Cathode R&D for Future Light Sources

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Abstract

This paper reviews the requirements and current status of cathodes for accelerator applications, and proposes a research and development plan for advancing cathode technology. Accelerator cathodes need to have long operational lifetimes and produce electron beams with a very low emittance. The two principal emission processes to be considered are thermionic and photoemission with the photocathodes being further subdivided into metal and semi-conductors. Field emission cathodes are not included in this analysis. The thermal emittance is derived and the formulas used to compare the various cathode materials. To date, there is no cathode which provides all the requirements needed for the proposed future light sources. Therefore a three part research plan is described to develop cathodes for these future light source applications.

keywords: cathode research; electron source; photocathodes; quantum efficiency; thermal emittance; high average current; Energy Recovery Linacs

Introduction

The development of the photocathode gun has become a significant enabling technology for x-ray free electron lasers and other 4th generation light sources. As the first x-ray FEL user facility, the performance of LCLS is impressive, lasing 10-orders of magnitude higher in peak energy than previous x-ray light sources [1]. And there are opportunities for improving even this performance. The emission processes of the cathodes used in the LCLS gun are not completely understood. The quantum efficiency needs to be made reliable and the low-charge, thermal emittance is nearly a factor of two higher than given by theory. In addition, it operates at a low repetition rate (120 Hz) and it is anticipated that future applications will require repetition rates of 100 kHz and higher with CW operation. Therefore a principal technical challenge for ERL's as well as for other high repetition rate light sources will be the production of LCLS-like beams in a lower peak field but high average power gun producing up to 100 milliamps of

average current. The combination of high average current and ultra-low emittances required by the ERL and x-ray FEL oscillator has never been achieved in a CW gun. An area requiring significant support is photocathode R&D since there are presently no cathodes meeting the known requirements. Thus there is strong motivation for two overlapping lines of cathode R&D: one of cathodes for low-repetition rate and ultra low emittance guns like LCLS, and a second of cathodes for high-average current guns to be used in ERL's and other CW applications.

Drive Laser and Cathode Requirements

The operating range of the injector and the corresponding drive laser system can be divided in to three distinct regimes: $< 1 \mu A$, $1 \mu A - 1 mA$, and > 1 mA. The cathode and drive laser requirements are presented in Figure 1 where the average drive laser power is given vs. the cathode quantum efficiency (QE). Lines of constant average current are plotted in the log-log graph. Three shaded regions schematically show the QE range for metal, antimonide and Cs:GaAs cathode types along with the vacuum required for them to survive several hours. For the low average current injectors (< 1 µA), metal photocathode irradiated by UV laser provide ultra high brightness beams as evidenced by the LCLS. Properly conditioned metal photocathodes such as Mg or Pb, along with a few watts of UV would be able to service the injector in the current range of 1 μA – 1 mA, delivering peak brightness comparable to LCLS. For high average current injectors, in order to make the drive laser practical, it is necessary to limit its average power to less than approximately 25 watts, operate it at visible or near IR wavelengths and require cathodes with QE's of 1 to 10 percent. Although there has been significant progress in the high average power lasers such as diode pumped fiber lasers that deliver up to 100 W in IR, at present, these lasers are generally less reliable, and the stringent beam shaping and stability necessary to produce a bright electron beam would necessitate additional laser R&D. One to ten percent QE's can be reached using Cs:GaAs or K₂CsSb, however the reliable fabrication and operation at 100 mA in a gun has not been demonstrated and represents a major technical challenge.

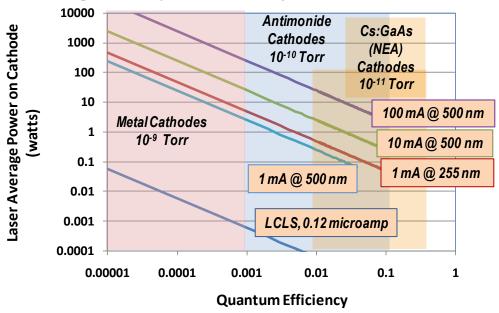


Figure 1: Plot of the average laser power vs. quantum efficiency to produce various average beam currents. The QE ranges for the general cathode types are shown along with their vacuum requirements.

Having the drive laser operate at either visible or near-IR wavelengths allows a more equitable sharing of the technical risks between the laser and cathode for high average current injectors. While cathodes at the longer wavelengths are more difficult, the laser challenges at UV wavelengths are greater. Current UV laser systems begin with an IR beam which is then frequency-tripled or -quadrupled using non-linear conversion crystals. Good conversion efficiency requires focusing the laser to a small spot in the crystal which can damage the crystal. In addition, shaping the laser pulse is difficult and inefficient at UV wavelengths as are the diagnostics. The option of frequency up conversion followed by power amplification is viable at specific wavelengths, but has not yet been fully investigated. In this approach, the beam shaping can be accomplished at lower average power UV beam that is subsequently amplified. In this scheme, the losses due to the shaping can be compensated by the amplification. The major drawbacks of this path is the UV gain media are limited to specific wavelengths such as 248 nm and the repetition rate is limited to < 10 kHz. With the current laser technology, the maximum obtainable UV average power is ~ 2 W resulting in a maximum obtainable average current with a metal photocathode to hundreds of microamperes. UV-beam and enhanced metal or CsTe cathodes with ~1-10% QE are viable alternatives for a few mA-tens of mA injectors, especially since excimer lasers operating at multi kHz can be very efficient amplifiers, having very large bandwidth and gain. Other laser options are on the horizon such as diode-pumped, cryogenically-cooled Yb:YAG, which show promise for scaling to kilowatt average power with the required stability and beam quality. Due to the superior quantum efficiency (91%) of Yb:YAG, and the possibility of extracting the full stored energy of the crystal at liquid nitrogen temperature while maintaining the high beam quality, this source can develop average optical power of hundreds of watts using about 3 kW electrical wall-plug power while maintaining the ultra short pulse duration.

Additional complications of using the UV radiation stem from transport optics. Typically, the AR coating of UV lenses and dielectric coating of mirrors are also much more susceptible to damage. For example, the vacuum windows on the laser transport tube for LCLS were prone to damage even at the low average fluencies at LCLS, forcing a re-design of the optics to increase the beam size on the windows to eliminate the damage. In general, UV optics are more sensitive to laser damage and are less efficient requiring an even higher power laser to make up for the losses. The additional laser power can be significant. Optical damage thresholds in these applications are limited typically by the peak power and not the average power. However, since most of the laser transport line would be in vacuum to minimize the beam fluctuation, the damage threshold and air absorption can be minimal.

The desired characteristics for the drive laser are sub-ps stability, micron level position stability, uniform transverse and longitudinal beam profiles are required for cathodes and gun types. There is one distinct difference between low and high average current systems and that is the amount of allowed photocurrent outside a nominal temporal and spatial window of the laser. For example, in a typical RF gun, laser light reaching the cathode +/-20 degRF or more away from the nominal laser-RF phase will produce off energy and different trajectories, potentially producing beam halo with significant average power for a high average current system. Laser-related beam halo is also produced by scattered laser light striking the cathode either at the wrong time or wrong location. The allowed laser-related beam halo is typically 1 part in 10^{-6} of the total beam current.

The desired laser parameters at low average current can be achieved through the use of diodepumping of the Titanium-Sapphire laser medium [2]. This type of laser system is also very reliable, operating for 18 months with better than 97% uptime. However one technical issue deserving further research is temporal pulse shaping. The desired three dimensional pulse shape has been difficult to attain even with state-of-the-art techniques.

Due to this interdependence of the cathode and laser, there should be parallel laser R&D concentrating on developing reliable and stable high-repetition rate systems with the capability of pulse shaping in three-dimensions. As noted above, the cathode drive laser is expected to operate at visible to near IR wavelengths and should have limited bandwidth to minimize the production of electron beam instabilities, unless the drive laser is also being used to seed the FEL. Laser pulse shaping allows preforming of the electron bunch to maintain linear space charge forces and to manipulate space charge waves.

Beyond the high-QE and its survival in a gun environment, the cathode has to emit a beam of exceptional quality with very little dark current. Recent results for the LCLS gun and the Cornell injector show significant progress in the practical applications of emittance compensation to control space charge effects, and the generation of near perfect RF and magnetic fields to eliminate optical aberrations. Combined with the overall advantages of operating at lower bunch charge, we are now reaching the thermal emittance as the limit to increasing the beam brightness. However, the effective thermal, or "intrinsic", emittance depends on several effects, including the crystallinity, surface roughness, surface impurities and QE non-uniformity. Thus it is very challenging to measure and combine all these phenomena into a complete and useful physical model. For example, it is relevant to note that the measured thermal emittance from the LCLS gun is nearly twice the theoretical value [3, 4] and the source of this difference is not understood. If the thermal emittance had the theoretical value, the already excellent LCLS emittance would be 20% lower still. Therefore the second major challenge for cathodes is to understand the origins of the low charge emittance and its interaction with the space charge forces.

The Three Part Cathode R&D Plan

In order to address these and other cathode related issues we suggest the following R&D program consisting of three interrelated parts:

- 1. Studies of optimal cathode formation methods and cathode emission characteristics, using available surface and material diagnostics
- 2. Modeling of cathode emission physics and electron dynamics near the cathode
- 3. Operational testing in the gun and injector system and validating models.

