithm structure searches to investigate its effect on surface reconstructions. As the phase space of possible surface structures is enormous, performing DFT calculations is not feasible, especially for large surface reconstructions. Moreover, the temperature can cause a change in the arrangement of atoms on the surface. To expand our surface structure search to incorporate large unit cells and explore the effect of temperature, we are developing machine learning (ML) based interatomic potentials, which provide DFT-level accuracy at a fraction of the cost. So far, we have developed a UF3 ML model for the Nb-Sn system. Figure 2 shows the performance of the ML model on the testing dataset.

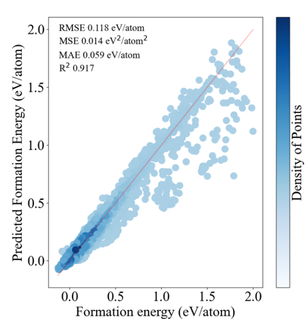


Figure 2: ML predicted vs. DFT formation energy calculated on the testing dataset.

To test the extrapolative ability of the ML model, we calculated the melting point of bulk Nb_3Sn. Our estimated melting point of 2140K for Nb_3Sn is much closer to the experimental value of 2400K than the value estimated using currently available interatomic potential in the literature.

We also implemented the UF3 framework in the LAMMPS molecular dynamic package. These implementation efforts enable massive dynamic simulation studies of the growth process of Nb_3Sn, diffusion of Nb and Sn in various phases, grain boundary, and surface structure evolution as a function of temperature. Further work on improving the current model and generating ML models for the Nb-Sn-O system is ongoing.

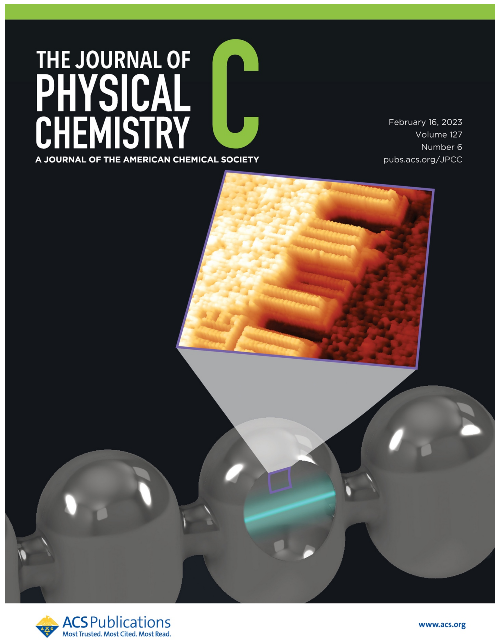
Validation of computational predictions through experimental collaboration.

To validate our computational methods and provide insights into the surface structures and kinetics, we collaborate with the Sibener group. We have investigated the NbO(3x1) ladder structure to shed light on the Sn adsorption on the substrate for growing Nb_3Sn. Using DFT calculations, we have identified multiple sites on the NbO(3x1) ladder structure for Sn adsorption. Sibener group's experimental results, supported by our theoretical calculations, contributed to a better understanding of Sn adsorption on Nb substrate.

In will continue our collaboration. The ML models being developed for the Nb-Sn-O system will be used to study the nucleation of Sn on the oxide surface.

Search for SRF superconductors beyond Nb, Zr-doped Nb, and Nb_3Sn.

We are also working on finding novel materials that might have the potential for higher efficiency and/or higher fields above Nb_3Sn. For this objective, we have been studying a new series of superconductors- $Nb_{(1-x)}Mo_xB_2$. MoB_2 undergoes a phase transition under pressure and becomes a superconductor with a T_c of 32K at 60GPa. We sort out to stabilize this high-pressure superconducting phase of MoB_2 at ambient pressure. Our DFT calculations predicted that adding Nb could stabilize MoB_2 in the correct structural phase at ambient pressure. With the help of our experimental collaborators at the University of Florida, $Nb_{(1-x)}Mo_xB_2$ samples were synthesized, and X-ray diffraction studies were performed. We discovered that as little as 10% Nb substitution ($Nb_{0.1}Mo_{0.9}B_2$) successfully stabilizes the high-pressure phase of MoB_2 at ambient pressure. Among our samples, $Nb_{0.25}Mo_{0.75}B_2$ had the highest T_c of 8K, with an interestingly high upper critical field (H_{c2}) of 6.71T. For reference, H_{c2} of Nb is 0.28T with a similar T_c of 9K. Further work is needed to estimate the suitability of $Nb_{0.25}Mo_{0.75}B_2$ for SRF cavity application.



PROJECT UPDATE: Thermodynamics and Superconducting Properties of Novel SRF Superconductors

Hennig students: Sam Dong (graduate student), Jason Gibson (graduate student)

Jason Gibson is working with Sam Dong on completing a manuscript on the structure prediction for the Cs-Sb surface phase diagram. Jason started last summer working as an intern at Los Alamos National Lab on machine learning methods. He continued after the summer and is still working at Los Alamos as an intern. The machine learning methods he is developing will impact his CBB work as they provide the opportunity to accelerate the structure predictions for surface phase diagrams. He has continued working on the side on the CBB project without being directly funded.

Sam Dong is working on implementing the machine learning methods into our structure prediction methods and will take over the project from Jason Gibson.

Hines, Melissa (Cornell)

Publications

Talks

1. Telluride Summer Res. Conf. on Semiconductor Surface Chem. (Telluride CO) (Hines invited talk)

Collaborating Group

1. Cornell University: Jared Maxson

PROJECT UPDATE: Air-stable, high-performance photocathodes (PHC)

Hines students and postdocs: Annabel Selino (graduate student), Mariam Hasany (graduate student), Amy Zhu (graduate student), Azriel Finsterer (graduate student)

The ultimate goal of this project is to develop a protocol for the production of air-stable, atomically flat, atomically clean, high QE epitaxial photocathodes via "upside down" growth on 2D-material-coated substrates. The successful realization of this requires four things: (i) deposition of clean single-layer graphene (SLGr) or hexagonal boron nitride (hBN) on a growth substrate, (ii) growth of the photocathode on the 2D material, (iii) successful capping of the photocathode with an inert material, and (iv) successful delamination of the protected photocathode from the growth substrate. Although we have been able to produce protected photocathodes in the Maxson MBE chamber, these photocathodes suffered from two problems. First, the deposited material is CsSb, not Cs_3Sb. Second, the photocathodes are lightly oxidized in the region near the 2D material (i.e., the region of initial growth) even though the photocathodes have good QE when measured from the backside (i.e., the region of final growth. Both issues point to a problem with film nucleation.

Early Cs_3Sb photocathodes were produced with an *atomic* Sb source, not a Sb_4 source such as that used in the Maxson lab. Sommer showed that the two sources are not interchangeable: the atomic source produces Sb films that are chemically more reactive than the cluster source. To produce atomic Sb, I synthesized a PtSb bead, which was installed in the Maxson MBE chamber. We found that the photocathodes had 3× higher QE than those synthesized with a standard Sb source (*i.e.,* 6% vs 2% QE); however, this did not solve our nucleation problem.

Last summer we realized that we needed to study the nucleation of the photocathode *in situ,* which is not possible in the Maxson chamber. To fix this issue, we purchased a three cell evaporation source — a “mini MBE” — which was installed on the Hines surface analysis chamber in January. Crucibles for the source arrived in February. We have grown atomically clean Cr films with the new source. We are in the process of synthesizing new CsIn and PtSb sources that are compatible with the mini-source. Once synthesized, we will begin to study film nucleation using both STM and XPS analysis.

Our progress has been slowed by two factors. First, Mariam Hasany was unexpectedly detained in Pakistan for the entire Fall semester. As a result, Annabel Selino had to cover Mariam's TA duties unexpectedly, which slowed her research. Second, the chemistry department laid off the glass blower, so I am having to learn to blow quartz for the PtSb synthesis, as Pt and Sb metal must be sealed in quartz under vacuum and then heated to ~1100°C. On the bright side, a new Hispanic graduate student, Azriel Finsterer, has joined the project. Annabel, Mariam, and Azriel will be replacing Amy, who is due to graduate in ~1 year.

Fig. 1: ~5 mm diameter PtSb bead synthesized in a quartz boat prepared by a professional glass blower. The dark deposit to the right of the bead is an Sb film from the cool-down process.

Hoffstaetter, Georg (Cornell)

Not available.

Karkare, Siddharth (Arizona State University)

Publications

Papers

P. Saha, O. Chubenko, J. Kevin Nangoi, T. Arias, E. Montgomery, S. Poddar, H. A. Padmore, and S. Karkare, "Theory of photoemission from cathodes with disordered surfaces," *Journal of Applied Physics*, vol. 133, no. 5, p. 053102, Feb. 2023, doi: 10.1063/5.0135629.

C. J. Knill, H. Yamaguchi, K. Kawahara, G. Wang, E. Batista, P. Yang, H. Ago, N. Moody, and S. Karkare, "Near-Threshold Photoemission from Graphene-Coated Cu(110)," *Phys. Rev. Appl.*, vol. 19, no. 1, p. 014015, Jan. 2023, doi: 10.1103/PhysRevApplied.19.014015.

A. Kachwala, O. Chubenko, D. Kim, E. I. Simakov, and S. Karkare, "Quantum efficiency, photoemission energy spectra, and mean transverse energy of ultrananocrystalline diamond photocathode," *Journal of Applied Physics*, vol. 132, no. 22, p. 224901, Dec. 2022, doi: 10.1063/5.0130114.

Posters and Talks

ORAL CONTRIBUTION P. Saha, S. Karkare#, E. Echeverria, A. Galdi, J.M. Maxson, C.A. Pennington, E.J. Montgomery, S. Poddar, Epitaxial Alkali-Antimonide Photocathodes on Lattice-matched Substrates, North American Particle Accelerator Conference, Albuquerque, NM (2022) TUYD5

ORAL CONTRIBUTION G.S. Gevorkyan, T.J. Hanks, A.H. Kachwala, C.J. Knill, C.A. Sarabia Cardenas S. Karkare#, Commissioning of the ASU Cryocooled 200 kV DC Electron Gun, North American Particle Accelerator Conference, Albuquerque, NM (2022) TUYD6

C.J Knill, S.S. Karkare#, H. Ago, K. Kawahara, E. Batista, N.A. Moody, G.X. Wang, H. Yamaguchi, P. Yang Near-Threshold Photoemission from Graphene-Coated Cu Single Crystals, North American Particle Accelerator Conference, Albuquerque, NM (2022) WEPA66

A.H. Kachwala, O. Chubenko, S.S. Karkare, R. Ahsan, H.U. Chae, R. Kapadia, On-Chip Photonics Integrated Photocathodes, North American Particle Accelerator Conference, Albuquerque, NM (2022) WEPA65

J.I. Mann, G.E. Lawler, J.B. Rosenzweig, B. Wang, S.S. Karkare, T. Arias, J.K. Nangoi Simulations of Nanoblade Cathode Emissions With Image Charge Trapping for Yield and Brightness Analyses, North American Particle Accelerator Conference, Albuquerque, NM (2022) TUPA86

C.M. Pierce, I.V. Bazarov, J.M. Maxson,S. Cabrini, D.B. Durham, D. Filippetto, A. Kachwala, S.S. Karkare, F. Riminucci, A. Minor, Towards High Brightness from Plasmon-Enhanced Photoemitters, North American Particle Accelerator Conference, Albuquerque, NM (2022) TUYD4

J.W. Lewellen#, J. Smedley*, T. Vecchione, D. Filippetto, S.S. Karkare, J.M. Maxson, P. Musumeci The Quest for the Perfect Cathode, Albuquerque, NM (2022) TUYD3

Collaborating Groups

1. Cornell University: Tomás Arias, Ivan Bazarov, Jared Maxson

PROJECT UPDATE: Optical, X-ray and surface characterization of Alkali-antimonides (PHC)

Karkare students: Pallavi Saha (graduate student)

Alkali-antimonides are excellent candidates for photocathodes, namely for their high quantum efficiency (QE) and low mean transverse energy (MTE), in the visible light. Existing studies have mostly focused on QE and emittance, however the experimentally measured values of MTE from Cs-Sb cathodes did not go down to the thermal limit when operated near photoemission threshold, at both room temperature and cryogenic temperatures. The thermal limit to MTE was not achieved when the cathodes were operated near photoemission threshold. The discrepancy between experimental and theoretical values was attributed to various factors like surface non-uniformities and bulk defects/impurity states in the band gap, which were thought to limit the electron beam brightness from such cathodes.

We formulated a theoretical model to explain photoemission from cathodes with disordered surfaces like alkali-antimonides and derived expressions for QE and MTE. The only theoretical model which exists in literature is Dowell and Schmerge's photoemission model which is based on some assumptions like conservation of transverse momentum and flat distribution of density of states, which do not hold true for polycrystalline materials like Cs-Sb cathodes. We modified Dowell-Schmerge approach to develop a model under the assumption that for a given energy, electrons get uniformly emitted in the transverse momentum space. We used the joint density of states coupled with Spicer's 3-step photoemission model. The band structure of Cs3Sb calculated by Kevin J. Nangoi (Arias group) was used and defect states was incorporated by adding an exponential tail of width 0.04eV to the valence band density of states. Our simplistic theoretical model predicts QE and MTE values which are in excellent agreement with experimentally measured values from Cs3Sb cathodes, both at room temperature and cryogenic temperatures. The near-threshold QE and MTE performance is largely explained by the exponentially decaying defect density of states near the valence band maximum. Our results have got published in J. Appl. Phys. as "Theory of Photoemission from Photocathodes with Disordered Surfaces"

We performed a detailed first time measurement of the MTE and kinetic energy distribution spectra from cesium-antimonide photocathodes grown on different substrates Si and STO, using the Photoemission Electron Microscope (PEEM). The energy distribution spectra indicates that the photoemission threshold of these materials lies around 1.55 eV (shown in Fig. 1) as opposed to the previous thought value of 1.8 eV. The occurrence of threshold at 1.55 eV is further corroborated by the MTE data which shows that the MTE approaches the thermal limit of k_BT as photon energy is equal to the threshold (Fig. 2). However, the QE at threshold is 10^{-7}, which suggests that it may not be feasible to operate these cathodes near threshold.

Pallavi also participated in the x-ray/RHEED studies of the Cs3Sb cathodes using the NSLS-2 synchrotron light source in collaborations with John Smedley (SLAC), Mengja Gaowei (BNL) and Jared Maxson (CBB-Cornell).

