# DEVELOPMENT OF ALKALI-BASED HIGH QUANTUM EFFICIENCY SEMICONDUCTORS FOR DISPENSER PHOTOCATHODES\*

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## Abstract

Photocathodes as electron beam sources are expected to meet the stringent requirements of high performance FELs but exhibit a lifetime-efficiency tradeoff. High quantum efficiency (QE) cathodes are typically semiconductors, well described by recently enhanced theory [1]. Cesium dispenser technology, proven to extend lifetime of Cs-W cathodes [2], is proposed to be extended to high OE via the development of suitable semiconductor coatings which are for rejuvenation. Rejuvenation occurs via controlled cesium diffusion through a sintered substrate to resupply the surface (as described by models of pore and surface diffusion). Compatible coatings must be thermally stable materials with a cesium-based surface layer. Following standard fabrication processes, we discuss alkali antimonides and alkali aurides as cesium dispenser photocathode coatings and analyze future prospects. We also describe improvements to experimental techniques.

## INTRODUCTION

Historical Contenders for High QE and Lifetime

Quantum efficiency, or QE – that is, the ratio of emitted electrons to incident photons – is a key parameter for high performance FELs. Given constrains on drive laser power, both from technological limits and from material damage considerations, ultimate demands for high average electron current require the best available QE.

As seen in Fig. 1, the high lifetime cathodes are the simplest ones: polished metal surfaces such as copper or molybdenum are commonly used under demanding injector conditions. But with deplorable QE stemming from high work function, extracting high average power is simply not attainable using a metal photoemitter. Other facilities have seen extended lifetime from more complex cathode structures such as [Cs]GaAs and Cs<sub>2</sub>Te. But a truly long-lived high QE photocathode remains elusive.

In the early days of photoemission the S-1 (Cs-O-Ag) photocathode was the only photoemitter not a metal or metal oxide. Its QE varies from 0.3% to 0.5% in the visible region with excellent long wavelength response, and throughout the 1930s and 1940s this was the best QE available. In the 1950s and 1960s the alkali antimonide family was discovered through a series of lucky accidents [4] and has been well studied since. QE of the S-20 (K<sub>2</sub>CsSb) exceeds 20% at 400 nm. In the sealed, alkali-

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Figure 1: Life-QE trade for injector cathodes, [Cs]W dispenser for comparison, adapted from Ref. [3].

rich environment of phototubes, such cathodes also exhibit long life. But when exposed to hard vacuum they typically last only a few hours or days before they must be rejuvenated by recoating the surface with Cs [5], and this does not even mention the significant RF heating and back-bombardment concerns in the accelerator environment. Since the pioneering work by Spicer, Sommer, and colleagues, multialkali phototubes have neared theoretical limits. For instance, QE of the commercially available Hamamatsu "Ultra Bialkali", by some as-yet unrevealed alchemy, tops an incredible 43% at 400 nm. Unfortunately such phototube performance has little hope of being duplicated in the accelerator environment, where QE is often less than ideal and lifetime suffers as well from the presence of reactive gases such as water vapor,  $CO_2$ , and  $O_2$  [6].

In an effort to provide more stable and more efficient sources, negative electron affinity cathodes were proposed and tested in the 1960s and have become serious contenders on both fronts. [Cs]GaAs has a QE which exceeds 14% in the green. Lifetime is given as weeks [7], although this follows a more rapid initial decay, and involves operation under excellent vacuum conditions and relocation of the laser spot across the cathode face during its life cycle, as well as periodic recessiation.

Significantly not on this list of historical high QE contenders is cesium auride. This is due to Spicer's early work exhibiting 0.02% QE at 400 nm, which led to many workers writing off the Cs-Au compound entirely as a photocathode candidate [8]. Based on results reported here, that dismissal may have been premature.

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## The Dispenser Photocathode

High efficiency with extended lifetime is the promise of the novel dispenser photocathode concept [9] under development by the University of Maryland (UMD) and its collaborators. Unlike thermionic dispensers, the UMD prototype uses cesium as the active coating, and temperatures of operation are significantly cooler. Dispenser photocathodes replenish QE *in situ*, so in principle there is no need for the mechanical complexity of a load-locked, retractable cathode stalk, transfer cart, or preparation chamber. In practice, unless the cathode surface is air-stable prior to introduction of cesium, a separate preparation chamber may still be required, and unless the cathode's cesium reservoir can be activated ingun without unwanted emission, a retractable cathode stalk may also be required.

In the dispenser configuration, cesium from the aforementioned sub-surface reservoir diffuses to the surface through a microscopically diffusion barrier to replenish the degraded surface layer. The goal of this research is to engineer and develop more efficient, robust, long-lived regenerable photocathodes in support of predictive photocathode modeling efforts and suitable for photoinjection applications.

# **EXPERIMENTAL PROGRESS**

## Apparatus and Upgrades

The UMD test stand has been extensively described previously [3], and in it QE is measured while surface conditions are systematically varied (coverage or layer thickness, temperature, gas composition, etc.). Recent upgrades include a more stable UV diode laser, cathodes as small as 1 mm dia., and measurement of dispenser cesium emission at high temperature. A monochromator/Hg lamp for 253 nm is under installation.