1. Studies of optimal cathode formation methods and cathode emission characteristics, using available surface and material diagnostics

Optimal performance of a cathode can only be achieved with understanding of the material properties, such as surface and bulk crystallinity, band structure, surface morphology, material optical properties and surface chemistry. Such understanding will provide feedback to allow optimization of growth and processing of cathodes, and will provide performance data that will be used to validate modeling codes and ultimately predict cathode behavior. These techniques should supplement a program including direct measures of cathode performance, such as spectral response, lifetime (both dark and operational) and sensitivity to chemical poisoning by gasses typically found in an injector. With the advent of modern user facilities (principally light sources and nanocenters), techniques such as diffraction, photoemission spectroscopy and high resolution imaging are available to explore these material properties.

New growth methods should be investigated for the creation of accelerator cathodes, such as atomic layer deposition and molecular beam epitaxy (MBE is already used for GaAs cathodes). For the longer-term, advances in the synthesis of novel materials, nano-engineering in particular, raises the possibility of designing photocathode materials optimized for specific properties [5] after validation of the design tools based on data from existing cathodes. Such cathodes should be studied using analytic tools such as Density Functional Theory (DFT) analysis [6] with promising candidates being fabricated and characterized [7].

2. Modeling of cathode emission physics and electron dynamics near the cathode

The fine details of the emission process need to be included to the electron simulation codes. This should include the physics of the emission process, using models such as Spicer's three step model of photoemission [8] or the exact one-step model [9]. The results of these models should be used to predict thermal emittance values based on full energy and angular distribution curves of the emitted electrons. Given the improvements made in RF and magnetic optics, and emittance compensation, the next step is for the computational dynamicists to put the physics learned in the cathode labs into the particle codes. These enhanced codes can then be used to simulate and design the complete injector and be verified in the gun and injector studies part of the R&D plan. Genetic algorithms have already been employed is some areas of beamline design [10]; a program to integrate this capability into modeling codes along with a complete emission physics package could lead to much better optimization of cathode, gun and laser properties.

3. Operational testing in the gun and injector system.

It is of course essential that these lab results and computer studies be tested in a operating gun. Some experiments can be performed in a low duty factor system but will also require testing in a CW gun. Among the current photocathode guns being proposed: DC, NCRF and SCRF, all are viable platforms for cathode testing since each has it's own application niche. Some tests of cathode performance, such as thermal emittance, operational lifetime and response time, are best made in an injector.

Cathode Technology

The semiconductor photocathodes in wide use today as high-brightness electron sources for accelerators derive in large part from cathode R&D performed decades ago. Much of the development work on photocathodes was focused on photoemissive detectors, where the most important criteria are quantum efficiency, reliability, low dark current, spectral response, and response time. The emission distribution, or emittance, was not a high priority, and thus these cathodes were not optimized for ultrahigh brightness. In order to meet the requirements for future light sources, a new wave of R&D is needed with collaboration among accelerator physicists and materials and surface scientists.

Thermionic cathodes can deliver thousands of coulombs reliably and have been used in several FELs around the world. To reduce their large thermal emittance though, grid structures must be eliminated, making pulse generation difficult. To offset that problem and to increase the peak charge density that can be extracted from the cathode, a pulsed DC structure is used at SCSS with a CeB₆ cathode. The interesting approach of Spring8 SCSS FEL is to use pulsed HV which can more easily reach 500 kV, use a single crystal thermionic cathode and modulate the beam energy at high frequency with a RF cavity and slice out short bunches using energy slits in a magnetic chicane. The CeB₆ gun has been very reliable and successfully delivered stable 500-keV beams to the SCSS test accelerator for three years, and is now operating for various EUV-FEL user experiments [11].

Metal photocathodes are predominately used in high gradient RF guns, in particular the s-band BNL/SLAC/UCLA gun. Improvements in this gun design were incorporated into the LCLS gun to produce the high brightness beam for LCLS [12]. The relatively low thermal emittance of metals contributes to the low gun emittance, especially at low charge. Metal cathodes are more tolerant to vacuum contamination and unlike other photocathodes can be transferred and installed at atmospheric pressures and thus do not require a load-lock. The disadvantage of the metal cathode is its low QE and need for a UV drive laser, which limits these cathodes to applications requiring \approx 1mA average current. Fortunately recent advances in laser technology have greatly improved the reliability of these lasers through the use of diode pumping of the gain medium. Therefore, while costly, fully integrated laser systems are commercially available.

Photoemission and thermionic cathodes are currently being used in ERL-based FELs. The Energy Recovery Linac-based FELs at Jefferson Laboratory in the U.S. and at Daresbury Laboratory in the U.K. use a Cs:GaAs photocathode in a DC gun illuminated with laser pulses at ~532 nm [13, 14], while the BINP FEL/ERL and the HEPL Recyclotron used a thermionic cathode [15,16]. To date, no other type of cathode has delivered beam for an ERL-based machine. The JLab FEL DC gun delivered over 900 hours and 7000 Coulombs at 2-9 mA CW from a single GaAs wafer between 2004 and 2007 with a lifetime of 550 Coulombs or 30 hours at an average current of 5 mA CW [17]. In 1992 the Boeing normal conducting RF (NCRF) gun demonstrated 32 mA with a K₂CsSb photocathode and still holds the record for the highest average current from a photo cathode gun [18]. Cs₂Te cathodes have been in operation for 120 continuous days in a normal conducting RF gun at PITZ with minimal QE degradation. [19].

Materials Science Modelling and Analysis of Cathodes

Numerous materials analysis tools are available to assist in cathode development. These can be broken into three broad classes – those that analyze the structure of the cathode, those that analyze the chemical makeup of the cathode and any contaminants and those that evaluate its function as an electron emitter. For structural analysis, surface imaging techniques such as atomic force microscopy and scanning electron microscopy (SEM) provide surface roughness values; the SEM can also provide spatially imaged chemical data via energy dispersive X-ray spectroscopy (EDS) and crystalline makeup of the cathode via electron backscattered diffraction (EBSD). Other electron diffraction techniques provide surface crystalline information, including local reconstruction due to surface termination (such as hydrogen on diamond). X-ray diffraction (XRD) is capable of providing crystalline information on both the surface and the bulk (by varying angle of incidence). XRD can be used to determine the grain size of grown cathodes (alkali antimonides and tellurides), and this can in turn be used to optimize the growth parameters to improve grain size and orientation. Diffraction imaging techniques such as X-ray topography can "see" strain in crystalline cathodes, possibly providing insight into damage caused by ion bombardment in GaAs.

Chemical analysis techniques include X-ray photoemission spectroscopy (XPS), secondary ion mass spectroscopy (SIMS) and X-ray absorption spectroscopy (XAS). These techniques can provide feedback to the growth process of grown cathodes and can provide data on adsorbed contaminants on all cathodes [20]. SIMS is capable of providing a depth profile of the chemical makeup of a cathode, allowing variations in the cathode make-up to be observed. XAS and XPS are sensitive to chemical bonding in addition to elemental make-up, and can be used to distinguish similar chemical forms (K_2CsSb and KCs_2Sb , for example).

The function of a cathode as an electron emitter can be evaluated using a variety of photoemission spectroscopy techniques, including photoemission electron microscopy (PEEM) and angle-resolved photoemission spectroscopy (ARPES). The PEEM allows the emitted electrons to be spatially imaged, uncovering variation in material work function and spatial non-uniformity. ARPES provides the energy and angular distribution of the emitted electrons. Together, these tools provide the spatial, angular and energy distribution of the beam. This represents all of the data necessary to determine the initial phase space volume of the beam from the cathode – the "thermal" emittance. Density Functional Theory (DFT) is a fully quantum mechanical approach for solving the electronic structure of solid surfaces. Many contributed to the idea of using density as the basic variable for the description of the energies of electronic systems. Kohn and Sham [21] demonstrated that the electron density of a fully-interacting system could be rigorously obtained from a simple one-electron theory. Much current understanding of metal surfaces comes from using the simplest DFT approximation of plane-wave pseudopotentials within the local density approximation (LDA). Other approximations are suitable for studying strongly-correlated systems [22].

DFT analysis has been used to compute the work function of various metal crystals and the agreement with experimental values is reasonable (within 10%) [23]. The surface bands computed in this model give the highest-energy partially-occupied bands that fall below the Fermi level, and the electrons in this "Fermi pool" have a bounded surface-parallel momentum, kmax (i.e., the transverse momentum in the accelerator physics convention). The laser energy determines what fraction of the Fermi pool can be photoemitted. Most notably, the measured angular distribution of photoelectrons has been shown to correspond with the calculated k_{max} , e.g., see ref. [24]. These results suggest that DFT analysis or other analytical methods are promising tools in studying candidate ultralow emittance photocathodes, both single crystals and more complex structures. This is an area that requires R&D.

Initial investigations were made of MgO monolayers on Ag, a well-studied material in catalysis [25]. DFT computations suggest that the surface-parallel momenta in the surface band for this system are well limited, and the corresponding emittance can potentially be reduced below 0.1 mm-mrad [7,26] Furthermore, thin oxide films induce a significant change in the work function [27]. For the MgO monolayers on Ag, a reduction in the work function of >1 eV relative to Ag is both computed and observed [28]. A possible practical device based on this material or a similar principle should be developed and studied.