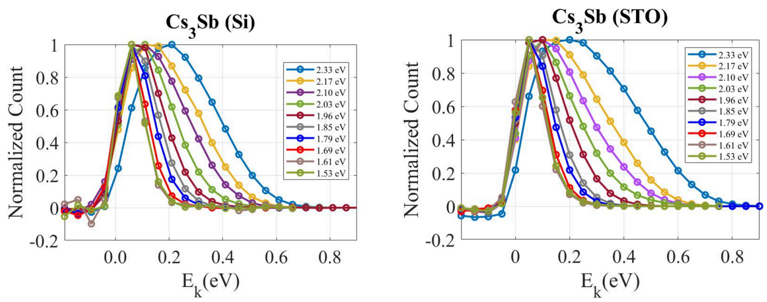


Fig. 1: Energy distribution spectra from Cs3Sb cathodes grown on Si and STO.

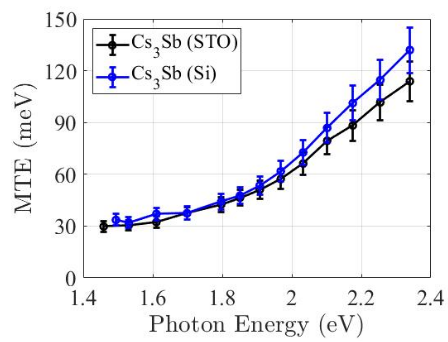


Fig. 2. Spectral response of MTE measured from Cs3Sb cathodes grown on Si and STO substrates. The MTE approaches k_BT as photon energy equal to the photoemission threshold ~1.55 eV. At higher photon energies, MTE does not scale as one-third of the excess energy owing to the scattering losses, which the electrons suffer with the phonons in the conduction band before emission.

PROJECT UPDATE: Measurements of low energy electron distributions (PHC)

Karkare students: Chris Knill (graduate student)

In this project Chris Knill performed a detailed study of photoemission from Cu(110) in terms for its QE, MTE and non-linear photoemission effects. It was found that graphene coated Cu(110) deliver thermally limited MTEs, however, the low QE would increase the MTE to several 100 meV for high charge densities due to non-linear photoemission effects.

First, we collaborated with several non-CBB members at Los Alamos National Laboratory and Kyushu University in Japan in order to investigate the use of a graphene protective layer to improve the stability of a photocathode. In particular we performed photoemission measurements on a graphene-coated Cu(110) single crystal at room and cryogenic temperatures in order to investigate the impact that a graphene layer has on the MTE. We found that aside from slightly shifting the work function of the photocathode, the graphene layer had no effect on the photoemission process. We were able to measure sub-10 meVs at the cryogenic temperature of 77 K and we were able to measure the thermal limit of 25 meV at room temperature (300 K). The thermal limit is the minimum achievable MTE from these photocathodes at a given temperature. The results are shown in the Fig. 1 below. We also performed MTE measurements over the course of a week and found that we could achieve MTEs right near the thermal limit for the duration of the time period. Lastly, we found that a graphene protective layer significantly reduces the surface preparation that is typically needed in order for single crystalline photocathodes to reach the thermal limit (Phys. Rev. Applied 19, 014015 (2023)).

Second, we worked on was the problem of low QE from the record low MTE Cu photocathode. For such low QEs, a very high laser fluence must be used to extract the necessary charge for high charge density applications like XFELs and single-shot UED/UEM. At these high fluences the nonlinear photoemission effects of electron heating and multi-photon emission begin to dominate the photoemission process and the small MTEs that made these photocathodes so promising are no longer attainable. We performed detailed measurements of these nonlinear effects at three different pulse lengths for both above and below threshold photoemission in order to gain a detailed understanding of nonlinear photoemission. Using our measured data with a QE of 10^(-9) we developed an extrapolation technique to determine the impact that nonlinear effects have on the MTE at high charge densities for a wide range of photocathode QEs. Our results (shown in the Fig. 2 below) indicate that in order to mitigate non-linear photoemission effects, photocathodes that have QEs on the order of 10^(-4) must be used.

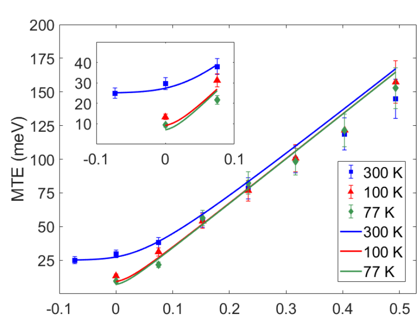


Fig. 1: Plot showing the MTE as a function of the excess energy (difference between photon energy and work function) for room and cryogenic temperatures. The points represent our measured data while the lines represent a theoretical model that we compared our results

with. Looking at the inset in the top left corner we see that for 77 K we measured sub-10 meV MTEs and that for 300 K we measured 25 meV. This shows that the graphene protective layer does not impact the MTE.

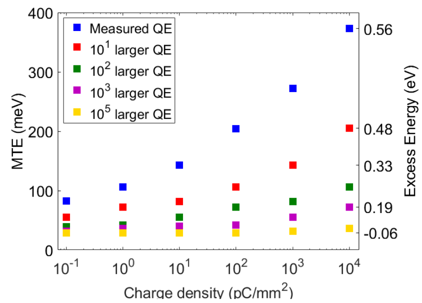


Fig. 2: Plot showing the minimum MTE for a 130-fs pulse length that can be measured for high charge densities and the excess energies where the minimum occurs. This plot shows that in order to mitigate nonlinear photoemission effects, the QE of the photocathode must be 5 orders of magnitude larger than our measured QE of 10^{-9}. This puts the necessary QE at 10^{-4} which points to the use of semiconductor photocathodes like Alkali-antimonides.

PROJECT UPDATE: Study of Nano-scale photoemitters using PEEM

Karkare student and theme postdoc: Ali Kachwala (graduate student)

We studied nitrogen incorporated Ultrananocrystalline diamond (UNCD) film using the PEEM. Recently, UNCDs have been considered for high peak current applications because of its mechanical, thermal, and electrical stability. However, the electron emission mechanism from (N)UNCDs is not very well understood. In one of my work, I have studied the QE, photoemission electron energy spectra and MTE of (N)UNCD photocathode and have published the same in the Journal of Applied Physics (J. Appl. Phys. 132, 224901 (2022)). This work was done in collaboration with Dr. Evgenya Simakov at LANL.

We also investigated plasmonic spiral structures and work function patterned structures in collaboration with Daniele Filippetto (exLBNL – CBB affiliate) and Prof. Jared Maxson (CBB-Cornell). The photoemission results from the plasmonic spirals indicate nanoscale hot spots due to fabrication defects. The work function patterned samples showed no emission from work function areas, likely due to contamination during the fabrication process. Our results show that the nanofabrication processes for both need to be improved obtain the desired performance.

PROJECT UPDATE: Nano-scale photoemitters via light focusing nano-structures

Karkare students: Mansoure Moeini Rizi (postdoc)

Little progress was made in this project as most of the year was spent in a post-doc search. Mansoure Moeini Rizi was hired as started working on this project in mid-February. She has been developing a new way to fabricate plasmonic spiral structures that will minimize the defects that produce localized emission hot spots using e-beam lithography and lift-off techniques. The first spirals have been successfully fabricated using this process and are awaiting testing in the PEEM.

PROJECT UPDATE: Development of ASU DC cryogun and beam diagnostics

Karkare students: Mansoure Moeini Rizi (postdoc)

In this project, we have commissioned the ASU DC cryogun to 34 K temperature and 200 kV fields. We obtained the first beam and developed the beamline to measure the 4-D phase space (figure 1). The motivation for this project is to combine the tools and resources of surface science with that of high voltage accelerators, and to demonstrate the use of novel photocathodes which were not previously possible due to the design of modern photoinjectors.

We have also performed simulations of electron-electron repulsion from nanoscale (sub-100 nm sized) photoemitters used in the DC beam mode for electron microscopy applications. Such photoemitters show potential to beat the brightness of field emission tips even in the DC beam regime. Our simulations show that the electron-electron repulsion will limit the beam current from such sources will be limited to around 1 um. They also show that there will not be any significant effect on the MTE down to the single meV scale and a minimal effect on the longitudinal spread on the 10 meV scale.

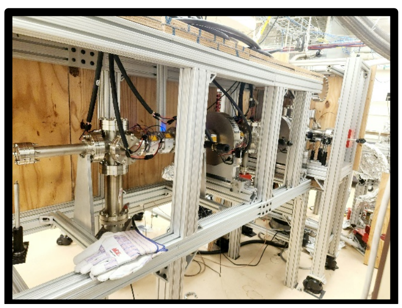


Fig 1: Commissioned beamline connected to the ASU cryocooled 200 kV DC electron gun. The vacuum system is at a steady 10^-10 torr and connected to the entire transfer line meaning there is direct vacuum access to our photocathode diagnostic instruments and growth chamber. We are developing a LabVIEW to automate our beamline magnets and cameras, outputting a full 4D phasespace measurement of the photoemitted beam.

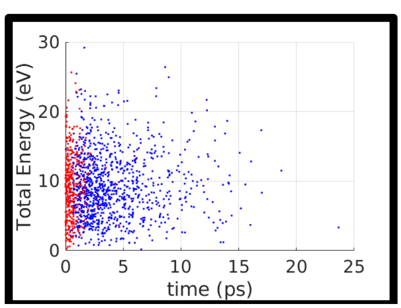


Fig 2: Plot of total energy of emitted electrons verses emission interval time. Blue electrons escape from the cathode and are counted toward the output current, and red electrons fall back into the surface of the photocathode. There is a distinct cutoff interval suggesting a current saturation limit.

Kim, Young-Kee

Not available.

Liepe, Matthias (Cornell)

Not available.

Maxson, Jared (Cornell)

Publications

Papers

M. Gordon, W. H. Li, M. B. Andorf, A. C. Bartnik, C. J. R. Duncan, M. Kaemingk, C. A. Pennington, I. V. Bazarov, Y.-K. Kim, and J. M. Maxson, "Four-dimensional emittance measurements of ultrafast electron diffraction optics corrected up to sextupole order," *Phys. Rev. Accel. Beams*, vol. 25, no. 8, p. 084001, Aug. 2022, doi: 10.1103/PhysRevAccelBeams.25.084001.

J. K. Bae, M. Andorf, A. Bartnik, A. Galdi, L. Cultrera, J. Maxson, and I. Bazarov, "Operation of Cs-Sb-O activated GaAs in a high voltage DC electron gun at high average current," May 2022, doi: 10.48550/arXiv.2205.13632. [Online]. Available: http://arxiv.org/abs/2205.13632

C. M. Pierce, D. B. Durham, F. Riminucci, S. Dhuey, I. Bazarov, J. Maxson, A. M. Minor, and D. Filippetto, "Experimental Characterization of Photoemission from Plasmonic Nanogroove Arrays," Phys. Rev. Appl., vol. 19, no. 3, p. 034034, Mar. 2023, doi: 10.1103/PhysRevApplied.19.034034.

M. Andorf, J. K. Bae, A. Bartnik, L. Cultrera, J. Maxson, and I. Bazarov, "HERACLES: A high-voltage DC test beamline for high average current photocathodes," Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, vol. 1052, p. 168240, Jul. 2023, doi: 10.1016/j.nima.2023.168240.

Collaborating Groups

1. University of California at Los Angeles: Pietro Musumeci, James Rosenzweig
2. Cornell University: Tomás Arias, Ivan Bazarov, Melissa Hines, Kyle Shen
3. Northern Illinois University: Philippe Piot
4. Arizona State University: Siddharth Karkare

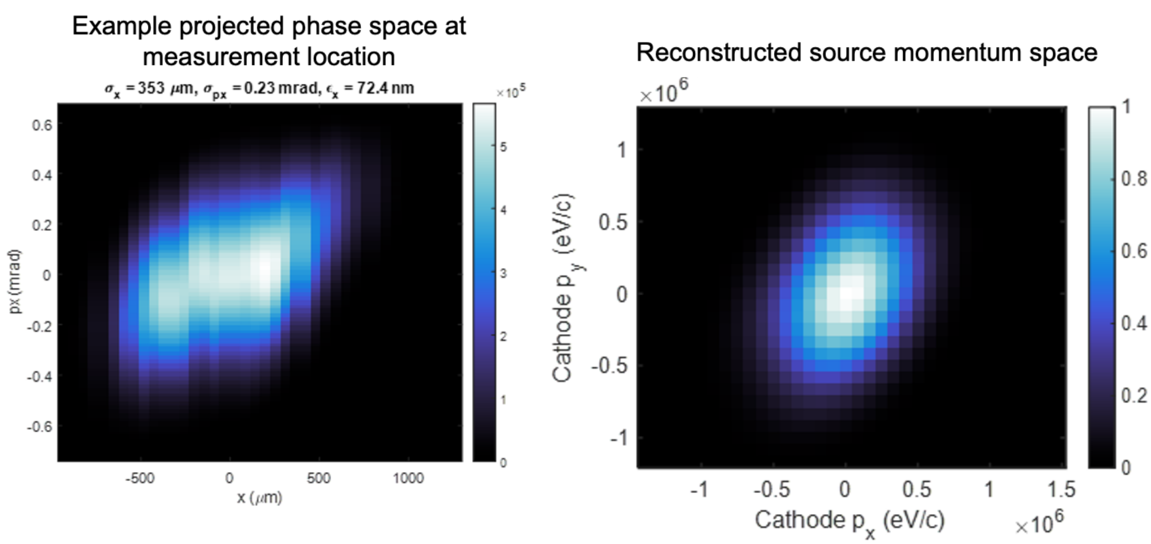
PROJECT UPDATE: MTE measurements on CBB cathodes: studying temperature, wavelength, crystallinity, and nonlinearity

Student: Charles Zhang (graduate student)

Charles is the primary graduate student assigned to the cryo-MTE meter at Cornell. This system directly interfaces with the Cornell photocathode MBE system, which is the only system in the world growing single crystal alkali antimonides. Charles is a second-year student who has just begun in my group this year. Charles has already made significant advances for our measurement of photocathode MTE.