The same test stand is used for dispensers and high QE coatings. The latter are described below on dispenser-type substrates (70% dense sintered tungsten) but have not yet been combined with an operational dispenser cathode due to thermal compatibility.

### Potential of Alkali Aurides and Antimonides

Cesium auride was studied by Spicer et al. in 1959 [8]. With QE at 400 nm of 0.03% and just a third that in the green, it was no wonder researchers stopped there. However, recent results at UMD indicate different fabrication methods for Cs-Au (denoting constituents, not stoichiometric ratios of necessarily 1:1) may lead to much higher QE. As shown in Fig. 2, we have observed not only more than an order of magnitude increase in QE over the 1959 results, but also an extended lifetime at 1 nTorr. Thermal stability, as with any cesium-based bulk semiconductor, continues to be a concern above 50°C. However, the ready interdiffusion of Cs with Au even at room temperature may allow novel solutions using modified dispensers. The fabrication process also offers



Figure 2: Best CsAu QE and life in the green at 1 nTorr



Figure 3: Cs-Au heating post-formation, optimizing QE.

room for further QE optimization beyond that shown in Fig. 2. The QE optimization process currently being used at UMD is shown in Fig. 3. This follows a standard fabrication of Cs-Au and the heating first to 50°C and then to 70°C begins at room temperature with the cesiated gold layer already formed. The results in Fig. 3 are not the best results in the green among the Cs-Au cathodes fabricated by UMD – those are shown in Fig. 2 – but the curves in Fig. 3 exhibit exceptional UV QE of 1.5%, all the more striking when compared to the 1959 Spicer value of 0.03%. And commenting on the variability of QE in the UMD results, note this bodes well for future optimization of high QE Cs-Au – there is room in the parameter space for it. It appears Cs-Au may yet be a candidate for high QE dispenser photocathodes.

As previously reported [10], we have measured for comparison the results of the simplest cesium-based antimonide,  $Cs_3Sb$ . QE was achieved of 11% at 375 nm and 3.3% at 532 nm for an initial 100 Å antimony layer. However, thermal stability is very poor above 50° C,

negating dispenser compatibility. In addition, the Cs may leave both surface and bulk, causing irreversible losses to stoichiometry and QE. Nor can the fabrication process be performed at room temperature: the range is normally 120° to 140°C with rapid cooling following formation.

But despite the shortcomings of Cs<sub>3</sub>Sb, its close relative the S-25 trialkali [Cs]Na2KSb is in fact also a candidate material for a high QE rejuvenable dispenser With a more temperature stable photocathode. photoemissive bulk and a cesium-based surface-only layer, it might be able to maintain surface Cs levels with a minimum-temperature dispenser. Such a test is planned at UMD.

### SUMMARY

Dispenser photocathodes will only meet the need for higher QE with longer life as the semiconductor coatings are developed and tested which are compatible with the dispenser architecture. Highlighted here are Cs-Au and [Cs]Na2KSb as possible materials of choice. Both offer OE of order 1% in the green.

Experimental results continue to be applicable to the development of a predictive photoemission theory. Through our ongoing collaboration with NRL, a photoemission theory for semiconductors is being developed [11]. Recent advances in understanding of scattering dynamics during electron transport to the photocathode surface will prove crucial in advancing this theory to a predictive capacity [1].

Future experimental work will involve the characterization of the CsAu cathode, fabrication for the first time of the S-25 trialkali at UMD, and incorporation of semiconductor coatings with operational dispenser architectures to test rejuvenation of QE and extended lifetime.

#### REFERENCES

- [1] K. L. Jensen, E. J. Montgomery, D. W. Feldman, P. G. O'Shea, J. J. Petillo, "Corrections to Quantum Efficiency Predictions for Low Work Function Electron Sources", (this conference).
- N. A. Moody, K. L. Jensen, D. W. Feldman, E. J. [2] Montgomery, P. G. O'Shea, J. Appl. Phys. 102, 104901 (2007). [3] E. J. Montgomery, Ph.D. dissertation, University of Maryland (2010).
- [4] A. H. Sommer, J. Vac. Sci. Tech. A 1, 119 (1983).
- [5] A. H. Sommer, Photoemissive Materials (Wiley, New York, 1969).
- [6] P. Dolizy, F. Groliere, J. Phys. D: Appl. Phys. 19, 687 (1986).
- R. Calabrese et al., Nucl. Instrum. Methods Phys. [7] Res. A 340, 109 (1994).
- [8] W. E. Spicer, A. H. Sommer, J. G. White, Phys. Rev. 115, 57 (1959).
- [9] N. A. Moody, K. L. Jensen, D. W. Feldman, P. G. O'Shea, E. J. Montgomery, Appl. Phys. Lett. 90, 114108 (2007).
- [10] E. J. Montgomery et al., Proc. IEEE Particle Accel. Conf. TUPMS010 (2007).
- [11] K. L. Jensen et al., J. Appl. Phys. 104, 044907 (2008).