The Spicer 3 step model is widely used to compute the QE of a cathode and seems to do a reasonable job for normal incidence light. However, even in this case there are ambiguities that can significantly affect the results. The most important perhaps is the fact that many real cathodes are polycrystalline, but have in reality a preferred crystallographic texture. Evaporation of thin films onto non-comensurate substrates often leads to this effect; for example, Al on glass has a <111> texture with only a few degree variation from the surface normal. In the case of the LCLS Cu cathode for example, micro-XRD has shown that the surface consists of an equal mixture of 111 and 110 grains. At the LCLS injector photon energy and field, the 111 grains would emit roughly 30 times less than the 110 grains, and so modelling really ought to take into account the statistical distribution of grains and corresponding work functions. A more intrinsic deviation from the Spicer model is seen when using p-polarized light off normal incidence. Recent work on Cu(111) has shown that 0.5 eV above the work function, the p-polarized QE peaks at around 70 degrees off-normal incidence, at 14 times the normal incidence yield [29]. Earlier work on Cu showed the same effect in polycrystalline Cu [30]. The same effect has been observed in Al, and in Mo, the enhancement is around 40. In recent work on annealed and ion-damaged

Cu, the effect can clearly be associated with sharpness of the metal – vacuum interface. Such effects are qualitatively predicted from theory [31] when accurate models of the surface potential are taken into account, but so far there is no universal predictive model. The general point is that the very rapid change in electric potential from outside to inside the surface causes sufficient uncertainty in electron momentum that many more initial and final states can be coupled, thus increasing yield. We need to advance to a point where details of the electron structure, electron transport and emission are all taken into account within a self consistent framework so that these complicated phenomena can be completely understood.

The Theoretical Thermal Emittance

In order to compare the various cathode types it's necessary to first define the thermal emittance for each emission process. If the electrons from a cathode are assumed to have no correlation between location of emission and the emission angle then the normalized thermal emittance per unit beam size, ε_v/σ_v , with units of microns/mm(rms) can be written as

$$\frac{\varepsilon_n}{\sigma_x} = \frac{\sqrt{\langle p_x^2 \rangle}}{mc}$$
 [1]

Here σ_x and p_x are the rms transverse beam size and momentum, respectively. The rms momentum is obtained from the electron distributions (the electron density of states) for each of the emission processes and reflects the electronic properties for that emitter.

The electron distribution for a thermionic emitter is given by the Maxwell-Boltzmann distribution which leads to the well-known thermal emittance in terms of the electron temperature,

$$\frac{\varepsilon_{th,n}}{\sigma_x} = \sqrt{\frac{k_B T}{mc^2}}$$
 [2]

For photoemission from a metal, the electron distribution is assumed to be Fermi-Dirac distribution at zero temperature convoluted with a uniform the density of states. In this case, the emittance is given in terms of the effective work function and the photon energy [4],

$$\frac{\varepsilon_{pe,n}}{\sigma_x} = \sqrt{\frac{\hbar\omega - \phi_{eff}}{3mc^2}} \ . \tag{3}$$

The effective work function includes the effect of Schottky reduction of the barrier in the presence of an applied electric field, E_a ,

$$\phi_{eff} = \phi_W - \phi_{Schottky} = \phi_W - e \sqrt{\frac{eE_a}{4\pi\varepsilon_0}}$$
 [4]

For the comparison purposes of this paper the Schottky work function, $\phi_{Schottky}$, is assumed to be zero.

At this point it is necessary to discuss an important approximation leading to the simple form of Eqn. [3]. The emittance derivation involves angular and energy integrations constrained by the surface boundary condition conserving the transverse momentum across the cathode-vacuum boundary. Eqn. [3] is simple because the initial state electrons are assumed to be in s-wave states with an energy distribution given by the Fermi-Dirac function. The s-wave assumption gives a simple isotropic angular distribution and the Fermi-Dirac function for zero temperature electrons (a very good approximation at 300 degK) becomes the Heaviside step function. The combination leads to Eqn. [3]. However in general, the electron density of states will be more complicated, involving states with higher angular momentum such as d- and p-wave states oriented by the crystalline planes and having a structured energy distribution.

This complication is significant in non-ideal electron gas metals such as lead [32]. It is also relevant in the interpretation and conversion of the electron energy distribution curves (EDC's) measured in a laboratory surface science chamber to the normalized photoelectric (thermal) emittance. Simply measuring the energy spectrum doesn't provide enough information, therefore determining the emittance requires knowing the correlated angular-energy distributions. Hence angular resolved photoelectron spectra (ARPES) at the operating photon energies will be necessary. These same comments also apply to the emittance analysis of semiconductors.

For semiconductors it is necessary to consider prompt and delayed emission separately. In prompt emission, the emittance is assumed to be determined by the electrons' excess energy in the vacuum. For example, the excess energy (ignoring the Schottky work function) of a metal is,

$$E_{excess,metal} = \hbar\omega - \phi_W$$
 [5]

A simple semiconductor with a band gap energy of E_G and an electron affinity of E_A is shown in Figure 2. If we assume most of the excited electrons come from the valence band, $E_G + E_A$ correspond to the material work function described above and the excess energy for a semi-conductor becomes,

$$E_{\text{excess,semi}} = \hbar \omega - E_G - E_A.$$
 [6]

Thus it follows that the emittance for emission from a semi-conductor can be approximated by,

$$\frac{\varepsilon_{semi,n}}{\sigma_x} = \sqrt{\frac{\hbar\omega - E_G - E_A}{3mc^2}} \ . \tag{7}$$

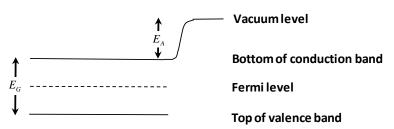


Figure 2: The energy levels of a simple semiconductor [33].

In delayed emission the excited electrons have time to thermally equilibrate with the lattice. Hence a special situation exists for delayed photoemission from semiconductor cathodes, especially negative electron affinity (NEA) cathodes such as Cs:GaAs. In these cathodes the excited electrons easily scatter with the lattice phonons, reaching thermal equilibrium with the ambient temperature phonons before escaping. Since the electrons are all thermal, the expression for thermionic emission should be used. Thus for Cs:GaAs cathodes, one should use the emittance formula for thermionic emission corresponding to the ambient temperature of the lattice,

$$\frac{\mathcal{E}_{GaAs,n}}{\sigma_{x}} = \sqrt{\frac{k_{B}T}{mc^{2}}} \ . \tag{8}$$

It is important to note that the expression for the cathode emittance given by Eqn. [8] only applies for emission with low energy photons, near 880 nm for Cs:GaAs. Emission with higher energy photons will lead to a mixture of prompt and delayed emission in which both Eqns. [7] and [8] apply. Because it is uncertain to know this mixture, for consistency Eqn. [7] will be used to compute the thermal emittance

of all semi-conductor cathodes, including GaAs, in Table III. In addition, the comments made earlier concerning how to interpret the EDC of metals in terms of the thermal emittance also apply to semi-conductor cathodes. The calculation for semi-conductors is complicated by the addition of electron-phonon scattering and the presence of the electron affinity energy level, both of which can be ignored in metallic photoemission. Electrons which leave from the bottom of the conduction band have energy with respect to vacuum equal to the magnitude of the electron affinity. Since they are likely to scatter elastically during emission, their momentum is not always normal to the surface. This effect is expected to be the primary source of thermal emittance from the diamond amplifier (see below), and it will be non-negligible for GaAs as well. The thermal emittance for photoemission using EDC's has been obtained for Cs_2Te [35] and GaAs [36].

Description of Cathode Properties

The above definitions will be used to estimate the emittances in the tables below. That is, Eqn. [2] for the thermionic emittance, Eqn. [3] for photoemission from metals, Eqn. [7] for prompt photoemission from a semiconductor.

Thermionic Cathodes

Table I gives the emission properties for CeB_6 which is used in the SCSS pulse high voltage gun. The thermionic emittance, Eqn. [2], has been used to compute the emittances from the temperature. Since a thermionic cathode naturally produces a DC beam, it is necessary to determine how long a bunch can be fit into the longitudinal acceptance of a RF cavity. The bunch length needed for a desired bunch charge, Q, is estimated by

$$t_{bunch} = \frac{Q}{4\pi\sigma_x^2 J_{thermal}} = \frac{Q}{\pi R_c^2 J_{thermal}}$$
 [9]

Using the surface current density, $J_{thermal}$, for CeB₆ and the emission size listed in Table I gives a bunch length needed for 250 pC as 84 ps. The 84 ps bunch can be sliced from the DC beam by producing an energy chirp and then sending it through a chicane to bunch and slice out the bunch with energy slits. The long bunch is then velocity compressed before injection into higher frequency accelerator sections. This is the scheme used for the SCSS FEL.

Thermionic Cathodes	Typical Temperature, $T(^{o}K)$, $k_{B}T$ (eV)	Emission Radius (mm)	Surface Current Density (A/cm²)	Work Function, $\phi_W(eV)$	Thermal Emittance (microns /mm(rms))
CeB ₆	1723K,	1.5	42	2.3	0.54
single crystal	0.15 eV				

Table I: Properties of the SCSS Thermionic Cathode

Metal Photocathodes

Metal photocathodes are commonly used in high gradient, high frequency RF guns and are the mainstay of the BNL/SLAC/UCLA s-band gun. The technological descendent of this device, the LCLS gun has produced the bright beam needed for the first x-ray FEL. Due to the high work function UV

photons are needed for reasonable QE, which makes them impractical for high average current applications such as ERLs. However, they are the most robust of all the photoemitters and can survive for years at the high cathode fields required to produce a high brightness beam. The current copper cathode installed in the LCLS gun has operated nearly continuously as the electron source for the x-ray FEL for over a year.

