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Project objectives and summary of results

The objective of the proposal was to develop graduate student training in materials and engineering research relevant to the development of particle accelerators.

Many components used in today's accelerators or storage rings are at the limit of performance. The path forward in many cases requires the development of new materials or fabrication techniques, or a novel engineering approach. Often, accelerator-based laboratories find it difficult to get top-level engineers or materials experts with the motivation to work on these problems. The three years of funding provided by this grant was used to support development of accelerator components through a multidisciplinary approach that cut across the disciplinary boundaries of accelerator physics, materials science, and surface chemistry.

The following results were achieved: (1) significant scientific results on fabrication of novel photocathodes, (2) application of surface science and superconducting materials expertise to accelerator problems through faculty involvement, (3) development of instrumentation for fabrication and characterization of materials for accelerator components, (4) student involvement with problems at the interface of material science and accelerator physics.

Physics results from fabrication of novel photocathodes

It was verified that emission properties of photocathodes can be manipulated through the engineering of the surface electronic structure[1]. An ultrathin multilayered structure of MgO/Ag(001)/MgO films was chosen for testing due to the availability of a theoretical prediction that surface band structure can be modified with MgO layer thickness[6]. The surface band deformation affects the Quantum Efficiency (QE), work function, and emittance characteristics of the photoemissive surface. These multilayered structures were grown by pulsed laser deposition, tuning the thickness, n, of the flanking MgO layers to 0, 2, 3, and 4 monolayers. We observed a systematic increase in quantum efficiency and simultaneous decrease in work function with layer thickness, as shown in Figure 1. The workfunction of the multilayer structures versus MgO layer thickness is shown in Figure 2 for two different crystal orientations. The results for the (001) orientation are compared with theoretical (Density Functional Theory) calculations. The scale and trend direction of measurements are in good but not excellent agreement with theory. Deviations from theoretical predictions are attributed to imperfections of real surfaces in contrast with the ideal surfaces of the calculation. For example, it is difficult to get a uniform surface for films that are only one or two monolayers thick. The surface bands of the photoemitter are modified to a degree that is dependent on the thickness of the flanking MgO layers. It is possible to change the range in surface parallel momentum of the electrons in the band closest to the Fermi surface within the material[3,6]. Since the surface parallel momentum is conserved in the absence of significant scattering, the electrons emitted from this band would have a narrower angular spread, and so the beam would have a smaller emittance. This good effect relies on emitting only those electrons in the band nearest the Fermi surface, and would thus require a light source with an appropriately restricted energy range, so as not

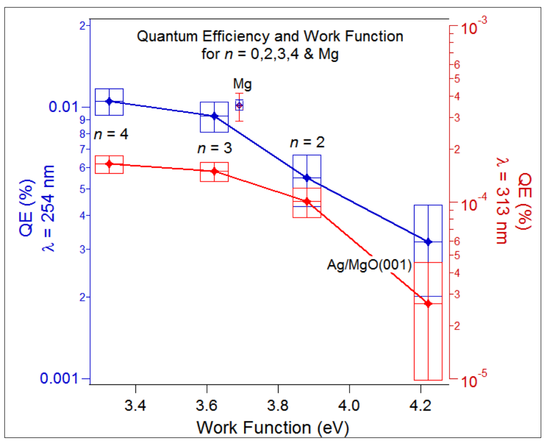


Figure 1: Workfunction and Quantum Efficiency (QE) of MgO/Ag/MgO as a function of flanking layer thickness.