Achievement: Charles developed an analytic method to reconstruct photocathode emission momenta without any a-priori knowledge of the laser distribution or the electron optics between source and detector. The latter is often a practical limitation in accelerators: the precise electron optics between source and detector are often challenging to determine with the precision needed for low MTE measurement. The former is a fundamental limitation in cases where the photoemission is partially nonlinear: in this case the exact spatial probability density distribution of the emitted electrons depends on the microscopic details of both the photocathode and laser distribution. Beyond reconstruction of the entire momentum distribution (not just the MTE); Charles' method also enables the reconstruction of the entire initial transverse phase space. This is an important step forward in our ability to reconstruct source information in downstream phase space measurements.

Charles is in the process of finalizing proof of principle data for this method. Example (preliminary) experimental data on the TE meter is shown below.

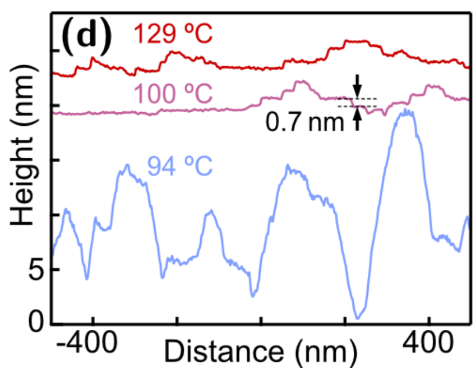


PROJECT UPDATE: Growth and Characterization of the Epitaxial Alkali Antimonides: Exploring growth parameters, MET, QE, and robustness (PHC) and Growth and characterization of high efficiency, low MTE photocathodes: epitaxy and performance high field

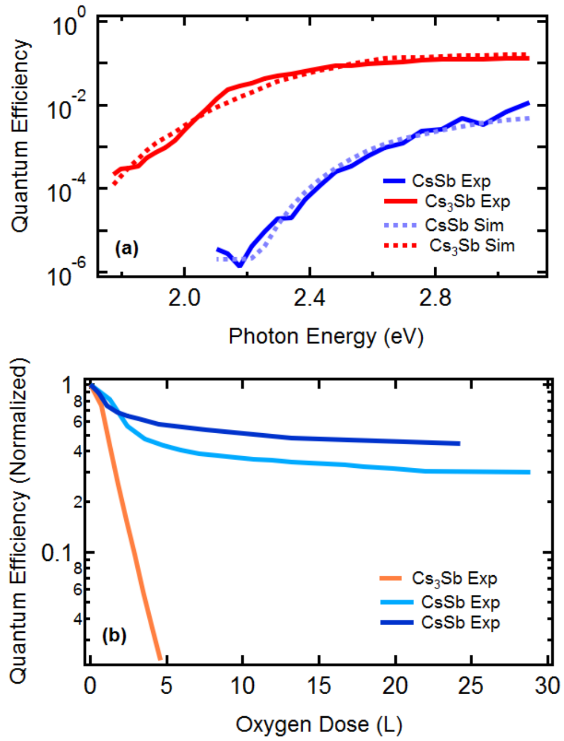
Maxson theme postdoc: Chad Pennington (Graduate Student), Elena Echeverria (theme postdoc)

I am grouping Chad Pennington and Elena Echeverria in one report section as many of their achievements have been made in close collaboration with each other. Chad and Elena are the primary photocathode growth scientists in my laboratory at Cornell. We have several achievements to report this year.

- Achievement: Demonstration of high quantum efficiency and dramatically reduced oxygen sensitivity (as compared to Cs_3Sb) in the Cs_1Sb photocathode system. A manuscript for publication on this "new" photocathode material is nearly complete.
    - In the optimal growth conditions, this material is near-atomically smooth. STM data from the Hines lab shown below:



    - It has high quantum efficiency in the blue region of the spectrum, and it is far more robust than Cs_3Sb in terms of oxygen exposure:



- Achievement: Our group participated in the first characterization of Na_3Bi photocathodes, which may support the generation of low MTE beams via the Dirac cone in its band structure. This project is led by the Shen group with lead graduate student Vivek Anil.

- We participated in a series of synchrotron beamtimes with BNL and SLAC wherein we characterized nearly-epitaxial photocathodes via x-ray fluorescence, reflectivity, and diffraction. We are aiming to fully transfer our know-how of epitaxial photocathode growth to national labs, starting with BNL and SLAC.

- We successfully grew Cs-K-Sb photocathodes in the Cornell MBE chamber and will soon explore the growth parameter space for epitaxy. Even these polycrystalline photocathodes display enhanced red response as compared to Cs_3Sb.

Muller, David (Cornell)

Publications

Papers

N. S. Sitaraman, Z. Sun, B. Francis, A. C. Hire, T. Oseroff, Z. Baraissov, T. A. Arias, R. Hennig, M. U. Liepe, D. A. Muller, and M. K. Transtrum, “Theory of Nb-Zr Alloy Superconductivity and First Experimental Demonstration for Superconducting Radio-Frequency Cavity Applications,” Aug. 2022, doi: 10.48550/arXiv.2208.10678. [Online]. Available: http://arxiv.org/abs/2208.10678

Z. Sun, Z. Baraissov, L. Shpani, R. D. Porter, Y.-T. Shao, T. Oseroff, M. O. Thompson, D. A. Muller, and M. U. Liepe, “Smooth, homogeneous, high-purity Nb3Sn RF superconducting films by seed-free electrochemical synthesis.” arXiv, Feb. 03, 2023 [Online]. Available: http://arxiv.org/abs/2302.02054

Posters and Talks

1. Desheng Ma, Chenyu Zhang, Yu-Tsun Shao, Zhaslan Baraissov, Cameron Duncan, Adi Hanuka, Auralee Edelen, Jared Maxson, and David Muller. "Physics-informed Bayesian Optimization of an Electron Microscope." Bulletin of the American Physical Society, March Meeting (2023).

2. C. Zhang, Yu-Tsun Shao, Z. Baraissov, C. Duncan, A. Hanuka, A. Edelen, J. Maxson, D. Muller, "Bayesian Optimization for Multi-dimensional Alignment: Tuning Aberration Correctors and Ptychographic Reconstructions", Microsc. Microanal. 28 (S1), 3146 (2022) Available: https://doi.org/10.1017/S1431927622011692

3. Desheng Ma, Chenyu Zhang, Yu-Tsun Shao, Zhaslan Baraissov, Cameron Duncan, Adi Hanuka, Auralee Edelen, Jared Maxson, and David Muller. “Machine Learning for Beam Emittance Measurement and Aberration Correction of an Electron Microscope”. 3rd ICFA Beam Dynamics Mini-Workshop on Machine Learning Applications for Particle Accelerators (Chicago, IL - Palmer House Hilton) – Nov. 1-4, 2022

4. Desheng Ma, Chenyu Zhang, Yu-Tsun Shao, Zhaslan Baraissov, Cameron Duncan, Adi Hanuka, Auralee Edelen, Jared Maxson, and David Muller. "Physics-informed Bayesian Optimization of an Electron Microscope." MIT nano workshop - Automation, AI and Machine Learning in Electron Microscopy: Current Trends and Future Developments - Nov. 7, 2022

5. Z Baraissov, Z Sun, YT Shao, N Sitaraman, T Arias, M Liepe, D Muller, “Nanoscale lattice and chemical composition measurements of superconducting Nb3Sn films using scanning transmission electron microscopy”, Bulletin of the American Physical Society, March Meeting (2023)

Licenses

1. Thermo Fisher Scientific – collaboration on high-brightness cold field emission gun (CFEG) technology. 2nd generation electron microscope pixel array detector licensed to Thermo Fisher Scientific. TFS funded Sol Gruner and I to develop a 2^{nd} generation detector, which is now completed. It which will take approximately 2 years to bring to market. (Licensing fees from the 1^{st} generation are now over $600k).

Awards and Honors

David Muller, American Association for the Advancement of Science
Zhaslan Baraissov, Microscopy Society of America

Collaborating Groups

1. Cornell University: Jared Maxson, Matthias Liepe
2. SLAC: Auralee Edelen
3.

PROJECT UPDATE: Electron Microscopy characterization of the microsctructure of materials for SRF cavities

Muller student: Zhaslan Baraissov (graduate student)

Goal: Characterize the impact of growth conditions on surface oxides and performance for Nb3Sn and Nb-Zr alloys.

In the search for improved SRF cavity, we have examined potential alternative materials to Nb, including NbZr and optimized processing of Nb3Sn.
NbZr was identified by CBB theorists as having improved superconducting properties, and based on these predictions, using his recently developed electrochemical synthesis method, Zeming Sun has successfully grown a layer of the ZrNb alloy on top of the bulk Nb. Thomas Oseroff then measured the resistance of these samples, and found the critical temperature to be between 13 to 16K, depending on the degree of Zr doping and annealing extent. We examined the structure and composition of these films at the nanoscale using Scanning Transmission Microscopy (STEM), Fig 1 shows the results of the energy dispersive x-ray analysis. Here, we find four principal elements present in this sample: Nb,Zr,C,O. In this system, there is ZrO2 the top of the surface, while bulk Nb crystal is used as the substrate for the deposition. Unexpectedly, at the interface between the two materials we observe a high concentration of carbon, that forms a thin layer right on top of the Nb and also diffuses into the underlying bulk Nb. Elemental analysis shows that the crystalline whiskers have Nb2C composition and the corresponding diffractive studies indicate an orthorhombic structure, that is non-superconducting. On the other hand, upon a closer investigation of the composite film, as shown in we have found that the layer is polycrystalline. The diffraction pattern from the film area represents a set of concentric rings that can indicate the type of crystal symmetry in the material. From the least-square fit shown in Fig 2C, one can conclude that the film has NbC composition and Fm-3m space group (rock-salt structure). As this type of NbC crystal is known

to possess superconducting properties with critical temperature of 12K, we can thus propose that the niobium carbide plays a significant role in the excellent superconducting performance of the annealed material.

We have also investigated the structural and morphological changes in niobium oxides, as a function of their treatment history. For this project we have used energy dispersive x-ray analysis for mapping elemental distributions (Figure 2) and Electron Energy Loss Spectroscopy (EELS) for oxygen bonding information. The signal from the anodized sample exhibits increasing order and oxidation state. What is remarkable is that the oxide in anodized sample is layered where the top layer is less oxidized than the bottom layer. With further oxidation the first peak becomes narrower and more elevated, indicating that the oxide structure reached maximum oxidation level -niobium pentoxide. We find a striking similarity between the bottom layer anodized Nb oxide and the oxide in Nb3Sn, suggesting that the structure in both cases is mostly Nb2O5. Using this method, we can investigate the changes in oxide structure with different thermal and chemical treatment.

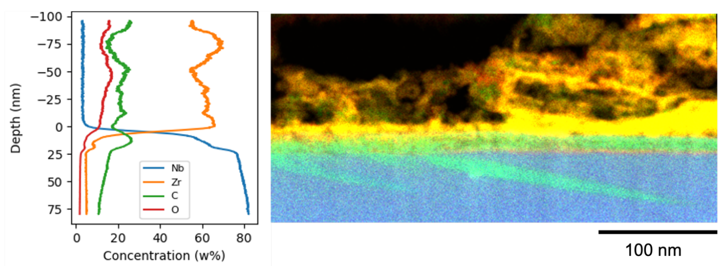


Figure 1. NbC precipitates in Superconducting NbZr. Left: Concentration depth profile of each of the principal elements found in the sample. Right: Composite EDX map, obtained by combining elemental maps in different color channels. Colors are assigned the same way as on the left figure.

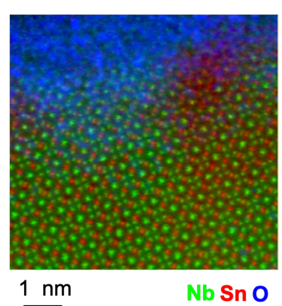


Figure 2. Atomic-Resolution map of oxidized Nb3Sn from energy-dispersive x-ray spectroscopy in a scanning transmission electron microscope, showing the oxide is predominantly Niobium oxide with very little tin remaining in the oxide.

PROJECT UPDATE: Deliverable 3.2 Electron Microscope Aberration Correction by Machine Learning

Muller student: Desheng Ma, (graduate student)

Aberration-corrected scanning transmission electron microscopes (STEM) use a series of multipole magnets to generate a sub-Ångstrom-sized electron beam for atomic resolution imaging and chemical composition. The alignment of the aberration corrector requires specialist and specific training, and it often takes hours to finish. In this work, we aim to automate tuning the aberration corrector using an optimizer called Bayesian optimization (BO) to align the probe by minimizing the beam emittance.

There are three parts to the project:

a. Theoretical proof of the equivalence between aberration correction and phase space beam emittance minimization. We have shown that the wave function of electrons characterized by their aberration function can be translated to beam emittance via Wigner-Weyl transform. This lays the foundation of our optimization scheme by providing a single valued metric as the response of the black-box function.

b. Design of a convolutional neural network to predict beam emittance from electron Ronchigrams. This has been demonstrated by the previous postdoc (CBB alumnus) Chenyu Zhang. As continuation, we have tested several new architectures, including ResNet and DenseNet with improved performance.

c. Design of Bayesian optimization (BO) scheme to execute the optimization. We apply BO to efficiently optimize the emittance by modelling the full posterior of the response over control parameters to account for uncertainties at each query. We have demonstrated the method with a simulation by the General Particle Tracer (GPT) model with the help of Cameron Duncan (CBB alumnus). We have now succefully implemented the algorithm on three different electron microscopes, a Nion UltraSTEM (Chenyu Zhang), a ThermoFisher Spectra 300 S/TEM and ThermoFisher Titan Cryo-S/TEM.

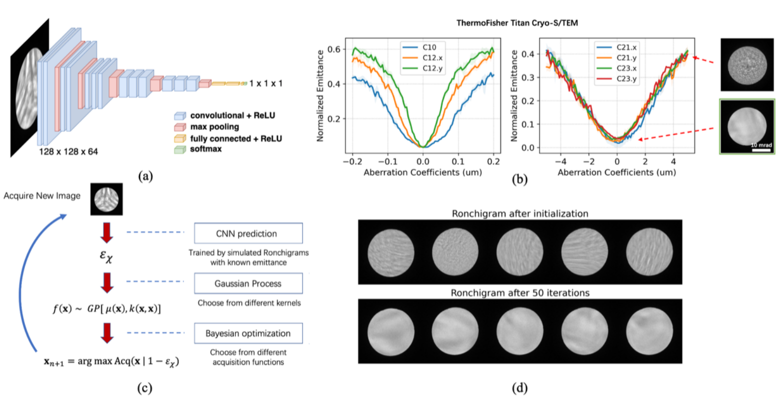


Figure 1. Bayesian optimization of a real microscope: ThermoFisher Titan Cryo-S/TEM. (a). VGG-16 architecture to map Ronchigrams to beam emittance. (b). CNN predicted emittance values from Ronchigrams at varying aberrations of first order and second order, averaged from five individual scans. (c). Workflow of the online Bayesian optimization of an electron microscope. (d). Ronchigrams before and after optimization with 50 iterations.