An interesting and significant increase in the QE occurs when the metal is coated with just a few angstroms of CsBr. Although CsBr alone has a large band gap energy, the metal acts as a reservoir of electrons which can be excited into new states formed by color centers at the interface between the two materials. Experiments show the QE can be enhanced by a factor of 50-times in copper and 350-times in niobium [37, 38]. Since the CsBr coating is a few to tens of angstroms thick, it is much thinner than the coherence length of the Cooper pairs in a superconductor. This means a CsBr coating on niobium would retain its superconductivity and one would effectively have a superconducting photocathode. This could significantly simplify present SCRF gun designs by eliminating the need for thermal isolation between a warm cathode and the cryogenic RF cavity. A QE amplification of 350-times for niobium is enough for initial testing with CW RF at low charge per bunch. Further research may lead to high QE superconducting cathodes which would greatly simplify future SCRF gun designs.

Table II: Properties of Metal Photocathodes.

The thermal emittances are computed using the listed photon and work function energies in Eqn. [3] and expresses the thermal emittance as the normalized rms emittance in microns per rms laser size in mm. The known experimental emittances are given with references.

Metal Cathodes	Wavelength& Energy: λ_{opt} (nm), $\hbar\omega(eV)$	Quantum Efficiency (electrons per photon)	Vacuum for 1000 Hr Operation (Torr)	Work Function, $\phi_W(eV)$	Thermal Emittance (microns/mm(rms))	
					Eqn. [3]	Expt.
Bare Metal						
Cu	250, 4.96	1.4x10 ⁻⁴	10 ⁻⁹	4.6 [34]	0.5	1.0±0.1 [39]
						1.2±0.2 [40]
						0.9±0.05 [3]
Mg	266, 4.66	6.4x10 ⁻⁴	10 ⁻¹⁰	3.6 [41]	0.8	0.4±0.1 [41]
Pb	250, 4.96	6.9x10 ⁻⁴	10 ⁻⁹	4.0 [34]	0.8	?
Nb	250, 4.96	~2 10 ⁻⁵	10 ⁻¹⁰	4.38 [34]	0.6	?
Coated Metal						
CsBr:Cu	250, 4.96	7x10 ⁻³	10 ⁻⁹	~2.5	?	?
CsBr:Nb	250, 4.96	7x10 ⁻³	10 ⁻⁹	~2.5	?	?

Semiconductor Cathodes

Table III lists properties of many of the known semi-conductor materials which are possible candidates for study in the cathode R&D plan. In all cases the thermal emittances have been computed using Eqn. [7] combined with the photon, gap and electron affinity energies given in the table.

Besides having good QE and low thermal emittance, the ideal photocathode should also have low thermionic emission. While the thermionic and photoelectric work functions are the same in metals, they

differ in semiconductors. The thermionic work function is the energy difference between the Fermi and vacuum levels. Referring to Figure 2, it can be seen that the thermionic work function for semiconductors

is then $\frac{1}{2}E_G + E_A$. One could apply this relation to the cathodes listed in Table III to form an estimate of

the thermionic emission, however it is better to use experimental values instead, since the emission is dependent upon many other factors such as defects and surface condition. Sommer also lists the thermionic emission at room temperature for some of the materials shown in Table III. In particular he notes that the K_2CsSb has one of the lowest emissions of the bi-alkali cathodes, 10^{-11} microamps/cm² [42], adding to its suitability as a photocathode for use in RF and DC guns.

Table III: Properties of the Semiconductor Cathodes.

The thermal emittances are computed using the listed photon, gap and electron affinity energies in Eqn. [7] and expresses the thermal emittance as the normalized rms emittance in microns per rms laser size in mm.

Cathode Type	Cathode	Typical Wavelength, $\lambda_{opt}(nm)$, (eV)	Quantum Efficiency (electrons per photon)	Vacuum for 1000 Hrs (Torr)	$Gap\ Energy + \\ Electron\ Affinity, \\ E_A + E_G\ (eV)$	Thermal Emittance (microns/mm(rms))	
						Eqn. [7]	Expt.
		211, 5.88	~0.1	10 ⁻⁹	3.5 [42]	1.2	0.5±0.1 [35]
	Cs ₂ Te	264, 4.70	-	-	دد	0.9	0.7±0.1 [35]
	2	262, 4.73	-	-	"	0.9	1.2 ±0.1 [43]
PEA:							
Mono-alkali	Cs ₃ Sb	432, 2.87	0.15	?	1.6 + 0.45 [42]	0.7	?
	K_3Sb	400, 3.10	0.07	?	1.1 + 1.6 [42]	0.5	?
	Na ₃ Sb	330, 3.76	0.02	?	1.1 + 2.44 [42]	0.4	?
	Li ₃ Sb	295, 4.20	0.0001	?	?	?	?
	Na ₂ KSb	330, 3.76	0.1	10 ⁻¹⁰	1+1 [42]	1.1	?
PEA:	(Cs)Na ₃ KSb	390, 3.18	0.2	10^{-10}	1+0.55 [42]	1.5	?
Multi-alkali	K ₂ CsSb	543, 2.28	0.1	10-10	1+1.1 [42]	0.4	?
	K ₂ CsSb(O)	543, 2.28	0.1	10 ⁻¹⁰	1+<1.1 [42]	~0.4	?
NEA	GaAs(Cs,F)	532, 2.33	~0.1	?	1.4±0.1 [42]	0.8	0.44±0.01 [44]
		860, 1.44	-	?	>>	0.2	0.22±0.01 [44]
	GaN(Cs)	260, 4.77	-	?	1.96 + ? [44]	1.35	1.35±0.1 [45]
INICA							
	GaAs(1-x)Px x~0.45 (Cs,F)	532, 2.33	-	?	1.96+? [44]	0.49	0.44±0.1 [44]
S-1	Ag-O-Cs	900, 1.38	0.01	?	0.7 [42]	0.7	?

NEA Cathodes for ERL's:

A large variety of photocathodes are employed for production of bright electrons: from metallic cathodes (Mg, Cu, Nb, and Pb) typical of RF and superconducting RF guns [41, 32, 39] to high quantum efficiency alkali-antimonide and multi-alkali cathodes (Cs₂Te and K₂CsSb) [46] as well as III-V semiconductor photocathodes activated to negative electron affinity [47, 48]. For the production of high average current beams as required for ERLs only high QE cathodes are practical, with those having a

good response in the visible being preferred to keep the requirements on the laser system realistic (typically frequency doubled high power IR lasers). While good quantum efficiency is an important consideration for the new generation of high current high brightness sources, it is by far not the only figure of merit. Other key factors are longevity of the photocathodes during the operation as well as a short (picosecond or less) temporal response of photoemitted electrons and low transverse intrinsic (thermal) emittance [49]. The longevity aspect of photocathodes itself has several components to it, including the vacuum condition, the state of the surface, especially for cesiated photoemitters, operational conditions such as beam losses downstream of the gun and ion back bombardment. Even though other physical mechanisms can dominate cathode lifetime in a particular setup, it is the ion back bombardment which sets the ultimate limit to the photocathode longevity. To meet the longevity requirement, therefore, a two-prong approach is necessary: 1) improving of the photoemitting materials by e.g. using stoichiometric compounds or large gap materials with stronger binding of the cesiated layer, or even eliminating Cs and achieving the NEA condition through delta-doping techniques [50]; and 2) improvement of operational conditions through achieving better vacuum, halo and beam loss minimization in the gun vicinity. Additionally, we note that a care should be exercised when reporting photocathode lifetime values so that the main cause of the degradation is properly identified and correctly attributed to (which may have very little to do with a particular photocathode material choice).

The accelerator community so far has primarily been users of known photocathode materials when employing them for high brightness beam production. Since the understanding of the requirements and demands on the photocathodes for new high brightness high current electron sources has grown tremendously over the last decade, there exists a well-defined incentive for accelerator scientists to be engaged in the effort of obtaining a comprehensive understanding, which will ultimately lead to the creation of better photocathodes. A notable example worth emulating in this regard is the use of GaAs for the production of polarized electrons. Once the need for photocathodes delivering a higher degree of polarization was identified, the accelerator community stayed engaged in the process of improving their performance, contributing to the creation of strained superlattice photocathodes now operating close to the theoretical limit of polarization (over 90% degree of polarization improved from the initial 30% for the bulk GaAs) [51]. A similar need remains to be addressed for high brightness high current unpolarized beams by providing careful photocathode characterization (transverse and longitudinal energy distributions, photoemission response time performed in a systematic and well-controlled environment) and then using these experimental data as an input for development of comprehensive and verifiable theoretical models, which will eventually allow engineering of new photocathode materials with the desired properties.