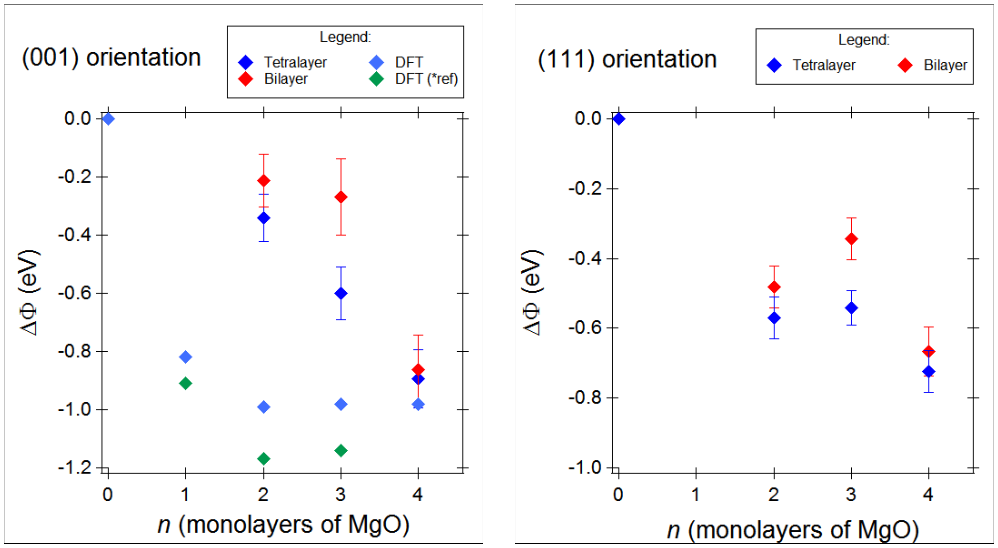


Figure 2: Workfunction versus layer thickness for MgO/Ag/MgO structures shown for two different crystal orientations (001, 111). Data for the (001) orientation are compared to Density Functional Theory (DFT) calculations. The comparitively gradual change in the experimental data is perhaps due to the inability to get an absolutely smooth surface coating for deposition of very thin layers.

to excite electrons in bands further from the Fermi surface. Angle Resolved PhotoEmission Spectroscopy (ARPES) is a measurement technique that gives information about photoelectron flux and energies versus angle, and can thus be used to measure emittance and also reconstruct the surface band structure of the material[3]. Since IIT does not have easy access to an ARPES system, we made only one measurement with a now decommissioned system at Brookhaven National Laboratory. Angle resolved photoemission data (shown in Figure 3) for the multilayered sample $n = 3$ shows that the emission profile has been modified from that of bare silver in a way consistent with the DFT calculations. The DFT results are shown for bare silver and for the MgO multilayer, along with corresponding measured data. While the theoretical and experimental results are not precisely aligned, the observed deformation of the band is consistent with the theoretical deformation. Considering that it was only possible to do a single measurement, these results fuel our optimism that we can design surfaces to have specific emission properties.

It was necessary to go through an extensive learning curve to gain precision control over our samples. The fabricated surfaces were of single crystal (epitaxial) layers of stringently controlled thickness. We found that we could also control the surface roughness of the fabricated samples. This opens the door to future studies that compare models how surface roughness

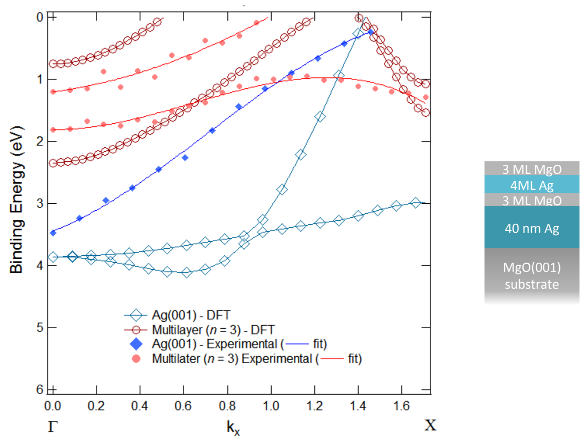
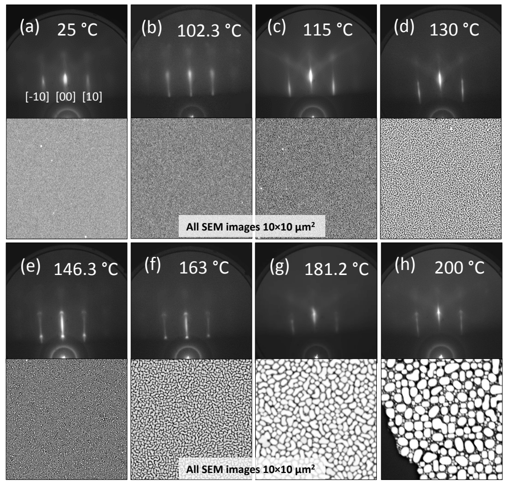


Figure 3: Comparison of theory (DFT calculation) to data (ARPES) on the bandstructure of multilayer (001) MgO/Ag/MgO structure with n=3 monolayers of flanking MgO.

affects emission with experimental data. Figure 4 shows silver films with systematically controlled roughness.