Musumeci, Pietro (UCLA)

Publications

Papers

F. Cropp, L. Moos, A. Scheinker, A. Gilardi, D. Wang, S. Paiagua, C. Serrano, P. Musumeci, and D. Filippetto, "Virtual-Diagnostic-Based Time Stamping for Ultrafast Electron Diffraction." arXiv, Feb. 09, 2023 [Online]. Available: http://arxiv.org/abs/2302.04916

C. V. Frederick (Eric), D. Filippetto, A. Gilardi, P. Musumeci, S. Paiagua, A. Scheinker, and D. Wang, "Toward Machine Learning-Based Adaptive Control and Global Feedback for Compact Accelerators," in *Proceedings of the 13th International Particle Accelerator Conference*, 2022, vol. IPAC2022, doi: 10.18429/JACOW-IPAC2022-TUPOST055 [Online]. Available: https://jacow.org/ipac2022/doi/JACoW-IPAC2022-TUPOST055.html

Talks

Contributed oral presentation, A. Scheinker, D. Filippetto, and F. Cropp, "6D Phase space diagnostics based on adaptively tuned physics-informed generative convolutional neural networks." IPAC 2022, Bangkok, Thailand.

Posters

Poster presentation, F. Cropp, L. Moos, A. Scheinker, A. Gilardi, D. Wang, S. Paiagua, C. Serrano, P. Musumeci and D. Filippetto, "Virtual-Diagnostic-Based Time Stamping for Ultrafast Electron Diffraction." 3rd ICFA Beam Dynamics Mini-Workshop on Machine Learning Applications for Particle Accelerators, Chicago, Illinois, USA, 2022

Poster presentation, F. Cropp, A. Scheinker, A. Gilardi, S. Paiagua, D. Wang, P. Musumeci, D. Filippetto, "Toward Machine Learning-Based Adaptive Control and Global Feedback for Compact Accelerators," IPAC 2022, Bangkok, Thailand.

Poster presentation, C. Pennington, A. Ody, P. Denham, F. Cropp, Y. Park, A. Galdi, L. Cultrera, J. Maxson, P. Musumeci, "Alkali Antimonide Photocathodes at High Gradients." IPAC 2022, Bangkok, Thailand.

Collaborating Groups

1. Cornell University: Jared Maxson
2. Northern Illinois University: Philippe Piot
3. SLAC: Auralee Edelen

PROJECT UPDATE: Advanced photocathodes testing in high gradient RF gun at the UCLA Pegasus Laboratory

Musumeci students: Veronica Guo (undergraduate), A. Kulkarni (undergraduate student), David Garcia (graduate student)

This project has the ambitious goal to carry out the first tests of alkali antimonide cathodes in high gradient RF gun enviroments in the US. The cathodes are grown at Cornell and then transported to UCLA. The first successful transfers of the cathode have attracted the interest of the broader accelerator community and

A new student D. Garcia is developing schemes to shape the photocathode drive laser and optimize the beam brightness by controlling the laser spectrum and its spatio-temporal profiles. Part of this activity, in particular the adoption of four-wave mixing as the non linear optical process to generate the final photon energy, is carried out in collaboration with UCLA EE Assistant Professor S. Carbajo.

This topic is now strongly linked to the optimization of the beam dynamics in an high gradient photoinjector with low MTE cathodes. In collaboration with the Cornell group we will be working on demonstrating 100 nm-scale emittance with 100 pC beam charge combining the advatnages of these advanced cathodes with strategiaally placed apertures along the beamline.

Over the past 12 months the entire process has been completed twice with mixed results. We did achieve generation of an electron beam from an alkali antimonide cathodes in a S-band high gradient gun and the results were reported in an invited talk at the P3 workshop in November 2021. The photocathodes were characterized in terms of quantum efficiency and thermal emittance at different wavelengths (the three harmonics of the Ti:Sa laser). Unfortunately, due to uncontrolled environment during transport, the quantum efficiency of the cathodes measured at arrival at UCLA was orders of magnitude lower than what measured at Cornell before shipping. Still, QE approaching 1% level were measured in the UV. This represented a 3 orders of magnitude improvement for the UCLA Peagsus facility when compared to the Cu cathode previously installed and enabled a series of (very lossy) laser shaping manipulation benefitting some of the high profile (THz-FEL) experiments on the beamline.

In terms of photocathode science, we are nowhere near the conclusion of these efforts as we do need to complete a high QE cathode transfer and to characterize the cathode at high field close to threshold.

Second, our efforts have been noted by the broader community and SLAC invited Cornell and UCLA to a new DOE BES initiative aimed at determining the best cathode for the LCLSII-HE injector. This is a very important problem as lasing at the highest possible energy (20 keV) will require better than state-of-the-art performance from the photoinjector.

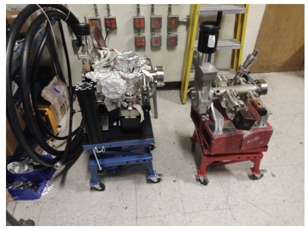


Figure 1. Two suitcases ready for installation/transportation at the UCLA Pegasus laboratory. The one on the right is being shipped back to Cornell to be loaded with cathode. The one on the left has an improved transfer system and it is currently installed on the UCLA Pegasus beamline.

PROJECT UPDATE: Advanced beam manipulations enabled by novel computational techniques in beam physics

Musumeci student: Eric Cropp (graduate student)

This project is connected to the application of machine learning techniques to improve the performances of the HiRes beamline at LBNL. UCLA graduate student E. Cropp spent one year at LBNL working with D. Filippetto on implementing new smart feedback algorithms that combine particle tracking simulations and actual data from the machine to ensure improved stability, shorter bunch lengths and smaller beam emittances.

The continuous presence of Eric at LBNL has been instrumental for the continuous operation of the facility which in the last twelve months has completed ultrafast electron diffraction studies on a number of novel samples and has strengthened the collaboration with machine learning groups at SLAC and LANL. Eric has been a key-member in all of these collaborations and in fact after Daniele’s departure from LBNL, he is one of the main candidates to take on the responsibility of running the beamline.

One notable achievement is the successful prediction of electron bunch time-of-arrival in the Hires UED setup using virtual diagnostics based on various machine parameters. Eric has demonstrated the technique's ability to predict time-of-arrival with an accuracy of 200 fs or better, exceeding the performance of closed-loop feedback systems at HiRes. This method has the potential to enhance the temporal resolution of pump and probe UED scans. The publication of these results is forthcoming in PRAB.

After experimental activities at LBNL ramped down at the end of last summer, Eric has switched his attention to the FAST beamline at Fermilab. This beamline is a candidate for the 100pC-100nm CBB quest and Eric carried out preliminary measurements there aimed at validating the simulation model which will be used in the optimization of the beam dynamics.

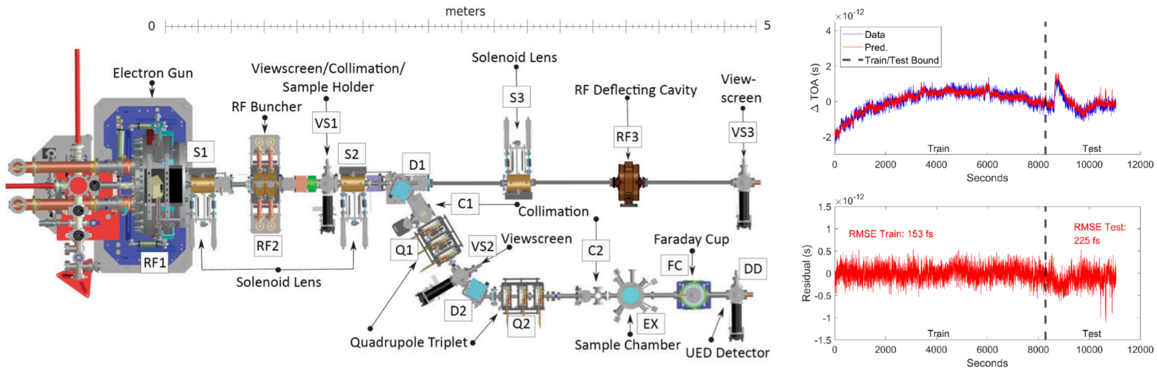


Figure 2. Left) The HiRES beamline. The UED beamline starts at D1 and goes through the dogleg to DD, while the diagnostic beamline goes straight from D1 to VS3. Right) Time of arrival fluctuations measured using the position of the beam on the final screen after the deflecting cavity while the feedback systems were engaged. Residual drifts not corrected by the feedback are still visible. The prediction of the time-of-arrival using non-destructive measurement of all the available machine parameters is also shown. Using this virtual diagnostic approach, time-stamping the uncertainty due to long-term drifts is reduced to 200 fs level (see residual plot on bottom right) similar to the shot-to-shot short-term jitter.

Piot, Philippe (Northern Illinois University)

Publications

Papers

N. Majernik, G. Andonian, W. Lynn, S. Kim, C. Lorch, R. Roussel, S. Doran, E. Wisniewski, C. Whiteford, P. Piot, J. Power, and J. B. Rosenzweig, “Beam shaping using an ultrahigh vacuum multileaf collimator and emittance exchange beamline,” *Phys. Rev. Accel. Beams*, vol. 26, no. 2, p. 022801, Feb. 2023, doi: 10.1103/PhysRevAccelBeams.26.022801.

J. Jarvis, V. Lebedev, A. Romanov, D. Broemmelsiek, K. Carlson, S. Chattopadhyay, A. Dick, D. Edstrom, I. Lobach, S. Nagaitsev, H. Piekarz, P. Piot, J. Ruan, J. Santucci, G. Stancari, and A. Valishev, “Experimental demonstration of optical stochastic cooling,” Nature, vol. 608, no. 7922, pp. 287–292, Aug. 2022, doi: 10.1038/s41586-022-04969-7. [Online].
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Talks

A. Al Marzouk, S. Benson, K. Deitrick, D. Doran, J. Guo, G. Ha, A. Hutton, C. Mitchell, G.-T. Park, P. Piot, J. Power, J. Qiang, R. Ryne, S. Wang, C. Whiteford, E. Wisniewski, and T. Xu, “Preliminary Tests and Beam Dynamics Simulations of a Straight-Merger Beamline,” Proceedings of the 5th North American Particle Accelerator Conference, vol. NAPAC2022, p. 4, 0.838 MB, 2022, doi: 10.18429/JACOW-NAPAC2022-MOPA72.

A.J. Dick, P. Piot, and J. Jarvis, “Modeling of the optical stochastic cooling at the IOTA storage ring using ELEGANT”, Proceedings of the 13th International Particle Accelerator Conference, IPAC2022, p. 2307, 2022, doi: 10.18429/JACoW-IPAC2022-WEPOMS029

A.J. Dick and P. Piot, “Electron beam shaping techniques using optical stochastic cooling”, Proceedings of the 13th International Particle Accelerator Conference, IPAC2022, p. 2307, 2022, doi: 10.18429/JACoW-IPAC2022-WEPOMS028

A. Afnan, P. Piot, and R. Ryne, “First-Principle Simulations of Electron-Bunch Compression Using a Large-Scale Lienard-Wiechert Solver”, Proceeding of the Advanced Accelerator Concept Workshop, November 2022 (accepter, in press)

A.J. Dick and P. Piot, “Application of the Optical Stochastic Cooling Mechanism to Electron-Beam Shaping in Storage Rings”, Proceeding of the Advanced Accelerator Concept Workshop, November 2022 (accepter, in press)

Posters

Collaborating Groups

1. Cornell University: Jared Maxson, James Rosenzweig
2. UCLA: Pietro Musumeci
3. FNAL: J. Jarvis, J. Ruan
4. LBNL: R. Ryan

PROJECT UPDATE: Optical Transport and Beam Manipulation for Optical Stochastic Cooling

Piot student: Austin (AJ) Dick (graduate student)

This project relates to the “beam-cooling” thrust of BDC and specifically focused on the development of hardware and numerical methods to support the optical-stochastic-cooling (OSC) experiment at the IOTA electron storage ring at Fermilab. In year 7, our group contributed to the analysis of the successful experiment on passive OSC with focus on benchmarking the experiment with the beam-tracking model developed in ELEGANT. The agreement between simulation and experimental observation is excellent and a paper was drafted with collaborator from Fermilab (planned submission to PRAB imminent). The validated model was also applied to investigate the performance of amplified OSC a phase II experiment in preparation at IOTA. Additionally, the OSC model was further used to demonstrate that controlling (e.g. modulating) the amplification of radiation on a turn-by-turn basis can provide precise control over the stored electron beam distributions. Several examples were studied and a model of a storage ring implementing two OSC insertions was developed to further illustrate the capability of the zOSC technique to beam control and tailoring.

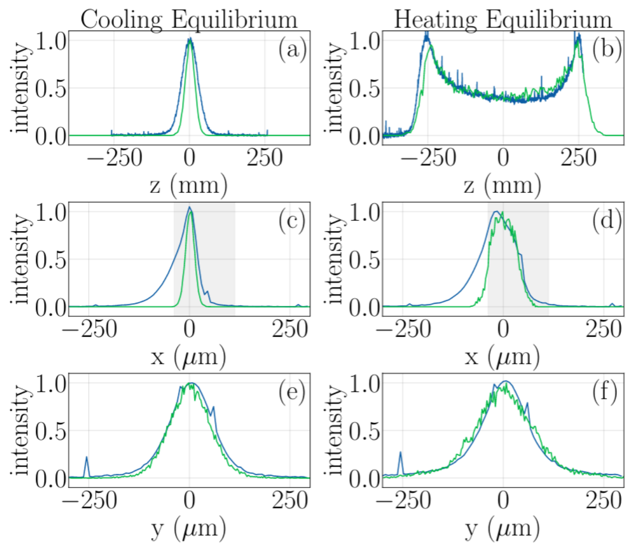

Figure OSC1: Comparison of the experimental (blue) and simulated (green) equilibrium distributions in longitudinal (a,b), horizontal (c,d) and vertical (e,f) directions for the cooling and heating modes of OSC. The shaded area in plots (c,d) represents the regions where the measurements are accurate (see text for details). The development of a tail toward negative values of x is a measurement artifact.