As a motivation reiterating the need for a systematic approach, one could point out the need for better understanding of photocathode thermal emittance, and in particular the thermal emittance of high quantum efficiency materials. For example, three different III-V materials were investigated at Cornell University over a wide range of photon energies (GaAs, GaAsP, and GaN), and a large variation of thermal transverse energies has been observed. GaN has been found to have a surprisingly large thermal emittance as compared to GaAs when excited with photons of energies above the bandgap by a similar amount. GaAsP has demonstrated very long response times as well as strong surface condition dependence on both thermal emittance and the response time. No such strong dependence was observed for GaAs and GaN. While several possible causes explaining these phenomena have been proposed [47,45], the results still remain to be quantitatively explained.

Another puzzling fact concerns the transverse momentum conservation and the role of the reduced mass on the thermal emittance. Some groups [36] have reported seeing the effect of momentum conservation at the surface from electrons thermalized to the Γ valley in GaAs and the resulting narrow cone emission (and sub-thermal intrinsic emittance as a result) by the virtue of the effective mass ratio for electrons inside the semiconductor and in vacuum (analog of Snell's law in optics). However, other measurements [52, 47] show that the emitted electrons essentially do not experience the energy spread reduction due to the effective mass change, and an energy spread equal to the lattice temperature is obtained for a near band gap photon illumination. This is in contrast with simple theoretical models which predict much smaller values in the case of GaAs with effective mass ratio of 0.067 (effective mass in Γ valley to vacuum mass). The surface morphology and preparation techniques may be the deciding factors behind these seemingly conflicting observations, and these need to be accounted for in a systematic fashion. All this underscores the importance of bringing thermal emittance and response time of photoemitted electrons into quantitative agreement with a comprehensive theory.

Development of theoretical models for NEA photocathodes:

As pointed out earlier, critical parameters for high brightness photocathodes, such as thermal emittance and response time, can be largely understood in the framework of Spicer's three-step model [53]. It should be noted that electron thermalization to the bottom of the conduction band in NEA photocathodes occurs quickly (e.g. 10^{-13} to 10^{-12} s in GaAs), thus, it is possible in principle to have both cold and sufficiently prompt electrons for ERL applications where a picosecond cathode response typically suffices.

The Spicer model has been extremely fruitful in explaining a wide range of photoemission-related phenomena. Its use has been typically limited to explaining quantum efficiency dependence on the wavelength using parameterized expressions with one or more adjustable parameters [54, 55]. The diffusion model has also been proven useful in explaining temporal response from GaAs [56, 57, 47]. Recently, Jensen et al. demonstrated excellent quantitative agreement of QE versus photon energy for Cs₃Sb as computed in the framework of the three-step model without relying on adjustable parameters [58]. The model is also starting to be used to explain thermal emittance from metals and cesiated metallic surfaces [4, 59]. The next logical step is to explain the transverse, longitudinal energy distributions and response time from high quantum efficiency photocathodes. Development of a sophisticated model should be a research goal of ERL photocathode R&D, the model will be refined with time as new data and theoretical insights become available. The ultimate objective is to obtain sufficient predictive power from the modeling to allow band structure and basic geometry engineering (active layer thickness, etc.) of new photocathodes to achieve favorable properties for high brightness average current photoinjectors. The essential model includes photon absorption, electron transport (diffusion and inelastic phonon scattering), as well as basic surface interface interaction [58, 60, 36]. It also becomes necessary to include multiple conduction band minima and the intervalley scattering between them as the effect has been shown to matter for a number of photocathodes. This proves to be important for indirect gap photocathodes such as GaP and is known to increase the thermal emittance [61]. Similarly, the effect of intervalley scattering may play a significant role in direct gap photocathodes when indirect conduction band minima are insufficiently separated in energy. Surface and geometry effects become critical when the photon absorption depth is comparable to the band-bending region (e.g. bend-banding region is about 10 nm for GaAs). The spatial variation of the potential near the surface then needs to be incorporated into the model. Similarly, effects of geometry matter when modeling epitaxially grown thin layer photocathodes [57]. Additionally, empirical parameters describing the effects of the surface roughness and surface states will have to be added to better account for these phenomena.

Cathodes by Design

Beyond the basic cathode types just discussed there are cathode systems designed to meet the needs of a particular application. In the simplest form, a laser producing photo-electrons from a thermionic cathode is a synthesis of two emission phenomena which can improve the QE and cathode robustness.

By far the most novel and technically challenging cathode is the diamond amplified cathode proposed by BNL [62]. In this scheme, a K_2CsSb cathode in transmission-mode is encapsulated in a single stage electron multiplier with a few KV across a ~1 mm gap formed between the cathode and a thin diamond film exit window. A laser generates electrons which accelerate in the gap which in turn produce secondary electrons in the diamond film. The secondary electrons escape and are accelerated in the gun. Multiplication of the photo-electrons by a factor of >100 is expected and ~ 40 has been measured [63] These cathodes are being developed for a SRF gun to drive a demonstration ERL. Very high average current densities (>10 A/cm²) have been transported through diamond.

The properties of metallic cathodes can be significantly enhanced if they are designed so that plasmons can couple to light. Plasmons are a quasi-particle of coherent oscillations of electron density but do not normally couple to light due to momentum mismatch. This momentum mismatch can be made up either by coupling with a grating on the emitting surface, or by back surface illumination through a prism. In both cases, the momentum matching manifests itself in a giant increase in yield through a small angular range of the incoming light. In the case of Al, it has been shown that for backface (Kretchmann) geometry, for a photon energy 0.5 eV above the work function, the increase in electron yield in this matched condition is remarkably around 100[64]. This increase comes from firstly a localization of field at the vacuum – metal interface, and a very large increase in absorption. Even though Al is a free electron metal, under these conditions the reflectivity becomes zero. Electron yield enhancements of 3,500, 1,000, 2,500 and 50 over the bulk emission have been measured from Ag, Au, Cu and Al respectively using multiphoton process with 100 fs laser operating at 625 nm wavelength and 50 mW average power [65] In the grating coupled case large QE increases have also been seen in the Ag-O-Cs system around 900 nm [66] and around 350 nm wavelength [67]. Plasmons can also be generated at sharp discontinuities and then trapped in nano-cavities. The plasmons generated can exist up to very high wave-vector and thus be trapped in very small cavities of the order of 10 nm in size [68] This may be useful for localizing emission on a grid to reduce the effects of stochastic electron – electron interaction, or simply to design a certain emission profile in a dot-matrix arrangement. When illuminated with short (10's of fsec) pulses, plasmon induced field localization can cause significant electron acceleration. In one example [69], 400eV electrons with 50% energy bandwidth were created using only 1.5nJ 27fsec 800 nm pulses on gold in prism coupled geometry using multiphoton excitation. This is both a good and bad result for photocathodes! On the one hand we must be careful if using plasmon enhancement to increase QE not to 'damage' the intrinsic momentum spread of the material but on the other hand plasmon acceleration might offer a way to impulsively accelerate electrons to high energy, offering a way to avoid the most damaging effects of electron – electron scattering at low energy. In recent work it has been shown that by control of the carrier envelope phase of the laser and use of small emission points or strips, quasi monochromatic emission at high energy can be obtained [70].

Summary and Conclusions

Physics Challenges

The challenges for accelerator-based cathode R&D have two principal aspects. One is to continue improving the peak brightness of beams at low repetition rate. It is advances in this area which lead to the success of LCLS. The second is to develop cathodes for use in CW, high-average current accelerators for ERLs and high-average-power FELs.

Previous accelerator R&D provided the development of high peak brightness guns as a key enabling technology for the 4th generation, x-ray FEL light sources. Further R&D leading to even lower emittance beams will allow future FELs to be built at lower beam energy and shorter high energy accelerators which will greatly reduce the overall facility cost. High-peak brightness also widens the possibilities for using advanced FEL concepts and ideas, such as the production of fully coherent attosecond x-ray pulses, in new user facilities. Already one can see the advantages where the brighter than expected beam from the LCLS gun allows the x-ray FEL to saturate in less than half the constructed undulator length, and at low charge produce a few micron long x-ray pulses which are nearly 100-times shorter than originally expected. If this was known before LCLS was built, it may have reduced the cost of the undulator system and allowed for a more aggressive FEL design.

On the other hand, the high-average current applications such as ERLs want both the very low emittance and high-average current. The ERL will require a cathode current density of approximately 1.3 mA/mm², QEs greater than 4% at visible or longer wavelengths, and an operational charge of several kiloCoulombs from a single cathode. The cathode should also have low thermal emittance as well as low thermionic and field emission to minimize beam halo. These needs present the greatest challenge for cathode technology and are perhaps where the most intense R&D should be performed.

The issues of beam halo and dark current deserve additional attention given their impact on high average current applications. Beam halo is associated with the photocurrent and results from a poorly shaped laser beam, scattered laser light, space charge interactions with the beamline impedance and poorly matched electron optics. Beam halo is minimized with good laser shaping and laser transport, and mitigation of wake fields in the low energy section of the accelerator. Dark current is produced without any laser light and is mainly due to thermionic and field emission. Typically for cathodes operating at or below ambient temperature the thermionic emission is small. (See value for K_2CsSb above.) However the field emitted current can be as high as a few mA from a high gradient gun, and comes not only from the cathode but also from any surface at high electric field and low work function cathodes may be more problematic in such an environment. Recent work at the Photo injector Test Facility at DESY, Zeuthen (PITZ) shows that cleaning the gun surfaces with dry ice can reduce the dark current by an order of magnitude [15].