The experimental work required to get these results was supported both by this grant and NSF funding, and would not have been possible without both sources. The NSF grant supported the graduate student Daniel Veláźquez, while the DOE funds supported research faculty member Dr. Zikri Yusof. Instrumentation commissioning (PLD, Kelvin Probe, and QE measurement system) was carried out under supervision of Dr. Yusof. Graduate student training on these systems (and other instrumentation) was carried out by Dr. Yusof. Dr. Yusof also participated in discussion and supervision of measurement results.

This grant supported summer students Chaoyue Cao and Mark Warren, advised by Professor John Zasadzinski, an expert in superconducting materials. Mark was initially researching breakdown in SRF cavities; for example, he performed Raman spectroscopy studies on Nb foils to study how various treatments may affect the impurity content of the surface. Since then he has shifted his thesis topic to the development of a novel high QE superconducting photocathode. Dr. Zasadzinski's expertise is the perfect compliment to the photocathode research. A high QE superconducting photocathode is the final piece of an entirely superconducting radiofrequency (SRF) linear electron accelerator that includes SRF accelerating cavities and photoinjector. Achieving high pulse rates will likely require a superconducting photoinjector and photocathode with high quantum efficiency (QE). A new approach to achieving a high QE superconducting photocathode (funded by a new DOE grant) will be taken by exploiting what is known as the proximity effect, a phenomenon that occurs when thin layers of different materials are fabricated into heterostructures. These are bilayers or multilayers that take advantage of high QE materials, along with QE enhancement from a dipolar layer, and merge them with superconductors.



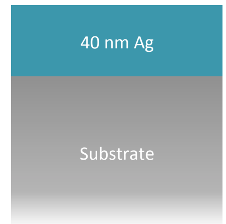


Figure 4: Diffraction pattern (above) and 10X10 μm^2 SEM images (below) of silver films fabricated with PLD. These show the surface roughness varying systematically with substrate surface temperature.

Development of instrumentation and material science collaboration

This grant promoted collaboration between Prof. Spentzouris (accelerator physicist), Prof. Zasadzinski (material science expert in superconducting materials, including state-of-the art thin film deposition methods, and in use of sophisticated probes of surface superconductivity), and Prof. Terry (trained as a physical chemist with a specialization in a variety of analytical techniques to examine surface chemistry). Dr. Yusof, who was funded by this grant, worked with these professors, graduate students, and undergraduate students, making a cohesive group of researchers. He was intimately involved with the commissioning of the Pulsed Laser Deposition (PLD) system, the Kelvin Probe for measuring the work function of surfaces, and the Quantum Efficiency measurement system. He also trained students on other characterization instrumentation such as the FTIR UV/VIS (Fourier Transform Infrared Ultraviolet/Visible) spectroscopy system used for absorption or reflection spectroscopic measurements on materials, Raman spectroscopy, Atomic Force Microscopy (AFM) and a small XPS (X-ray Photoelectron Spectroscopy system). In addition, he supervised summer students, and collaborated on research related to the physics goals of the group.

Pulsed Laser Deposition

Pulsed Laser Deposition (PLD) is an ultra-high vacuum (UHV) epitaxial thin film deposition technique in which a high energy density laser beam is focused and pulsed onto a target. The system which resides in Prof. Terry's lab is shown in action in Figure 5. Upon absorption of