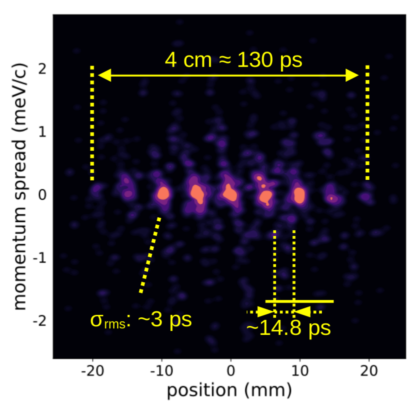

Figure OSC2: Generation of a train of electron bunches in an electron storage ring using a dual-OSC system. In this case the beam consists of 9-ps bunches with 14.8-ps separation.

PROJECT UPDATE: Exploring the Impact of Radiation Field on Brightness

Piot student: Afnan Al Marzouk (postdoc)

This project relates to the "beam-control" thrust and specifically focused on understanding the impact of radiation fields on brightness and beam control in general (e.g., beam shaping). The project uses and contribute to the development of a first-principal simulation program

(LW3D) to understand the impact of radiation field on the beam dynamics, During Year 6, a self-consistent model (generation of field and update of particle according to the corresponding force) was implemented and tested. A model of a four-bend bunch-compressor chicane was implemented and preliminary results on emittance dilution were generated. The current effort is to minimize this emittance dilution using well-known approaches (e.g., control of the beam lattice function) and gain further confidence in LW3D. Additionally, LW3D was used to model a straight merger concept consisting of a cavity superimposed with a dipole magnet. Such a merger was proposed as an alternative merger to inject “fresh” bright beam in an energy-recovering linac while recycling the “spent” bunch. The modeling includes the impact of radiation effects as the fresh bunch propagates in the time-dependent RF field superimposed with the magnetostatic field. A related experiment was performed at AWA that demonstrated the main feature of the straight merger and the data analysis and comparison with numerical simulations is underway.

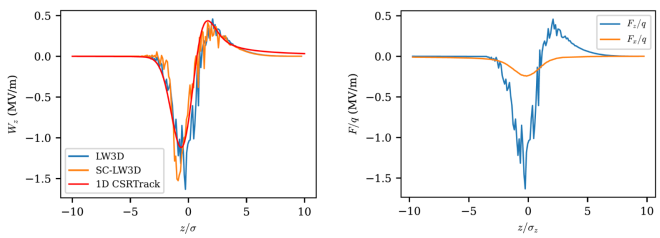

Fig. RAD1: **left:** Comparison of the CSR longitudinal wakefield field simulated with LW3D and self-consistent (SC) LW3D with the 1D semi-analytical model (1D CSRTrack). **Right**: Comparison of the longitudinal Fz and bending-plane transverse Fx forces.

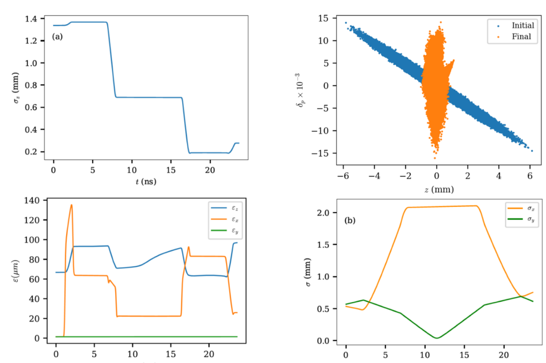

Fig. RAD2: SC-LW3D simulation of a bunch-compression chicane for a 1-nC bunch. **top-left:** evolution of the bunch length, along the chicane; **top-right:** initial (blue) and final (orange) longitudinal phase spaces, **lower-left:** emittances evolution along the chicane; **lower-right:** transverse beam sizes evolution.

Publications

Papers

J. I. Mann, Y. Li, J. K. Nangoi, T. Arias, and J. B. Rosenzweig, “Material normal energy distribution for field emission analyses from monocrystalline surfaces,” *J. Phys.: Conf. Ser.*, vol. 2420, no. 1, p. 012033, Jan. 2023, doi: 10.1088/1742-6596/2420/1/012033.

T. Paschen, R. Roussel, L. Seiffert, B. Kruse, C. Heide, P. Dienstbier, J. Mann, J. Rosenzweig, T. Fennel, and P. Hommelhoff, “Ultrafast Strong-Field Electron Emission and Collective Effects at a One-Dimensional Nanostructure,” *ACS Photonics*, Feb. 2023, doi: 10.1021/acsphotonics.2c01551. [Online]. Available: https://doi.org/10.1021/acsphotonics.2c01551

J. Mann and J. Rosenzweig, “A Coherent Bi-Directional Virtual Detector for the 1-D Schrodinger Equation,” arXiv, arXiv:2205.10461, May 2022 [Online]. Available: http://arxiv.org/abs/2205.10461

G. Lawler, N. Majernik, J. Mann, N. Montanez, and J. Rosenzweig, “Development of Nanopatterned Strong Field Emission Cathodes,” *Proceedings of the 5th North American Particle Accelerator Conference*, vol. NAPAC2022, p. 3 pages, 15.012 MB, 2022, doi: 10.18429/JACOW-NAPAC2022-THYD5.

J. Mann, T. Arias, S. Karkare, G. Lawler, J. K. Nangoi, J. Rosenzweig, and B. Wang, “Simulations of Nanoblade Cathode Emissions with Image Charge Trapping for Yield and Brightness Analyses,” *Proceedings of the 5th North American Particle Accelerator Conference*, vol. NAPAC2022, p. 4 pages, 0.859 MB, 2022, doi: 10.18429/JACOW-NAPAC2022-TUPA86.

G. Lawler, F. Bosco, A. Fukasawa, Z. Li, N. Majernik, J. Parsons, J. Rosenzweig, Y. Sakai, B. Spataro, and S. Tantawi, “Cyborg Beamline Development Updates,” *Proceedings of the 5th North American Particle Accelerator Conference*, vol. NAPAC2022, p. 4 pages, 6.730 MB, 2022, doi: 10.18429/JACOW-NAPAC2022-TUPA80.

G. Lawler, A. Fukasawa, N. Majernik, J. Parsons, J. Rosenzweig, Y. Sakai, and A. Suraj, “CrYogenic Brightness-Optimized Radiofrequency Gun (CYBORG),” *Proceedings of the 13th International Particle Accelerator Conference*, vol. IPAC2022, p. 4 pages, 2.884 MB, 2022, doi: 10.18429/JACOW-IPAC2022-THPOST046.

G. Lawler, A. Fukasawa, N. Majernik, and J. Rosenzweig, “Temperature Dependent Effects on RF Surface Resistivity,” *Proceedings of the 13th International Particle Accelerator Conference*, vol. IPAC2022, p. 4 pages, 1.555 MB, 2022, doi: 10.18429/JACOW-IPAC2022-THPOST045.

G. Lawler, N. Majernik, J. Mann, N. Montanez, J. Rosenzweig, and V. Yu, “Emittance Measurements of Nanoblade-Enhanced High Field Cathode,” *Proceedings of the 13th International Particle Accelerator Conference*, vol. IPAC2022, p. 4 pages, 8.059 MB, 2022, doi: 10.18429/JACOW-IPAC2022-MOPOMS033.

Talks and Posters

J. I. Mann, Y. Li, J. K. Nangoi, T. Arias, J. B. Rosenzweig, Material normal energy distribution for field emission analyses from monocrystalline surfaces, in Proc. IPAC’22, Bangkok, Thailand, 2022, pp. 713-716 doi: 10.18429/JACoW-IPAC2022-MOPOMS034

B. Wang, J. I. Mann, G. E. Lawler, J. K. Nangoi, S. Karkare, T. A. Arias, J. B. Rosenzweig, Simulations of Laser Field Emission from Nanostructures with Image Charge Trapping and Band Structure Transitions, in Proc. IPAC’22, Bangkok, Thailand, 2022, pp. 717-720 doi: 10.18429/JACoW-IPAC2022-MOPOMS036

J. I. Mann, B. Wang, G. E. Lawler, J. K. Nangoi, S. Karkare, T. A. Arias, J. B. Rosenzweig, Simulations of Nanoblade Cathode Emissions with Image Charge Trapping for Yield and Brightness Analyses, in Proc. NAPAC’22, Albuquerque, NM, USA, 2022, pp. 535-538 doi: 10.18429/JACoW-NAPAC2022-TUPA86

Gerard Lawler International Workshop on Breakdown Science and High Gradient Technology (HG2022)

Gerard Lawler IPAC22 (May, 2023)

Gerard Lawler NAPAC22 (Aug, 2023)

Gerard Lawler 10th International Workshop on the Mechanisms of Vacuum Arcs (MeVArc 2022)

Gerard Lawler C3 Workshop (Oct, 2022)

Gerard Lawler C3 Workshop (Feb, 2023)

James Rosenzweig “UCLA Cryo-RF Research for C3” C3 Workshop (Oct, 2022)

James Rosenzweig “UCLA Cryo-RF Research for C3” C3 Workshop (Feb, 2023)

Nathan Majernik ”Multi-leaf Collimator for 3D beam shaping” IPAC22 (May, 2023)

Collaborating Groups

1. Cornell University: Tomás Arias, Jared Maxson

2. UCLA – Pietro Musumeci
3. University of New Mexico: Sandra Biedron
4. Northern Illinois University: Philippe Piot

Awards and Honors

James Rosenzweig, 2022 Advanced Accelerator Prize
James Rosenzweig, European Physical Socitey 2023 Hannes Alfven Prize in Plasma Physics

PROJECT UPDATE: Extreme High Brightness Electron Source from Intense Laser Illumination of Nano-Blades

Rosenzweig student and postdoc: Joshua Mann (graduate student), Gerald Lawler (graduate student), Nathan Majernik (postdoc)

The brightness of an electron beam, as a conserved quantity, must be improved by increasing the brightness immediately at the cathode. While much work is being put into materials which produce a lower MTE more efficiently, there are two other (not necessarily independent) ways to improve brightness: increasing the extraction field and decreasing the emission area. Nanostructured photo-cathodes serve both purposes: the nano-plasmonic field enhancement localizes the field to a small area of nm scale and increases the intensity of the field according to the enhancement factor. Our primary subject, the nanoblade, is a nanopatterned ridge which naturally produces a flat beam and is more thermos-mechanically robust than the ubiquitous nanotip, as proven by experiment.

Increasing laser intensity spurs the process of non-linear photoemission, with surface fields exceeding $\sim 10\ \mathrm{V/nm}$. The field forces the ionized electrons into ponderomotive motion, resulting in rescattering which boosts the maximum electron emitted energy, remarkably, up to the keV range. The rescattering process is well-understood classically, with the electron elastically scattering at the surface. We studied this semi-classical model of rescattering (as part of our "$11U_p$" project) and found the peak electron energy as a function of the peak field, laser wavelength, and nano-structure design. The cutoff energy we found in this way proved to be an improvement over the standard $10U_p$ approximation, as seen by the comparison to our time-dependent Schrodinger equation (TDSE) simulation in Fig. 1 below. This work has been submitted to JOSAB.

We also made adjustments to our TDSE simulation. One improvement is in the fictitious loss of charge in the metallic simulation boundary which exacerbated the restoring image force, and reduced the yield at higher intensities. We now use an image charge model which treats the metal as a perfect conductor, with no internal fields. This eliminates the fictitious charge loss and gives the nearly linear yield-to-laser-intensity relationship at high intensities. We also concluded our efforts on including an effective mass transition at the metal surface. We found that the difference between calculations with and without this feature are mostly dominated by noise at low field and is only perturbative at high field. As our material of study, gold, has an effective mass of 1.1 vacuum electron mass, and we elide over this feature for now.

Additionally, the project investigating more analytical approaches to creating computationally efficient TDSE solutions, headed by Benjamin Wang, came to a close. All the options we considered provided good results at low fields but broke down at high fields, requiring more resources and precision as the fields increased. Thus, directly and numerically integrating the TDSE serves our purposes best in this case.

Lastly, we have been making progress on our material normal energy distribution (MNED) project, which aims to characterize various crystalline metals' abilities to field emit under illumination of a strong laser or a static field by finding the source current within the metal as a function of the normal energy. Yiming Li was the primary undergrad for this project, and Kevin Nangoi and Tomas Arias were integral to it. Thus far we have used density-functional theory (DFT) to create MNEDs for the Au, Cu, and W (100), (110), and (111) surfaces. Interestingly, we found that the Au(111) and Cu(111) surfaces may have a higher effective work function for tunneling as no electrons at the Fermi surface have zero transverse crystal momentum.

Experimentally, our new lab space at the UCLA Science and Technology Research Building has been renamed Megavolt InTense High-gradient Research Accelerator (MITHRA) in order to mark the turning on of the new accelerator including the production of photoelectrons. Additional emission data from the latest round of nanoblade measurements were analyzed and theoretical predictions for the emittance, yield, and brightness from nanoblades were refined further. The production of one nanoblade beamlet is shown imaged on a phosphor screen (Figure 2). Preliminary results were presented at IPAC and NAPAC in the context of the general development of nanopatterned cathodes*, i.e.,* those with nanoscale features. The peer reviewed publication of these results is now moving forward with this additional data analysis. Changes to the fabrication method to improve simplicity and uniformity of the existing nanoblade cathodes are under development (Figure 3). Changes to the spacing using these new methods has reopened the question of whether plasmonic effects can be observed from blade-to-blade interaction. A collaboration with the UCLA School of Engineering has begun examining nanoblade cathodes for spacecraft propulsion. One such test device which has been designed and produced is shown below.

In the beam dynamics work, we have produced a preliminary design of new, multi-pulse UCXFEL for 3D imaging purposes, based on ultra-high-brightness electron beams from cryo-RF sources. We are proceeding to understand the beam challenges in producing high fluxes of Angstrom level photons in the UCXFEL, particularly in short and long-range beam breakup. This next generation instrument will be co-developed with the wide UCXFEL collaboration to develop a proposal for constructing a CHIPS Act-funded coherent radiation source for ptychographic tomography-based 3D nm-scale metrology, an area of urgent national need.