In addition, achieving the high peak brightness and high current simultaneously will require meticulous three-dimensional shaping of the cathode drive laser pulse. The laser requirements are more stringent than for the low-duty factor beams because CW operation implies these guns will have lower accelerating fields making the space charge forces much more dominant, especially near the cathode. Therefore there should also be laser R&D in conjunction with the cathode research.

Summary of Cathode R&D Plan

Many of the details of the cathode R&D plan have been described both earlier in this paper and in outline form in Appendix I, therefore only a concise summary is presented here.

The proposed cathode R&D plan consists of the following three interrelated parts:

- 1. Studies of optimal cathode formation methods and cathode emission characteristics, using available surface and material diagnostics
- 2. Modeling of cathode emission physics and electron dynamics near the cathode
- 3. Operational testing in the gun and injector system.

In general terms, the first part can be performed in national lab and university surface science laboratories, likely in collaboration with user facilities. This research provides an excellent opportunity for the education of PhD level students. As its product it will fill in the knowledge gaps in indicated in cathode tables above and provide detailed information on the thermal emittance, QE, lifetime, etc. And will provide important engineering design requirements such as vacuum, temperature stability and other specifications needed to engineer an accelerator cathode system.

The second part will also use the data produced by 1) to incorporate improved physics into the electron beam simulation codes. These enhanced codes in turn will be utilized to provide more realistic beam dynamics from the cathode through the injector and entire accelerator to preserve the low cathode emittances. One result will be a physics design which can be used to engineer future guns and injectors.

The third part is then to use the knowledge gained of the previous two for testing cathodes in an operating gun and injector. If possible these tests should be performed in all three styles of guns: DC, NCRF and SCRF. For example, the test facility at JLab is already available to compare the performance of K₂CsSb and Cs:GaAs cathodes in a DC gun. And cathode studies in a DC gun can also be performed at Cornell. The VHF gun now being constructed at LBNL provides an excellent opportunity for cathode studies in a NCRF gun. The 700 MHz SRF injector in construction and 1.3 GHz operational SRF gun at BNL provide excellent test beds for cathode studies in superconducting RF environment.

Activities that must be supported within the next 5 years

In the near term adequate support (M&S and effort) is needed to continue cathode surface science activities such as reliable production of K_2CsSb cathodes. This should include development of load-lock capabilities so that cathodes can be characterized at existing user facility beamlines. This characterization should include measuring the quantum efficiency of various photocathode materials as a function of wavelength and testing performance for incident laser pulses with a variety of photon energies and temporal and transverse distributions. Analysis of the experimental results with density functional theory and other models will yield further understanding of photoemission processes which can be applied to future engineered photocathodes.

Within three to five years, a variety of cathodes should be tested in normal and superconducting RF and high voltage DC guns, coupled to a beam dynamics characterization line to verify cathode survivability and the required beam quality for ERLs and FELs. These studies can be performed at injector test stands which already exist or are under construction at ANL, BNL, Cornell, JLab and LBNL. In order to facilitate these tests a standard load lock system for transferring cathodes between the various cathode and accelerator labs should be developed. This will assure compatibility among the various research facilities.

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References

- [1] J. Murphy, plenary presentation at BES Workshop
- [2] D. H. Dowell *et al.*, "LCLS Injector Drive Laser", Proceedings of 2007 Particle Accelerator Conference
- [3] Y. Ding et al., PRL **102**, 254801(2009)
- [4] D.H. Dowell and J.F. Schmerge, "Quantum efficiency and thermal emittance of metal photocathodes", Phys. Rev. ST Accel. Beams, **12** (2009) 074201
- [5] J. G. Endriz et al., Applied Physics Letters 25, 1974, p. 261]
- [6] A. Michaelides and M. Scheffler to appear in K. Wandelt (ed.), *Surface and Interface Science*, vol.1, Wiley-VCH, (2010)
- [7] K. Harkay *et al.*, to be published in Proc. 2009 Particle Accelerator Conference, Vancouver, IEEE, MO6RFP045 (2010)
- [8] W.F. Krolikowski and W.E. Spicer, Phys. Rev. 185, 882 (1969)
- [9] S. Hüfner, *Photoelectron Spectroscopy*, pp 244, Springer 1995
- [10] I.V. Bazarov, I. Senderovich, C.K. Sinclair, "Use of multiobjective evolutionary algorithms in high brightness electron source design", Proceedings of the 2005 Particle Accelerator Conference, IEEE 0-7803-8859-3 (2005) 2188-2190
- [11] K. Togawa, et al., Proc. 2003 PAC, Portland, OR pp 3332-3334 (2003)
- [12] Akre *et al.*, "Commissioning the Linac Coherent Light Source injector", PRST-AB **11**, 030703 (2008)
- [13] C. Herrnandez-Garcia *et al.*, "A high average current DC GaAs photocathode gun for ERLs and FELs", Proceedings of the 2005 Particle Accelerator Conference, Knoxville, TN, May16-20 2005, pp 3117-3119
- [14] L. B. Jones *et al.*, "Photocathode Preparation system for the ALICE Photoinjector", AIP Conf. Proc. August 4, 2009, Vol. 1149, pp. 1089-1093
- [15] V. Volkov et al., Proc of EPAC08, Genoa, Italy 2213-2215(2008
- [16] C.M. Lyneis et al., IEEE Transactions on Nuclear Science, Vol. NS-26, No. 3, June 1979
- [17] C. Hernandez-Garcia *et al.*, "Status of the Jefferson Lab ERL FEL DC photoemission guns", proceedings of the 2009 ERL Workshop, Ithaca, NY June 2009, in press.
- [18] D. H. Dowell *et al.*, "First operation of a photocathode radio-frequency gun injector at high duty factor", Appl. Phys. Lett. (1993) **63** (15), pp 2035-2037
- [19] C. Boulware *et al.*, "Latest results at the upgraded PITZ facility", FEL2008 Conf. Proc., Gyeongju, Korea, August 2008
- [20] S. Lederer et al., FEL2007 Conf. Proc., Novosibirsk, Russia, 457 (2007)
- [21] W. Kohn and L. Sham, Phys. Rev. Lett. 140, 1133 (1965)

- [22] I. A. Nekrasov et al., Eur. Phys. J. B 18, 55 (2000)
- [23] Y. Ikuno, K. Kusakabe, e-J. Surf. Sci. Nanotech. 6, 103 (2008)
- [24] S.D. Kevan, Phys. Rev. B 28, 2268 (1983)
- [25] M. Sterrer et al., Phys. Rev. Lett. 98, 206103 (2007); and references therein.
- [26] K. Nemeth *et al.*, "High-brightness photocathodes through ultrathin surface layers on metals," Phys. Rev. Lett. 104, 046801 (2010).
- [27] U. Martinez, L. Giordano, G. Pacchioni, J. Chem. Phys. 128, 164707 (2008)
- [28] T Konig et al., J. Phys. Chem. C 113, Issue 26, 11301 (Jul. 2, 2009)
- [29] E. Pedersoli *et al.*, "Surface and bulk contribution to Cu(111) quantum efficiency", Applied Physics Letters, 2008. **93**(18): p. 183505.].
- [30] E. Pedersoli *et al.*, "Evidence of vectorial photoelectric effect on Copper", Applied Physics Letters, 2005, **87**(8): p. 081112-3
- [31] P.J. Feibelman, "Self-Consistent Calculation of the Surface Photoelectric Effect", Physical Review Letters, 1975. **34**(17): pp. 1092-1095
- [32] J. Smedley, T. Rao and J. Sekutowicz, "Lead photocathodes", Phys. Rev. ST Accel. Beams 11,013502 (2008)
- [33] A.H. Sommer, *Photoemissive Materials* (J. Wiley & Sons: New York, 1968), p.7
- [34] Ibid. pp. 21-26.
- [35] D. Sertore, D. Favia, L. Monaco, P. Pierini, "Cesium Telluride and Metals Photoelectron Thermal Emittance Measurements Using a Time-of-Flight Spectrometer", Proc. of EPAC 2004, Lucerne, Switzerland, p 408-410 (2004)
- [36] Z. Liu, Y. Sun, P. Pianetta, R.F.W. Pease, "Narrow cone emission from negative electron affinity photocathodes", J. Vac. Sci. Technol. B, 23 (2005) 2758
- [37] Juan R. Maldonado et al., Phys. Rev. ST Accel. Beams 11, 060702 (2008)
- [38] Juan R. Maldonado et al., submitted to J. Appl. Phys.
- [39] W.S. Graves, L.F. DiMauro, R. Heese, E.D. Johnson, J. Rose, J. Rudati, T. Shaftan, B. Sheehy, "Measurement of Thermal Emittance for a Copper Photocathode", Proceedings of the 2001 Particle Accelerator Conference, p. 2227.
- [40] J. F. Schmerge *et al.*, "Emittance and quantum efficiency measurements from a 1.6 cell s-band photocathode RF gun with Mg cathode", Proceedings of the 2004 FEL Conference, pp. 205-208
- [41] X.J. Wang, M. Babzien, R. Malone, and Z. Wu, "Mg cathode and its thermal emittance", Proceedings of LINAC2002, Gyeongju, Korea
- [42] Sommer, op. cit., p. 130.
- [43] M. Miltchev *et al.*, "Measurements of thermal emittance for cesium telluride photocathodes at PITZ", Proc. of 2005 FEL Conf., pp. 56—563, 2005.
- [44] I. V. Barazov *et al.*, "Thermal emittance measurements from negative electron affinity photocathodes", Proc. PAC 2007, p. 1221, 2007.
- [45] I.V. Bazarov, B.M. Dunham, X. Liu, M. Virgo, A.M. Dabiran, F. Hannon, H. Sayed, "Thermal emittance and response time measurements of a GaN photocathode", J. Appl. Phys. **105** (2009) 083715
- [46] Michelato *et al*, "Cs₂Te photocathodes for the TTF injector II", Proceedings of European Particle Accelerator Conference 1996, Barcelona, Spain, p. 1510
- [47] I.V. Bazarov, B.M. Dunham, Y. Li, X. Liu, D.G. Ouzounov, C.K. Sinclair, F. Hannon, T. Miyajima, "Thermal emittance and response time measurements of negative electron affinity photocathodes", J. Appl. Phys., **103** (2008) 054901