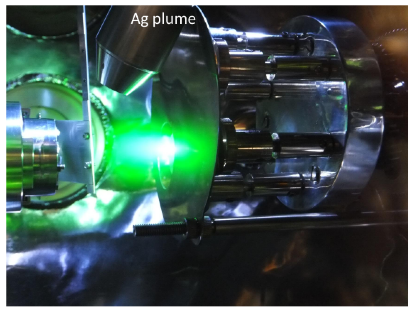
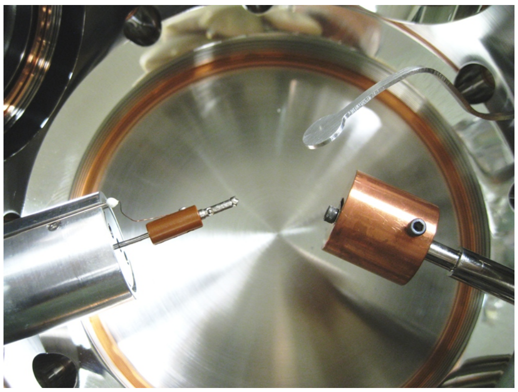


Figure 5: Pulsed laser deposition of silver.

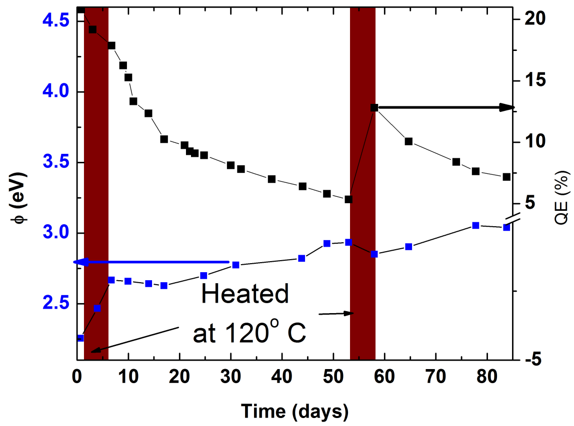
the pulse, the target undergoes a process in which material from the surface of the target is vaporized and formed into a plasma plume. After ablation the plume material is deposited on a heated sample facing the target. Multiple targets can be deployed without breaking vacuum, so layered structures with different materials in each layer can be fabricated. The in situ RHEED diffraction system is used to verify the crystal structure of deposited materials. Layer thickness is measured with calibrated thickness monitor, and can be controlled at the monolayer level. The temperature of the sample is controlled by laser heating from a second laser and can be used to control the surface roughness of the finished film. Dr. Yusof and graduate student Daniel Velazquez commissioned the PLD system with a new laser. They got the vacuum system operational, dealt with laser optics, and calibrated the RHEED diffraction system (Reflection High Energy Electron Diffraction).

The Kelvin Probe and Quantum Efficiency Measurement system

Dr. Yusof and graduate students assembled, commissioned and deployed a Kelvin Probe system to measure the work function of photocathode materials. The Kelvin Probe was provided by collaborator Dr. Kathy Harkay, and is located at the Argonne Wakefield Accelerator (AWA) facility. Prolonged studies of the behavior of the AWA Cs_2Te photocathodes were done soon after commissioning, including behavior changes during aging, exposure to UV light, and when undergoing a heating process known as rejuvenation. Data from these studies is shown in Figure 6(b).



(a) Kelvin Probe tip



(b) Work function and QE of Cs_2Te

Figure 6: Left: View of the Kelvin probe system, showing sample mount (bottom right), Kelvin probe tip (bottom left) and collection anode (top right). Right: Measured work function and quantum efficiency of AWA Cs2Te photocathodes versus time, while photocathode was still in the deposition chamber. The first shaded band is during the period of rapid change at initial cathode fabrication. The second shaded band covers the time when the cathode was rejuvenated. Note that the quantum efficiency is still around 7% after 80 days.

These studies resulted in a change in the preparation procedure of the cathodes used in the AWA accelerator. The new procedure has resulted in photocathodes that age better in the high field gun environment. Besides IIT graduate students, Euclid Techlabs (an SBIR company) has also been using the Kelvin probe for material studies. Dr. Yusof further upgraded the Kelvin probe system to add Quantum Efficiency (QE) measurement capability. This was necessary for getting the MgO/Ag/MgO multilayer data shown earlier, and has also been used for initial tests for the development of a high QE superconducting photocathode.