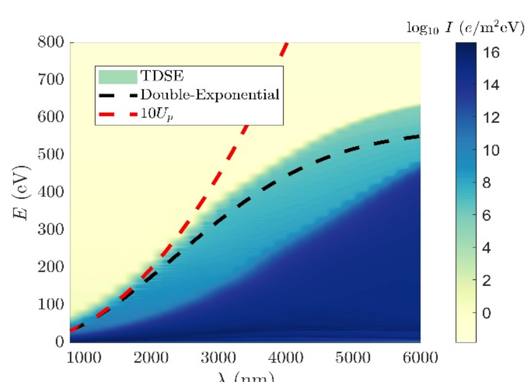

Figure 3. Comparison of the $\mathbf{10}\boldsymbol{U_p}$ approximation (red), the semi-classical result that we found (black) and the TDSE-derived emission spectrum (color). Our result better matches the TDSE calculation, particularly at high wavelengths.

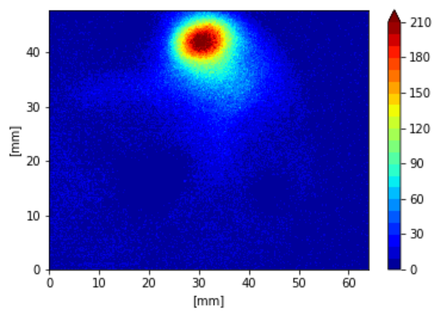

Figure 4. Production of one nanoblade beamlet from high field emission.

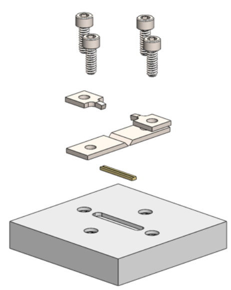

Figure 5. Assembly for nanoblade-based space-craft propulsion testing

PROJECT UPDATE: Optimization of ultra-compact free-electron laser performance with very low MTE photocathodes

Rosenzweig students: Joshua Mann (graduate student), Gerald Lawler (graduate student)

Construction and commissioning of the new 1 MeV CYBORG beamline at the MOTHRA (Multi-Objective Testing of High Gradient Radiofrequency Accelerators) lab is ongoing and a good deal of progress has been made. A MW rated Cband klystron and two functional 45K cryostats are now operational. The CrYogenic Brightness-Optimized Radiofrequency Gun (CYBORG) was delivered to UCLA. It was inspected and characterized by low level RF measurements. A photo of new photogun (right). Its operational temperature regime is 45-77K and is designed to produce fields greater than 120 MV/m with relatively low input power (around 1 MW). Several novel concepts are features of this design including its operation in the C-band frequency range (5.712 GHz @ 77K for example). Critical infrastructure such as a higher power cryocooler have been added increasing the cooling capacity by a factor of 3-4 at 45K. In addition, the RF subsystem which requires a new custom-made modulator for the C-band frequency has progressed to near completion. Functioning quadrupole and a dipole have been powered and precise characterization of the fields they produce has begun after the construction of a three-axis high precision measurement gantry. In addition, the initial solenoid for the beamline has been constructed and tested successfully up to 10 A. The components are shown in their approximate final location (left). The large cube is the cryostat which houses CYBORG; the green magnets are the quadrupoles; the red magnet is the dipole; and the octagonal element to the right is the newly assembled solenoid which will be added to the beamline once measurement is complete.

Because of the novel higher frequency in the C-band we are using for our existing and future photoguns. Additional studies of RF cavities at cryogenic temperatures in this frequency are required. These basic physics experiments are necessary to understand the thermal characteristics of copper, develop novel improvements to cavity shapes, and to establish the upper bounds on cavity fields all of which are used to improve beam brightness preservation off the cathode. We are collaborating primarily with Radiabeam Technologies, Los Alamos, and SLAC National Laboratories on these projects.

a. Next steps in research

The theoretical and computational nanoblade emission work has been brought to a high level of maturity and is mainly in need of writing up in final form, to lead to publications and Joshua Mann's thesis. The only remaining project with significant new work is the thermomechanical analysis of the fairly impressive fact the usable fields in the nano-blade scenario exceed those of nano-tips by a factor of over three. This gives an order of magnitude in ponderomotive energy increase. We are also evaluating future directions in use of such emission processes in HHG radiation production.

The near term (6 month) next steps for the CYBORG beamline are commissioning the gun to a very high gradient (>120 MV/m) at a cryogenic temperature (<77K). This will be followed by cryogenic dark current studies and the production of photoelectrons by the end of the summer.

The development steps have been refined into two phases. Phase1 will produce photoelectrons from a cryogenic Cu cathode to allow for continued parallel development of the cryogenic load lock and the higher precisions MTE measurement setup utilizing the TEM grid method. Once such example Cu cathode is shown below. Within the coming year, CYBORG commissioning will be complete and the associated beamline at the MOTHRA laboratory will produce photoelectrons for the first time from a high gradient photogun at cryogenic temperatures. Low precision measurement of MTE is the completion condition for Phase1:config1 leading to Phase1:config2 the addition of high precision MTE measurements on the CYBORG beamline.

Phase2 of the CYBORG beamline will incorporate the low temperature load lock and allow for the insertion of novel cathodes for low temperature high gradient testing. The nanoblade characterization will recommence as the diagnostic setup is moved to the new MITHRA laboratory laser room, the current home of the 800nm 35fs Ti:Sapphire laser setup. In addition, the new fabrication process which we hope will make more uniform blades will be implemented and the produced cathodes will be able to be measured in the new lab space.

In the beam dynamics sector, we are now proceeding to understand the beam challenges in producing high fluxes of Angstrom level photons in the UCXFEL. This next generation instrument will be co-developed with the wide UCXFEL collaboration to develop a proposal for constructing a CHIPS Act-funded coherent radiation source for ptychographic tomography-based 3D nm-scale metrology, an area of urgent national need.

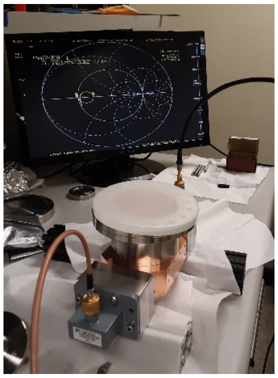

Figure 6. New 0.5 cell cryo-RF CYBORG photogun.

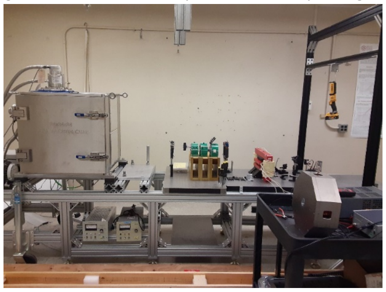

Figure 5. CYBORG beamline at UCLA MOTHRA

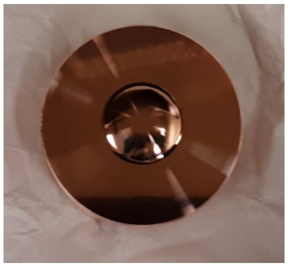

Figure 6. First Cu cryocathode in CYBORG RF photogun

Shen, Kyle (Cornell)

Publications

Papers

C. T. Parzyck, A. Galdi, J. K. Nangoi, W. J. I. DeBenedetti, J. Balajka, B. D. Faeth, H. Paik, C. Hu, T. A. Arias, M. A. Hines, D. G. Schlom, K. M. Shen, and J. M. Maxson, "Single-Crystal Alkali Antimonide Photocathodes: High Efficiency in the Ultrathin Limit," Phys. Rev. Lett., vol. 128, no. 11, p. 114801, Mar. 2022, doi: 10.1103/PhysRevLett.128.114801.

Talks

Vivek Anil, Christopher T Parzyck, Elena Echeverria, Alice Galdi, Chad A Pennington, Jared Maxson, Kyle M Shen. "Exploring the potential of Na3Bi thin films as high-brightness photocathodes", APS March Meeting 2023, Las Vegas, NV : https://meetings.aps.org/Meeting/MAR23/Session/S23.2

Posters

V. Anil and K. M. Shen. 2022. "Exploring the potential of Dirac semimetals as low MTE photocathodes." [Poster]. Center for Bright Beams Annual Meeting, 1-3 June, The University of California, Los Angeles.

V. Anil and K. M. Shen. 2022. "Exploring the potential of Dirac semimetals as low MTE photocathodes." [Poster]. NSF/DOE/AFOSR Quantum Science Summer School (QS3), 18-29 July, The University of California, Santa Barbara.

Collaborating Groups

5. Cornell University: Melissa Hines, Jared Maxson

PROJECT UPDATE: Atomically Ordered & Engineered Materials for Photocathodes

Shen student: Vivek Anil (graduate student)
We (Vivek Anil, together with other members of Jared Maxson's group and my own) are exploring of Dirac semimetals as low MTE photocathodes. Dirac semimetals are a class of materials that are promising for low MTE photocathodes. The brightness of an electron beam scales inversely with MTE, and so limiting MTE would improve photocathode performance. The MTE of a photocathode can be related to its band structure, which for Dirac semimetals is characterized by linearly dispersing bands in all three momentum directions at a particular momentum point. Vivek has simulated MTE versus excess energy by directly relating the band structure of Dirac semimetals to MTE. Our simulations predict that Dirac semimetals with steep Dirac cones centered at zero-transverse momentum points would be low MTE photocathodes.

To test this hypothesis, we wrote and submitted a PARADIM proposal to grow thin films of the Dirac semimetal Na_3Bi by molecular beam epitaxy (MBE) and characterize these films' utility as photocathodes by quantum efficiency (QE) and ARPES measurements. Our proposal was accepted by PARADIM, and we have commenced work on this project at the PARADIM Thin Film Facility. We have been successful in depositing epitaxial Na_3Bi films on sapphire and silicon substrates, and our ARPES spectra indicate the presence of the material's characteristic Dirac cone like band structure (Fig. 1a). ARPES Fermi surface maps taken at 21.2 eV (Fig. 1b) and 10.3 eV photon energies show that the overwhelming majority of the photoemission originates from the center of the Brillouin zone (states with zero transverse momentum), which suggest this material is very promising for low MTEs. Our QE spectral response measurements (Fig. 1d) show that the QEs of our films are ~10-4 and ~10-5 in the violet and blue, respectively, which are orders of magnitude better than currently used metallic photocathodes at these visible wavelengths. By biasing the sample with respect to the electron analyzer in the ARPES chamber, we were also able to measure the work functions of our films (Fig. 1c); we measure work functions that correspond to visible light energies, which is desirable for photocathode operation in accelerators. This project is progressing well – we are planning to perform a second round of experiments where we will directly measure the MTE by biasing the sample using our photoemission analyzer and manipulator, after which we plan to write a paper on these results.

For this project, Elena Echeverria, Chad Pennington, and Alice Galdi from Jared Maxson's group assisted with the growths and QE measurements. Christopher Parzyck helped with some of the initial setup of the QE measurements. Members of Prof. Darrell Schlom's group (outside of CBB) also helped: Anna Park helped with some of the growths and x-ray diffraction attempts, Dr. Yorick Birkholzer carried out some ex-situ x-ray photoelectron spectroscopy, and Dr. Matthew Barone helped with source and MBE chamber preparation for the growths. Dr. Brendan Faeth (outside of CBB) remotely provided instruction and helped with troubleshooting related to the ARPES measurements. Prof. Jared Maxson and Prof. Kyle Shen co-advised this project.

In addition to this project, we are continuing to perform experiments on complex oxide heterostructures as photocathodes. A recent report in Nature by Hong et al. has reported $SrTiO_3$ as a potentially revolutionary high-brightness photocathode. We intend to reproduce these experiments using our group's oxide MBE and ARPES system, together with Profs. Maxson, Karkare, and Padmore.

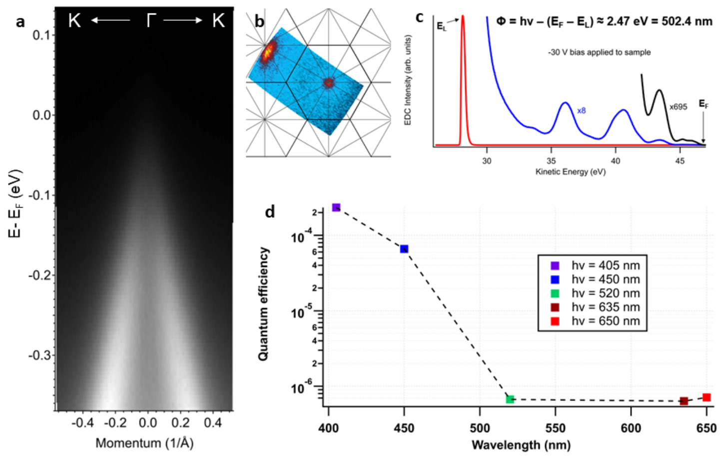

Figure 7: (a) ARPES spectra of Dirac cone and (b) Fermi surface of Na3Bi taken with He-I photon energy. (c) Measurement of work function of Na3Bi sample. (d) Measured quantum efficiencies at different visible wavelengths.

Sibener, Steve (University of Chicago)

Publications

Papers

1. S. A. Willson, R. G. Farber, A. C. Hire, R. G. Hennig, and S. J. Sibener, “Submonolayer and Monolayer Sn Adsorption and Diffusion Behavior on Oxidized Nb(100),” J. Phys. Chem. C, Jan. 2023, doi: 10.1021/acs.jpcc.2c08458. [Online]. Available: https://doi.org/10.1021/acs.jpcc.2c08458

[1]

2. A. A. McMillan, C. J. Thompson, M. M. Kelley, J. D. Graham, T. A. Arias, and S. J. Sibener, “A combined helium atom scattering and density-functional theory study of the Nb(100) surface oxide reconstruction: Phonon band structures and vibrational dynamics,” *J. Chem. Phys.*, vol. 156, no. 12, p. 124702, Mar. 2022, doi: 10.1063/5.0085653.

Talks and Posters

6. MRS Spring Meeting 2022, May 8-13, 2022, Talk, S.A. Willson, R.G. Farber, S.J. Sibener. “Optimizing Nb3Sn Growth for SRF Applications: Nanoscale Morphological and Electronic

Characterization of Intermetallic Adlayers on a Highly Ordered Nb Oxide" MRS Spring Meeting, Honolulu, HI, 2022.

7. 82nd Physical Electronics Conference, June 6-9, 2022, Poster, S.A. Willson, R.G. Farber, S.J. Sibener. "Optimizing Nb3Sn Growth: Nanoscale Morphological and Electronic Characterization of Superconducting Intermetallic Thin Films on Nb Oxides" 82nd PEC, Chicago, IL, 2022.