- [48] N. Yamamoto *et al*, "Thermal emittance measurements for electron beams produced from bulk and superlattice negative electron affinity photocathodes", J. Appl. Phys., **102** (2007) 024904
- [49] I.V. Bazarov, B.M. Dunham, C.K. Sinclair, "Maximum achievable beam brightness from photoinjectors", Phys. Rev. Lett., **102** (2009) 104801
- [50] D. Bell, "Recent progress in non-cesiated III-Nitride photocathodes", presented at 1st workshop on photocathodes: 300-500nm, University of Chicago, 20 July 2009
- [51] T. Maruyama *et al*, "Systematic study of polarized electron emission from strained GaAs/GaAsP superlattice photocathodes", Appl. Phys. Lett, 85 (2004) 2640
- [52] V.V. Bakin, *et al*, "Refraction of thermalized electrons emitted ballistically into vacuum from p+-GaAs-(Cs,O)", JETP Letters, **77** (2003) 167
- [53] W. E. Spicer, A. Herrera-Gomez, "Modern theory and applications of photocathodes", Proc. SPIE Vol. 2022, p. 18-35, *Photodetectors and power meters*, Ed. K.J. Kaufmann
- [54] W.E. Spicer, "Photoemissive, photoconductive, and optical absorption studies of alkali-antimony compounds", Phys. Rev. 112 (1958) 114
- [55] D.H. Dowell *et al*, "In situ cleaning of metal cathodes using a hydrogen ion beam", Phys Rev ST Accel. Beams **9** (2006) 063502
- [56] P. Hartmann *et al*, "A diffusion model for picosecond electron bunches from negative electron affinity GaAs photocathodes", J. Appl. Phys., **102** (2007) 024904
- [57] K. Aulenbacher *et al*, "Pulse response of thin III/V semiconductor photocathodes", J. Appl. Phys., **92** (2002) 7536
- [58] K.L. Jensen, B.L. Jensen, E.J. Montgomery, D.W. Feldman, P.G. O'Shea, "Theory of photoemission from cesium antimonide using an alpha-semiconductor model", J. Appl. Phys., **104** (2008) 044907
- [59] K.L. Jensen, N.A. Moody, D.W. Feldman, E.J. Montgomery, P.G. O'Shea, "Photoemission from metals and cesiated surfaces", J. Appl. Phys., **102** (2007) 074902
- [60] G. Vergara, A. Herrera-Gomez, W.E. Spicer, "Escape probability for negative electron affinity photocathodes: calculations compared to experiments", Proc. SPIE Vol. 2550, p. 142 (1995)
- [61] A.W. Baum, W.E. Spicer, R.F. Pease, K.A. Kostello, and V.W. Aebi, "Negative electron affinity photocathodes as high-performance electron sources", Part 2: Energy spectrum measurements", Proc. SPIE Vol. 2550, p. 189 (1995)
- [62] J. Smedley, I. Ben-Zvi, J. Bohon, X. Chang, R. Grover, A. Isakovic, T. Rao, Q. Wu, "Diamond Amplified Photocathodes", in Diamond Electronics—Fundamentals to Applications II, Mater. Res. Soc. Symp. Proc. 1039, Warrendale, PA, 2007, 1039-P09-02
- [63] Chang, X., Ben-Zvi, I.; Burrill, A.; Kewisch, J.; Rao, t.; Smedley, J.; Wang, Y-C.; Wu, Q., "First Observation of an Electron Beam Emitted from a Diamond Amplified cathode, PAC 09, Vancouver, Canada
- [64] T.A. Callcott and E.T. Arakawa, "Volume and surface photoemission processes from plasmon resonance fields", Physical Review B, 1975, **11**(8): p. 2750
- [65] T. Tsang, T. Srinivasan-Rao, J. Fischer, Phys. Rev. B Vol. 43, pp. 8870-8878, 1991
- [66] J.G. Endriz, Applied Physics Letters, 25, 1974, p.261.
- [67] F. Sabary *et al.*, "Silver-covered diffraction gratings as possible high-efficiency laser driven photoemitters", Applied Physics Letters, 1991. **58**(12): p. 1230-1232
- [68] J. Le Perchec *et al.*, "Why Metallic Surfaces with Grooves a Few Nanometers Deep and Wide May Strongly Absorb Visible Light", Physical Review Letters, 2008. **100**(6): p. 066408-4

[69] S.E. Irvine, A. Dechant, and A.Y. Elezzabi, "Generation of 0.4-keV Femtosecond Electron Pulses using Impulsively Excited Surface Plasmons", Physical Review Letters, 2004. **93**(18): p. 184801 [70] P. Dombi and P. Rácz, "Ultrafast monoenergetic electron source by optical waveform control of surface plasmons", Optics Express, 2008, **16**(5): pp. 2887-2893

Appendix I

Outline of Near and Long Term Cathode Research and Development

Near Term R&D

- Compile existing cathode database
 - Use the database to select promising cathode materials for various applications
 - Perform experiments to fill in the "data holes"
- Choose a prototype cathode material
 - Good QE in visible and can survive in expected operating gun environment
 - Develop fabrication techniques to reliably produce good QE
 - Characterize material properties of "good" cathodes, both to improve reproducibility and optimize growth parameters
 - Study surface characteristics: Lifetime, thermal emittance, robustness,...
 - Set specifications for survival in operational gun environment
 - Analysis using surface science theory, like Density Functional Theory,...
- Implement emission and surface properties into beam simulation codes
 - Study beam dynamics near cathode and beyond to include injector designs
- Pursue advanced cathode materials R&D in surface science lab
 - Surface science labs at BNL, LBNL and ANL are already studying cathode materials
- Use results of cathode studies to define specification for injector cathode system
 - Programming new physics into existing particle simulation codes
 - Photoemission models are being put into codes like IMPACT and Parmela
 - Load lock specifications and gun vacuum requirements
 - Cathode fabrication/transfer system for operational gun and injector
 - Develop a versatile load lock design that can be shared and copied by all the labs to encourage transfer of cathode materials and ideas between labs
 - Load lock should be compatible with both the lab surface science chambers and the gun to both fabricate new cathodes and test used ones in the surface science lab
- Initial testing in gun within 1 to 2 years

Long Term R&D

- Operation testing at full duty factor in support of beam physics experiments at injector test facility.
- Further develop and implement advanced cathodes
- R&D in support of injector operations

Appendix II Ongoing Cathode Research

Cathode Research at National Laboratories

Currently three Laboratories, ANL, BNL and LBNL have active R&D programs on K₂CsSb cathode preparation. This type of cathode can be driven with laser light at visible wavelengths and has demonstrated robust operation in vacuum environments in the 10⁻⁹ to 10⁻¹⁰ Torr range, typical of normal conducting radio frequency guns. Jefferson Lab has developed GaAs photocathodes for the CW 10 KW IR-FEL and has operated them at 10 mA of average current.

Argonne National Laboratory: Several groups are collaborating on cathode research for applications such as light sources and high-energy physics. For light sources, R&D on both cathode physics and high-brightness injector designs are being pursued. An ARPES lab is being commissioned that will be used to carry out fundamental photocathode studies for ultra-low emittance, including the benchmarking of DFT analyses. For emittance compensation, laser pulse shaping schemes have been developed and tested on the bench, and tests in the Injector Test Stand (ITS) are planned. Various injector design studies are underway. A thermionic RF injector is being studied that combines the SCSS-type CeB₆ thermionic cathode with a very-low frequency (~100 MHz) LBNL-type RF gun [P.N.Ostroumov, K.-J. Kim, P. Piot, Proc. 2008 Linac Conference, 676 (2009)]. To potentially shorten the pulse length in such an injector, studies of laser-gated emission using a standard APS thermionic RF injector are underway in the ITS. For high-energy physics and other applications, high-QE cathode preparation and study are underway for very-high-charge injectors and for photodetectors; these cathodes include Cs₂Te, bialkali, and III-V semiconductors (see also photon detection collaboration under University research).