Other equipment provided by the AWA facility includes a breakdown gun. This is a dedicated gun for testing cathodes, which can run at high field, up to 100 MV/m. Our collaborators at AWA are investigating the possibility of routing the laser to the breakdown gun to enable characterization of beams from photocathodes.

Summer students supported by this grant

Summer students supported by this grant include Teng Shen, Mingyu Xu, Medani Sangroula, Chaoyue Cao, Mark Warren and Anthony Ruth.

- Mark Warren is a graduate student doing his thesis work on high QE superconducting photocathodes, with John Zasadzinski as his advisor, and Linda Spentzouris as a collaborator.
- Chaoyue Cao, whose advisor was John Zasadzinski, graduated in 2014 with a dissertation entitled 'Surface characterization of niobium for superconducting RF cavities'.
- Medani Sangroula is a graduate student doing his thesis on Impedance measurement of the vacuum chamber components for the APS upgrade at Argonne National Laboratory. His Argonne advisor is Ryan Lindberg, and his IIT advisor is Carlo Segre.
- Teng Shen and Mingyu Xu were Master's students at IIT who spent the summer assembling, commissioning and testing a TiN coating rig at Fermilab. The rig was then used to make samples for the Secondary Electron Yield (SEY) test stand for studies of SEY conditioning by the beam. Once the rig was commissioned, they characterized some TiN samples at IIT using XPS and also imaged the surface.
- Anthony Ruth was an undergraduate student at IIT who worked with collaborator Karoly Nemeth and learned to do DFT calculations and investigated a novel photocathode material. The results of this study were published[5].

Summary

Support from this grant enabled students to be trained at the interface of material science and accelerator physics. Several of those students have gone on to do their thesis work at this junction. The funds from this grant enabled a cohesive effort to induct material science techniques to the development of accelerator components. The scientific results are primarily in the area of photocathode development, where it has been shown that it is possible to engineer photoemissive surfaces to improve photocathode figures of merit. The research carried out under this grant has continued. Research going forward will be on development of a high QE superconducting photocathode; work with Euclid Techlabs on the development of a functionalized plasmonic cathode; and continued work on thin layered structures, in particular, studies to determine their survivability at high field.

Relevant publications

(1) Tailoring the emissive properties of photocathodes through materials engineering: ultrathin multilayers, Daniel Velízquez, Rachel Seibert, Hasitha Ganegoda, Daniel Olive, Amy Rice, Kevin Logan, Zikri Yusof, Jeff Terry and Linda Spentzouris, Applied Surface Science, **360**, p. 762-766, (2016).

(2) Pulsed laser deposition of single layer, hexagonal Boron Nitride (white graphene, h-BN) on fiber-oriented Ag(111)/SrTiO3(001), Daniel Vel*acutea*zquez, Rachel Seibert, Hamdi Man, Linda Spentzouris, and Jeff Terry, Journal Applied Physics, **119**, 9 095306, (2016).

(3) A Metal-insulator photocathode heterojunction for directed electron emission, Timothy C. Droubay, Scott A. Chambers, Alan G. Joly, Wayne P. Hess, Karoly Németh, Katherine Harkay, and Linda Spentzouris, Phys. Rev. Lett., **112**, 067601, (2014).

(4) "Kelvin probe studies of Cesium Telluride Photocathode for AWA photoinjector", Eric Wisniewski, Daniel Velazquez, Zikri Yusof, Linda Spentzouris, Jeff Terry, Tapash J. Sarkar, and Katherine Harkay, *Nucl. Instr. Meth. A*, **711**, pp.60-64, (2013)

(5) "Searching for low-workfunction phases in the Cs-Te system: the case of Cs2Te5", Anthony Ruth, Károly Németh, Katherine C. Harkay, Joseph Z. Terdik, Linda Spentzouris, Jeff Terry, *J. Appl. Phys.*, **113**, 183703, (2013)

(6) High brightness photocathodes through ultra-thin surface layers on metals, Károly Németh, Katherine C. Harkay, Michel van Veenendaal, Linda Spentzouris, Marion White, Klaus Attenkofer, and George Srajer, Phys. Rev. Lett., **104**, 046801 (2010).