8. AVS 68 International Symposium, November 6-11, 2022, Talk, S.A. Willson, R.G. Farber, S.J. Sibener. "Development of a Predictive Model for Nb3Sn Thin Film Growth: Elucidating the Substrate-Mediated Diffusion Pathways Guiding Alloy Formation in Accelerator Infrastructure" AVS 68, Pittsburgh, PA, 2022.
9. R. G. Farber, S. A. Willson, A. C. Hire, R. G. Hennig, S. J. Sibener, "Investigating Materials Growth and Performance using Ultra-High Vacuum Surface Science Techniques", The University of Kansas, DyMERS Seminar, Lawrence, KS 2022 (Invited)

10. R. G. Farber, S. A. Willson, A. C. Hire, R. G. Hennig, S. J. Sibener, "Developing a Mechanistic Understanding of Nb_3Sn Growth: Sn Adsorption and Diffusion Behavior on (3×1)-O Nb(100)", *AVS 68th International Symposium and Exhibition*, Pittsburgh, PA, 2022

11. R. G. Farber and S. J. Sibener, "Basics of Ultra High Vacuum Systems", *CBB Annual Meeting*, Los Angeles, CA 2022

12. Caleb Thompson, Alison McMillan, Michelle Kelly, Tomas Arias, S. J. Sibener, High temperature persistence and bonding of the Nb(100) surface oxide reconstruction shown by high resolution helium diffraction, inelastic time-of-flight measurements, and density functional theory calculations, ACS Fall National Meeting, Chicago, IL 2022

13. Caleb Thompson, Alison McMillan, Michelle Kelly, Tomas Arias, S. J. Sibener, 82nd Physical Electronics Conference, Chicago, IL 2022

Collaborating Groups

1. Cornell University: Tomás Arias, Matthias Liepe, David Muller
2. University of Florida: Richard Hennig
3. FNAL: Sam Posen

PROJECT UPDATE: Investigating the Atomic and Micron-Scale Morphological Development of Nb3Sn Leading to Smooth Homogeneous Thin Films

Sibener students and theme postdoc: Rachael Farber (theme postdoc), Sarah Willson (graduate student), Van So (graduate student)

Year 7 of this project has directly contributed to CBB **Phase II, Objective 1**: Advanced SRF Materials Growth: Developing improved growth methods and understanding the impact of realistic (non-ideal) surfaces on performance. Particularly, this work has served **Deliverable 1.1**: Methods of growth and their refinement leading to annually significantly enhanced performance for Nb_3Sn on Nb and Cu substrates.

This project aims to address this objective and deliverable by scaling fundamental surface chemistry studies to inform Nb_3Sn film growth on realistic Nb surfaces. Our previous studies within the Center for Bright Beams developed an understanding of how thermal and oxidatio n conditions impact initial Sn behavior on Nb(100) with atomic-scale sensitivity. The Sn/Nb(100) interface was visualized using *in situ* scanning tunneling microscopy (STM). **This past year, we commenced alloy growth studies utilizing the same highly controlled surface preparation and metal deposition techniques to grow Nb_3Sn films on polycrystalline Nb surfaces.** The scaling of our Nb_3Sn growth studies to alloy films gives a thorough understanding of how various factors in the deposition procedure impact intermetallic interactions.

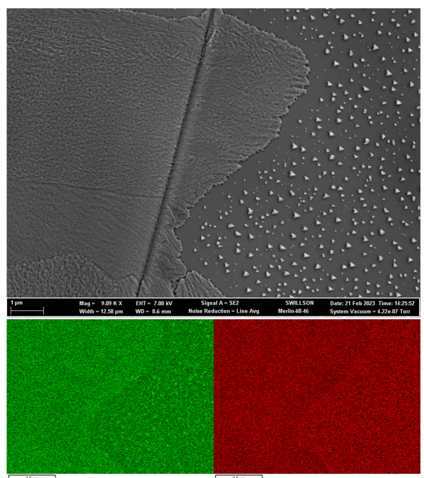

Fig 1. SEM of Nb_3Sn layer on polycrystalline Nb (top) with accompanying EDS maps for Sn (green) and Nb (red). Nb_3Sn layer is shown on the left half of the SEM/EDS images with the right consisting of unreacted elemental Sn on Nb. XRD analysis confirms Nb_3Sn unit cell. The Nb surface was an NbO overlayer and was exposed to Sn at 850 °C.

During Year 7, we continued our mechanistic studies looking at how Nb substrate and Sn vapor conditions impact Sn/Nb interfacial behavior during Nb_3Sn formation. The aim is to understand how deposition conditions, such as the Nb oxide morphology, impact the growth of homogeneous high performing Nb_3Sn films. To elucidate these thermally induced Nb-Sn surface-mediated interactions, Sn was deposited on polycrystalline Nb. *Ex situ* characterization tools, such as SEM (scanning electron microscopy), EDX (energy-dispersive X-ray spectroscopy), and XRD (X-ray diffraction), were employed in addition to the existing *in situ* surface characterization techniques. Thus far, we have determined that the crystallinity of the Nb substrate plays a vital role in alloy growth. Sn deposited on polycrystalline Nb has significantly greater thermal stability than on single crystal Nb(100). We have also observed that the NbO overlayers formed on Nb(110) and Nb(111) have distinct affinity for deposited Sn, similar to our results from CBB Year

6 that compared Sn on (3×1)-O/Nb(100) and Nb_2O_5/Nb(100). Notably, our SEM/EDS results have confirmed that Sn must be deposited on a Nb surface > 800 °C to form the N b_3Sn film. Annealing Sn on Nb following a room temperature deposition resulted in incomplete surface wetting and desorption dominating before any observed alloy formation. However, annealing > 800 °C *during* the Sn deposition results in the formation of micron-scale Nb_3Sn domains (top panel, **Fig. 1**). EDS maps (bottom panels, **Fig. 1**) confirm the intermetallic composition of these domains and suggest the triangular islands correspond to unreacted metallic Sn. XRD analysis shows the formation of the Nb_3Sn unit cell on the surface following this procedure and ongoing efforts aim to develop a more comprehensive understanding of the structural morphology of Nb_3Sn formed on varying Nb oxides surfaces.

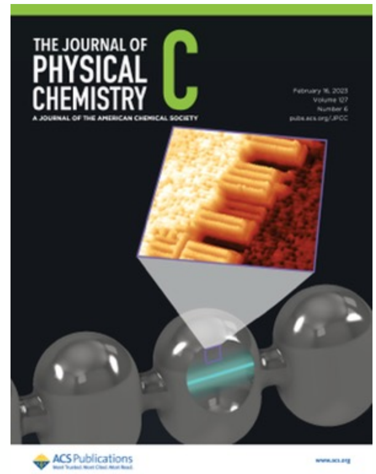

Fig 2. Cover of JPCC issue featuring publication with CBB research.

Additionally, the manuscript containing CBB research was published in the Journal of Physical Chemistry C. This work was a combined atomistic study involving experimental scanning tunneling microscopy (STM) and density functional theory (DFT) simulations of Sn adsorption, diffusion, and desorption on (3×1)-O/Nb(100). Theoretical calculations were done by CBB members Ajinkya Hire and Richard Hennig. A schematic depicting the surface of an SRF cavity being represented with an STM image was selected to be a supplemental journal cover (**Fig. 2**).

Our next step is to complete the ongoing Nb_3Sn growth studies on polycrystalline Nb. We aim to develop a matrix of how Nb oxidation, deposition temperature, and crystalline orientation each impact Nb_3Sn grain morphology. Completing this project will demonstrate how our previous Sn atomic studies can be applied to SRF optimization efforts, by examining how these subtle procedural changes impact properties of micron Nb_3Sn films.

The value in this work will be augmented by the utilization of *in situ* surface-sensitive structural characterization. A UV source, procured during Year 6 with CBB funds, is currently installed on our UHV chamber for the purpose of conducing UV photoelectron spectroscopy (UPS) measurements. UPS spectra provide valuable data evidencing intermetallic coordination by providing high enough resolution valence band spectra containing unique alloy signatures. These measurements will enable the study of the dynamic reactive Nb-Sn-O interface without surface contamination from *ex situ* analysis. This interfacial alloying study using surface sensitive UPS and our existing characterization techniques (XPS, AES, STM, etc.) gives valuable mechanistic information that cannot be obtained from examining already formed Nb_3Sn films. Along with *ex situ* structural and morphological analysis, the next steps in this project will leverage our experimental techniques to apply surface chemistry studies to inform procedural developments done by our collaborators in the SRF theme.

PROJECT UPDATE: Visualization of Nb_3Sn and Zr Doped Nb Growth Mechanisms to Inform Optimal Growth Procedures for Next-Generation SRF Materials

Sibener students and theme postdoc: Rachael Farber (theme postdoc), Sarah Willson (graduate student)

Our ongoing Year 7 research in the Sibener during CBB Year 7 investigated the growth behavior of Nb_3Sn works towards **Objective 2 – Multi-layers and inhomogeneous layers: Increasing RF performance via surfaces by design** and **Objective 3 – Advanced SRF materials growth: Developing improved growth methods and understanding the impact of realistic (non-deal) surfaces on performance** of the **Beam Acceleration** theme. Developing a mechanistic understanding of optimal Nb_3Sn growth will allow for Nb_3Sn films capable of operating with increased cavity efficiency in larger accelerating fields (**Deliverable**).

Our efforts during CBB Year 7 focused on developing codeposition procedures of Sn on Nb on Nb(100) and Nb foils to investigate Nb_3Sn growth behavior as it pertains to CBB Objective 2 – Multi-layers and inhomogeneous layers: Increasing RF performance via surfaces by design. We made considerable progress in this research topic, with X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), scanning tunneling microscopy (STM), and Auger electron spectroscopy (AES) data providing considerable insight into the mechanistic consequences of varying deposition procedures on optimal thin film Nb_3Sn growth. These mechanistic studies range from atomic-scale STM studies looking at early Nb-Sn interfacial interactions on single crystal Nb(100) (**Fig. 1)** to the growth of intermetallic films on polycrystalline Nb with both *in situ* and *ex situ* characterization. This work has provided insight pertaining to how changes to the Sn deposition procedure impact intermetallic interactions on both pristine and more realistic cavity surfaces.

In addition to this work looking at multi-layers and inhomogeneous layers as it pertains to Nb_3Sn growth, our collaboration with the Hennig group investigating Sn adsorption and diffusion on pristine (3×1)-O Nb(100) was published in *The Journal of Physical Chemistry C* in **2023** and the cover art created by Sarah Willson was featured as a supplemental cover. The calculated Sn adsorption energies and simulated STM images generated by the Hennig group are significant additions to the manuscript, and allow for more in depth conversations regarding the underlying physical phenomena guiding the observed Sn adsorption and diffusion behavior on pristine (3×1)-O Nb(100).

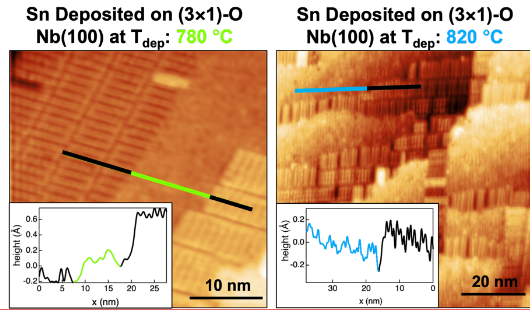

Fig. 1. STM images of the (3×1)-O Nb(100) surface following the deposition of 1 monolayer equivalence of Sn with the Nb substrate held at 780 and 820 °C during deposition (T_{dep}). Inset line scans highlight that the Sn adsorbate does not disrupt the Nb by incorporating into the subsurface at 780 °C while the Sn induces (3×1)-O Nb(100) reconstructions from incorporation at 820 °C.

PROJECT UPDATE: The Influence of Atomic Scale Surface Structural Changes and Composition on the Superconductivity of Nb_3Sn and Nb Due to Alloying, Doping, Domain Morphology, and Defects

Sibener students and theme postdoc: Rachael Farber (theme postdoc), Sarah Willson (graduate student), Caleb Thompson (graduate student), Michael Van Duinen (graduate student)

Our ongoing Year 7 research in the Sibener during CBB Year 7 contributed to CBB Phase II, Objective 1: Advanced SRF Materials Growth: Developing improved growth methods and understanding the impact of realistic (non-ideal) surfaces on performance. This project contributes towards Deliverable 1.1: Methods of growth and their refinement leading to annually significantly enhanced performance for Nb3Sn on Nb and Cu substrates.

A deeper understanding of atomic-scale surface structure's effect on the well-known superconducting state of the bulk would reveal the mechanisms determining which SRF cavity preparations aid or hinder the performance of a cavity. Our project, "The Influence of Atomic Scale Surface Structural Changes and Composition on the Superconductivity of Nb_3Sn and Nb Due to Alloying, Doping, Domain Morphology, and Defects" seeks to provide key fundamental knowledge of the physical properties and superconducting states that arise from surface structure. Understanding the role of surface atomic-scale structure and changes from SRF cavity preparations in accelerator performance and energetic efficiency would enable SRF cavity surfaces by design. **To obtain this, we must first understand the effect of the Nb surface on the superconducting states of Nb and then the effects on superconductivity of atomic scale surface structures resulting from SRF cavity treatment conditions.** To our knowledge, we have made the first measurement of electron phonon coupling (EPC) constant at any Nb surface and specifically the Nb(100) surface.

Helium atom scattering (HAS) has the unique capability to measure *in situ* high temperature atomic-scale surface structure and surface EPC constant. It has been shown through reasonable standard approximations such as the distorted wave Born approximation, Grimvall approximation, and ordinary second-order perturbation theory of the electron matrix elements that the HAS Debye-Waller (DW) factor is directly proportional to the EPC constant of metal surfaces. The DW factor is a multiplicative factor present in all scattered intensities that corrects for thermal excitations of phonons with changes in temperature. We have used HAS's unique capability to measure surface EPC constant and *in situ* high temperature atomic-scale surface structure to reveal the effects of the atomic-scale surface structure of Nb(100) on the superconductivity of Nb. The clean, metallic, unreconstructed Nb(100) surface was prepared and confirmed with HAS diffraction (Fig. 1) and AES. Diffraction scans were taken over the specular peak at a variety of temperatures. From the fitting parameters, a helium-to-surface well depth of 4.76 meV was obtained, and a surface Debye temperature of 296 K was determined from the well-known Debye model. The DW slope (Fig. 2) and well known tabulated (Ashcroft/ Mermin) values of the Fermi vector, work function, and number of free electrons per atom were used to calculate an EPC constant of 0.606 ± 2.38E-2 for the clean metallic Nb(100) surface.