Brookhaven Nation Laboratory: BNL researchers have been aggressively working for a number of years on a variety of photocathode related topics including development, optimization and characterization of metal, semiconductor and superconducting cathode materials, investigation of various photoemission processes including multi photon, surface plasmon and photofield assisted emission and modeling of photoemission. They have been recently concentrating their effort on K₂CsSb and its use in a diamond amplified cathode. In this scheme, the cathode in transmission-mode is encapsulated in a kind of single stage electron multiplier with a few KV across a ~1 mm gap formed between the cathode and a thin diamond film. A laser generates primary electrons which accelerate in the gap and produce secondary electrons in the diamond film. The secondary electrons escape and are accelerated in the gun. Multiplication of the photo-electrons by a factor of >100 is expected And 40 has been measured. These cathodes are being developed for an SRF gun as part of an ERL demonstration.

BNL has an extensive material characterization effort dedicated to cathode development, utilizing resources at both the National Synchrotron Light Source and the Center for Functional Nanomaterials. This effort includes surface morphology and chemical analysis with scanning electron microscopy (including x-ray analysis), atomic force microscopy, near-edge X-ray absorption fine structure and X-ray photoemission spectroscopy. Crystallinity studies are performed with X-ray diffraction, X-ray topography and electron diffraction. Angle resolved photoemission and total-yield spectroscopy are used to measure band structure and electron affinity of cathodes. Bulk impurities in cathode materials such as diamond have been investigated using IR spectroscopy, Raman imaging and photoluminescence. X-ray micro-beam mapping has been used to identify the causes of spatial non-uniformity in diamond cathode

response, and high-flux x-ray beams have been used to test the high current density performance of diamond.

BNL has three major photoinjector based accelerators (Accelerator Test Facility, Source Development Lab, Laser-Electron Accelerator Facility) in operation, and a fourth under construction (Energy Recovery Linac - ERL). The BNL ERL, with a design goal of 0.5A average current, will be a test bed for the high-current ERLs needed for light-source applications. The ATF injectors operating at normal conducting mode at 2.856 GHz has been used extensively in testing metal photocathodes to deliver electron beams of very high brightness and low average current. Copper and Magnesium cathodes tested in this gun has led the way to a number of very high brightness injectors including the LCLS injector. The superconducting RF injectors operating at 1.3 GHz, and 700 MHz and 112 MHz injectors currently under construction will be used for testing cathodes capable of producing very high brightness and very high average current.

Jefferson National Laboratory: Jefferson Lab's FEL is developing the next generation DC photoemission gun based on inverted insulators for reliable operation at 500kV and with the capability of photocathode change out via load-locked system. The aim is to study various types of photocathodes in the same gun environment, in particular K₂CsSb and Cs:GaAs. The gun will be located in the FEL's Gun Test Stand facility which has a 600 kV DC power supply, a drive laser system with the flexibility for 3D pulse shaping, and a diagnostics beam line that needs to be upgraded for measuring transverse and longitudinal emittance.

Lawrence Berkeley National Laboratory: LBNL is working on three aspects of photocathode research; 1) understanding the fundamental aspects of the interaction of light with the electronic system of a metal surface, leading to production of electrons and 2) understanding the chemistry and reliable production of Alkali Antimonide photocathodes for FEL applications and 3) the design of plasmonic metal surfaces for enhanced production of photoelectrons. A lab dedicated to Alkali Antimonide photocathode production and R&D and a second lab for characterization of cathodes using surface science techniques such as Angle Resolved Photo-Electron Spectroscopy (ARPES). There is also a UV PEEM to examine the microstructure of emitting surfaces, and synchrotron radiation surface analytical techniques at the ALS. The lab is in the final stages of construction of a 20 MV/m RF photogun that will be used as a facility for photocathode testing. The photocathode production and transfer system for this system is under design and construction.

Cathode Research at Universities

Maryland University: The photocathode research group at Maryland University focuses on studying dispenser cathodes for robust perfomance at high average current. This type of cathode is based on a metal substrate where Cesium is dffused trhough the bulk of the cathode replenish contantly the quantum efficiency. In collaboration with the **Naval Research Laboratory** matematical and computational models of density functional theory for emission from cesium-coated metals and some semiconductor materials have been developed for many years.

Cornell University: As a part of ongoing ERL R&D effort, Cornell University is in the process of establishing a dedicated gun and photocathode research laboratory. The photocathode research program involves the study of high quantum efficiency photocathodes, their properties as they pertain to high

brightness electron beam creation, namely thermal emittance and response time. So far the photocathodes under investigation have belonged to III-V semiconductor group (GaAs and GaAsP [I.V. Bazarov, B.M. Dunham, Y. Li, X. Liu, D.G. Ouzounov, C.K. Sinclair, F. Hannon, T. Miyajima, "Thermal emittance and response time measurements of negative electron affinity photocathodes", J. Appl. Phys., 103 (2008) 054901; I.V. Bazarov, D.G. Ouzounov, B.M. Dunham, S.A. Belomestnykh, Y. Li, X. Liu, R.E. Meller, J. Sikora, C.K. Sinclair, F.W. Wise, T. Miyajima, "Efficient temporal shaping of electron distributions for high brightness photoemission electron guns", Phys. Rev. ST Accel. Beams 11 (2008) 040702], GaN [I.V. Bazarov, B.M. Dunham, X. Liu, M. Virgo, A.M. Dabiran, F. Hannon, H. Sayed, "Thermal emittance and response time measurements of a GaN photocathode", J. Appl. Phys. 105 (2009) 083715]). Alkali-antimonide photocathodes are being evaluated, and a setup that allows the growth of Cs₃Sb, K₂CsSb, and Na₂KSb has been designed and now being constructed. In addition to the time resolving diagnostics which now allows characterization of photocathode response times to a 0.1ps level, a dedicated setup is being built for simultaneous characterization of transverse and longitudinal energy spectra outside of high voltage DC gun environment based on the method originally implemented at Max Plank Institute [D.A. Orlov et al, "Energy distributions of electrons emitted from GaAs(Cs,O)", Appl. Phys. Lett., 78 (2001) 2721]. Another research direction is developing Monte-Carlo models incorporating the photocathode physics to explain the salient features of the semiconductor photocathodes and their dependence on the laser wavelength, band-gap structure, and electron transport parameters in materials. One of the early successes of such modeling has been qualitative explanation of wavelength dependence of the response time from GaAs photocathode [P. Hartmann et al, "A diffusion model for picosecond electron bunches from negative electron affinity GaAs photocathodes", J. Appl. Phys., 102 (2007) 024904; K. Aulenbacher et al, "Pulse response of thin III/V semiconductor photocathodes", J. Appl. Phys., 92 (2002) 7536]. In collaboration with EE Department at Cornell University and SVT Associates, the work is underway to identify new promising structures with good quantum efficiency when excited with visible light suitable for low emittance beam operation for trial measurements. Independently from the gun and photocathode laboratory, the operating 10 MeV ERL injector prototype accelerator allows investigations in realistic running conditions of high average current performance of existing and new photocathodes. The highest average current obtained so far was 8 mA at 6 MeV (20 mA DC beam demonstrated from GaAs after the high voltage gun), and operation at significantly higher currents is planned for this year. Detailed lifetime studies at high average current will commence then also.

Vanderbilt University: The group within the Physics Department and in collaboration with the Electrical Engineering Department studies field-emitter arrays (FEAs) cathodes for production of bright electron beams for compact free-electron lasers (FELs). This type of cathodes are rugged, require no laser driver, and generate little heat, which makes them attractive to test in normal conducting RF guns, but have only been tested in ~50 kV DC guns. The group has developed two methods to fabricate diamond FEAs, in the first method, pyramids are formed on a Si substrate and sharpened by microlithography and then coated with CVD nanodiamond. Typically, tip radii on the order of hundreds of nanometers are formed on 20-μm pyramids. In the second method, all-diamond pyramids are formed by a mold-transfer process in which they become sharpened from an oxide layer in the mold process, with tip radii smaller than 10 nm formed on 10-μm pyramids.

Old Dominion University: Through the applied research center in the Jefferson Lab Campus, the Electrical Engineering Department has performed over the last decade many surface analysis studies on GaAs photocathodes for both the Continuous Electron Accelerator Facility polarized electron gun and for the Free Electron Laser un-polarized, high current DC photoemission gun. The director of the applied research center has several publication on cesiated GaAs photocathode studies and has a strong graduate research program.

The College of William & Mary: The Applied Physics Department has various surface science laboratories located in the Applied Research Center, also in the Jefferson Lab Campus. Establishing a collaboration with the College of William & Mary to conduct R&D on photocathode preparation and surface analysis would be very easy. Some of the equipment potentially available include Time of Flight Ion Mass Spectromenter, Scanning Probe Microscope, Atomic Force Microscope, DekTak Surface Profilometer, Fourier Tranform-Infrared Spectrometer, Scanning Electron Microscope, Hirox High Resolution Digital Microscope, Variable-angle Spectroscopic Ellipsometer, Scanning Tunneling Microscope, etc.

Cathode Research for Photon Detection Applications

Additional efforts in photocathode research are being performed by an interdisciplinary collaboration that is devoted to the development of Large Area Photodetectors. Under the leadership of Argonne National Laboratory, the University of Illinois Urbana Champaign, the University of Illinois Chicago, Washington University, and the Space Science Laboratory of the University Berkeley, these institutions are working on novel design concepts of fast and robust photocathodes with high quantum efficiency, low dark current, and long life time. The efforts address engineering, design, simulation, and industrial production aspects of standard bialkali and III-V based photocathodes as well as nano-engineered materials. There is sizable overlap in this R&D effort with future light source accelerator applications.