This is the first recorded value of an EPC constant for any Nb surface. The recorded values for bulk Nb fall in the range of 1.0 – 1.2. This shows that the Nb(100) surface decreases the electron phonon coupling at the surface relative to the bulk. This indicates that the termination of the Nb lattice with a Nb(100) surface should be expected to significantly decrease the superconducting gap and increase the RF resistance relative to the bulk. To more deeply understand how this modification of the EPC constant would affect the surface's performance in an SRF cavity, we estimate appropriate superconducting properties and SRF cavity performance with well-known equations. Using McMillan's well known equation, our measured Debye temperature of 296 K, and the commonly used $\mu^* = 0.15$ for Nb, we obtain a critical temperature of 3.1 K from this EPC constant. Using this value to estimate the critical field, $H_c(T)$, of the material with the empirical parabolic relationship and a literature value for $H_c(0)$, we obtain a $H_c(2K)$ of 0.127 T at standard operating temperature, 2 K. Furthermore using a recently derived equation for the superheating field, H_{sh}, our calculated H_c at 2 K, and the literature value for κ we obtain a H_{sh} of 1.39 T at the standard operating temperature of 2 K. The H_{sh} obtained from the EPC constant measured for the Nb(100) surface is significantly less than values calculated for bulk Nb, cutting H_{sh} by 1/2. This clearly shows that the unreconstructed Nb(100) surface atomic-scale structure decreases the superconducting properties and should significantly lower the H_{sh}, limiting the accelerating fields of SRF cavities. These results are being written up into a paper for submission.

While the metallic (oxygen free) Nb surface is not in SRF cavities, this begins to build a fundamental understanding of the effects of the surface on superconducting properties, and now we can begin to understand the changes to the surface by oxides, carbon impurities, N doping, Sn/Zr alloying, and other SRF cavity preparations. This is an excellent demonstration of our capability to make *in situ* high temperature surface structure measurements and surface EPC measurements, allowing us to connect atomic scale surface structure to changes in superconducting properties and SRF cavity performance.

Currently, we are working on the (3x1)-O/Nb(100) surface, performing *in situ* high temperature atomic-scale surface structure and surface EPC constant measurements. This will build upon our understanding of the unreconstructed clean Nb(100) surface showing how the atomic-scale surface structure of the oxide reconstruction affects EPC and the properties of the superconducting state relevant to SRF cavity performance. Upcoming plans are to dose Sn, Zr, and N in varying amounts and mimic SRF cavity preparation procedures. We will measure simultaneously both *in situ* high temperature atomic-scale surface structure and surface EPC constants for these modified surfaces. Through the EPC constants, we can calculate superconducting properties such as critical temperature, superconducting gap, critical field, and superheating field, estimating the SRF cavity performance produced while making *in situ* high temperature atomic-scale structural measurements. In this way we plan to build a fundamental understanding of the atomic-scale surface structure produced by these treatments and their resulting effects on the superconducting state, enabling SRF cavity surfaces by design.

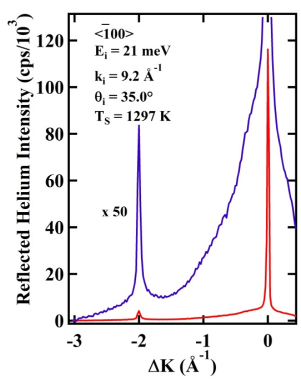

Figure 1: Helium diffraction from clean Nb(100) surface showing atomic scale structure.

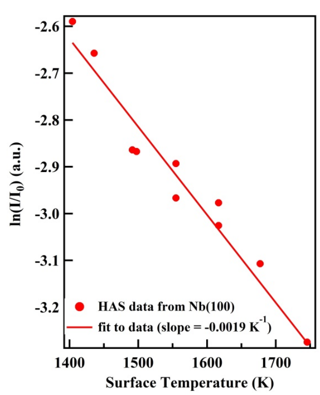

Figure 2: DW measurement on Nb(100) that gives λ of 0.61. Intensity values were taken from specular peak ($\Delta K = 0$ Å^{-1}) on diffractions scans at various surface temperatures (1400 K to 1750 K).

PROJECT UPDATE: Bonding, Diffusion, and Structure of Nb and Nb_3Sn Surfaces with Nitrogen Doping, Oxidation, Defects, Impurities and Sn or Zr Alloying

Sibener students and theme postdoc: Caleb Thompson (graduate student), Michael Van Duinen (graduate student)

Our ongoing Year 7 research in the Sibener during CBB Year 7 contributed to CBB Objective 1 of Beam Acceleration within CBB Phase II: Advanced SRF Materials Growth: Developing improved growth methods and understanding the impact of realistic (non-ideal) surfaces on performance. This project is focused on Deliverable 1.1: Methods of growth and their refinement leading to annually significantly enhanced performance for Nb3Sn on Nb and Cu substrates, with potential to branch into Deliverable 1.2, methods of growth and their refinement leading to annually significantly enhanced performance for Nb-Zr alloys, in the near future.

This project reflects work on the project titled "Bonding, Diffusion, and Structure of Nb and Nb_3Sn Surfaces with Nitrogen Doping, Oxidation, Defects, Impurities and Sn or Zr Alloying." Developing and improving SRF cavity material surfaces requires a precise, fundamental, atomic-scale chemical understanding of key driving forces for the formation of surface structures, their defects, and resulting film quality for a variety of preparation conditions. Understanding the structure, bonding, and dynamics of these material surfaces would enable SRF cavity surfaces by design. We must first access the chemical characteristics of the niobium substrate upon which these materials are built, and then from there we can obtain a better understanding of the chemical processes that drive the development of well-ordered, homogeneous new materials.

Helium atom scattering (HAS) has the unique capability to access structure, bonding, and dynamics *in situ* at high temperatures relevant to SRF cavity preparation conditions. We choose the Nb(100) face as our system of study for its periodic and characteristic (3x1)-O reconstruction. Single-crystal diffraction of the Nb(100) surface can reveal *in situ* high temperature atomic-scale periodic structure, with induced defects, and after N, Sn, or Zr doping and alloying. For example, high-temperature *in situ* diffraction data has revealed the persistence of the (3x1)-O reconstruction at temperatures as high as 1130 K. Time-of-flight (TOF) measurements on phonons through inelastic scattering can reveal information about surface interatomic forces that drive or hinder the adsorption, incorporation, and development of new materials at SRF cavity-relevant surfaces. Inelastic scattering studies also allow us to understand the strength and stability of these interatomic forces after doping or alloying processes. For example, phonon data on the (3x1)-O reconstruction mapped onto DFT data by Michelle Kelley of the Arias group comparing surface phonons of the bare Nb(100) surface with the (3x1)-O reconstruction demonstrate that the (3x1)-O ladder crests contain strengthened Nb-O and Nb-Nb bonds, corroborating the high-temperature stability of the (3x1)-O reconstruction. Combining diffraction and TOF capabilities, quasi-elastic scattering can allow us to access surface diffusion rates and mechanisms to better understand the interaction of SRF cavity-relevant surfaces after N, Sn, or Zr doping and alloying formation processes.

Within the past year, we have made adjustments to our electron beam heater that have expanded our temperature range by 400 K and allowed us to access the bare, oxide-free

Nb(100) surface. Expanding our temperature range will allow us to experiment with a wider range of SRF cavity preparation conditions. Studies on structure, bonding, and dynamics before and after doping and alloy formation processes on the Nb(100) will allow us to better understand the effects of the (3x1)-O reconstruction on these processes in SRF cavity preparation. Thus far, we have taken diffraction scans on the bare Nb(100) surface at a wide variety of temperatures to find a Debye-Waller factor, as seen in Figure 1. There is no diffuse scattering background under the specular peak in these diffraction scans, indicating an absence of diffusion at the surface. We also find that the full-width half maximum (FWHM) of the specular peak does not change with temperature, indicating that the coherence length and domain size do not change with temperature. We proceeded to find this Debye-Waller factor for a variety of incident beam energies (Figure 2). This data can be used to find that the slope of the relationship between mean-squared displacement and temperature is 1.22×10^{-5} Å^2 K^{-1} for atoms at the Nb(100) surface. Knowing the mean-squared displacement can elucidate the dynamics at the surface and how readily a given surface may incorporate new materials. We were also able to find a surface Debye temperature of 296 K for the Nb(100) surface, providing insight into surface phonon modes.

The single-crystal Nb(100) surface is a fundamental and significant starting point towards getting a full understanding of the chemical characteristics of polycrystalline SRF cavity material surfaces. High-temperature, *in situ* studies at the single-crystal face provide atomic-scale chemical information that can be used to better understand the driving forces behind macroscopic phenomena in the formation of new SRF cavity material surfaces.

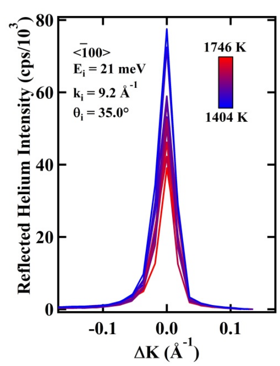

Figure 1: Attenuation of the specular diffraction peak with temperature. Unchanging FWHM of the specular peak demonstrates that the coherence length of the Nb(100) surface does not change with temperature. Also, the diffraction scan shows no diffuse scattering background, demonstrating that there is not diffusion at the surface.

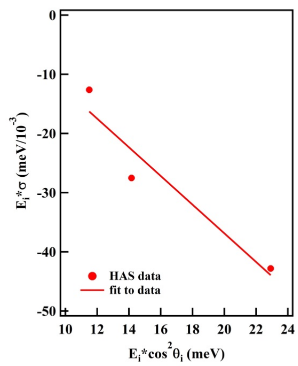

Figure 2: Debye-Waller slope for three different beam energies. The slope of this line leads to the determination of the Debye temperature (296 K) and mean squared displacement ($1.22*10^{-5}$ Å^2 K^{-1}).

Equipped with a strong understanding of the Nb(100) substrate upon which new SRF cavity materials are built, we hope to get a better understanding of the Nb(100) surface during and after doping and alloy formation processes. We are looking towards doping our Nb(100) surface with N and alloying it with Sn or Zr. Much remains to be known about the structure, bonding, and dynamics of SRF-cavity-relevant niobium surfaces doped with N or alloyed with Sn or Zr. We are equipped to resolve the atomic-scale structure of doped surfaces and alloys after formation and even during; the ability to make high-temperature *in situ* measurements allows us to directly observe the development of new reconstructions at the Nb(100) surface. We can also use inelastic TOF measurements to better understand the effect of doping and alloy formation processes on interatomic forces and their relative strengths. Lastly, we can use quasi-elastic scattering to elucidate the diffusion mechanisms that develop these material surfaces. Building these doped and alloyed material surfaces on a single-crystal will provide a starting point for understanding the driving forces and conditions necessary to design and develop homogeneous and high-performing materials for the next generation of SRF cavities.

Transtrum, Mark (Brigham Young University)

Publications

Papers

N. S. Sitaraman, Z. Sun, B. Francis, A. C. Hire, T. Oseroff, Z. Baraissov, T. A. Arias, R. Hennig, M. U. Liepe, D. A. Muller, and M. K. Transtrum, “Theory of Nb-Zr Alloy Superconductivity and First Experimental Demonstration for Superconducting Radio-Frequency Cavity Applications,” Aug. 2022, doi: 10.48550/arXiv.2208.10678. [Online]. Available: http://arxiv.org/abs/2208.10678

Talks

Harbick, A. V., Transtrum, M. K. (2023, March 7). *Simulation of the Field Enhancement Effect in*

Type II Superconductors for SRF applications. APS March Meeting 2023, Las Vegas, NV, United States.

Awards

Mark Transtrum, Edward and Betty Seppi Endowed Chair of Physical and Mathematical Sciences. (BYU College of Physical and Mathematical Sciences)

Collaborating Groups

1. Cornell University: Tomás Arias, Matthias Liepe, David Muller
2. University of Florida: Richard Hennig
3. FNAL: Sam Posen

PROJECT UPDATE: Time-Dependent Ginzburg-Landau Studies of Realistic Materials and Surfaces

Transtrum students: Aiden Harbick (graduate student), Eva Guevara (undergraduate)

Efforts in the past year have been devoted to improving to our group's existing TDGL modeling capabilities. In particular, we have improved our ability to calculate dissipation in superconducting systems as well as implementing 3D simulation capabilities. These improvements represent significant advances that have improved our ability to make predictions about superconducting systems relevant to SRF cavity development. We expect these tools to continue to guide CBB SRF research in the next few years.

One of our modeling focuses has been on 'island-like' material defects in superconducting materials. 'Islands' refers to small regions within a superconducting material that have a different chemical composition from the bulk material. Our island modeling has included 'hydrides' in Nb, where the island will be a region of NbH within the Nb bulk material or 'Sn-deficient islands' where the island is a Sn-depleted region within a Nb3Sn bulk material. Our results suggest that the mechanism of HFQS in Nb cavities comes from the dissipation due to hydride-induced magnetic vortices. This is a significant result as the mechanism for this effect has not yet been fully understood. These results also have implications for Sn-deficient islands in Nb3Sn. While the relevant length scales of the two materials are different, the qualitative picture remains very similar. These models are guided by close collaboration with the Liepe group (whose SRF cavity composition we attempt to model) and Arias and Hennig groups (who supply superconducting parameters from *ab initio* calculations)

Our other focus of the past year has been modeling field enhancement effects for rough superconducting surfaces. We have successfully achieved simulations of the field enhancement effect using a two-domain TDGL formulation in COMSOL Multiphysics software. Simulations are still ongoing.

PROJECT UPDATE: Superheating field for multi-layers and inhomogeneous layers

Transtrum students: Ben Francis (postdoc)

During this reporting period, this project only lasted a few months and concluded in August 2022. During this time, we co-authored a paper on niobium-zirconium (Nb-Zr) alloys, a material that our work previously indicated would be a good candidate material for SRF cavities. (See Figure below), which shows the predicted improvement in H_{sh} as Zr is added to Nb.) Together with the Liepe, Arias, Hennig, and Muller groups, we used both theoretical predictions and experimental results to show that improvement in several metrics could be expected by using Nb-Zr in cavities instead of just Nb. This paper has been submitted to Physical Review Applied and is currently under review.