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Experimental verification of the 3-step model of photoemission for energy spread and emittance measurements of copper and CsBr-coated copper photocathodes suitable for Free Electron Laser applications

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Abstract
This paper presents measurements and analysis of the quantum efficiency and intrinsic emittance of Cu and CsBr coated Cu photocathodes. The data analysis uses expressions for the quantum efficiency and the intrinsic emittance for metal cathodes previously derived from Spicer's three-step model of photoemission [1]. Data taken with a 257 nm CW laser on (100) Cu crystals indicate an emittance of 0.77 (μm/mm-rms) for CsBr coated and 0.42 (μm/mm-rms) for uncoated cathodes. The high QE and low emittance observed for CsBr coated cathodes have applications in Free Electron Laser and other devices requiring high brightness electron beams.

Two important parameters required for an effective photocathode for Free Electron Lasers (FEL) and other applications are electron emission Quantum Efficiency (*QE*) and energy spread under pulsed operation. In this paper, we demonstrate increased *QE* for CsBr coated Cu relative to untreated copper. In addition, we have developed a compact parallel plate energy analyzer to measure the energy spread of the electron beam. The resulting energy spread gives the CsBr/Cu intrinsic emittance which is compared with similar measures for untreated copper.

CsBr films deposited on several substrates [2-10] including, Cr, Ta, Mo, Cu, Nb, Au, Al and GaN single crystals, when illuminated with a 257nm CW laser, have shown low energy spread and a photo yield improvement >50X relative to the underlying uncoated substrate performance under similar conditions. The performance enhancement of CsBr coated photocathodes has been explained by color centers generated by the UV radiation [10-11]. The color centers form energy states lying inside the CsBr energy gap of approximately 7.3 eV and centered about 3.8 eV below the vacuum level. These gap states allow photoemission with a photon excitation energy

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of 4.8 eV which is less than the energy gap. In addition, Cs migration to the top of the CsBr film lowers the work function and enhances the photoemission [10-11]. It is important to note that the contamination problems of the vacuum system and its components seen in standard NEA photocathodes which use deposited Cs, are not observed with CsBr films since in this case the free Cs is produced locally only in the area illuminated by the UV radiation. Therefore, the Cs remains bound in the high resistivity CsBr film during film deposition and use.

For the CW measurements, the sample substrates were (100) single crystal Cu substrates 1x1 cm^2, 0.5 mm thick . The polished substrates were ultrasonic cleaned with acetone and methyl alcohol and dipped for a short time in dilute HCL to remove the oxide film. The substrates were mounted in the load lock of the high vacuum source development test stand (SDT) [3] and pumped down to $2x10^{-8}$ torr. CsBr films from 25-200 Angstroms thick were deposited with the effusion cell at 418 C by sublimation from a powder of 99.999% CsBr obtained from Alpha Aesar. The film thickness was measured with a crystal monitor inside the chamber. Subsequently, the samples were moved under vacuum to the SDT analysis chamber at $2x10^{-10}$ torr and were illuminated with a 257nm laser beam from a Coherent 300 FRED Argon laser to perform *QE* measurements.

Prior to the *QE* measurement, the uncoated samples were cleaned by exposure to the CW UV laser in the SDT for several hours. The coated samples were activated with the UV laser in the SDT. For the case of continuous wave (CW) operation at 257nm, the photocurrent was measured as a function of time illuminating the photocathode with a Coherent 300 Fred Argon laser until there was no appreciable change in current with time. For uncoated Cu samples, the *QE* increased with time due to UV cleaning of the surface and reached a maximum value of approximately $2\text{-}5x10^{-5}$ electrons/photon after several hours. For CsBr coated Cu (CsBr/Cu) samples the measured *QE* at high power density in the SDT system was more than 50-times

higher as shown in Figure 1 before and even after a 1 minute air exposure to simulate the transfer from a film growth chamber into an accelerator. Although the CsBr/Cu photocathode experienced some *QE* loss after the air exposure, subsequent exposure to 257 nm photons recovers the lost *QE* [10].

The energy spread was measured utilizing a parallel plate analyzer shown in Figure 2. The light from the 257nm laser is transmitted to the photocathode through a Au mesh (200 lines/inch) with 80% light transmission. The mesh is an anode collecting the photoelectrons making it across the retarding potential as shown in Figure 2. The experimental results produce an error function like curve [12] shown in Figure 3C which can be easily differentiated to indicate the energy spread shown in Fig. 3A, B. The gap between the photocathode and the collector mesh is about 1 mm. This geometry allows an electron collection solid angle of 2π which gives the total energy spread. We estimate the energy resolution of the analyzer to be less than 0.1 eV based upon measurements with a diamondoid film [13] low energy spread electron source. In addition, in a paper to be published elsewhere, we have validated the energy spread method with data obtained in diamondoid films with a parallel plate analyzer comparing it to data obtained with a conventional hemispherical analyzer. The electron energy spectra for a single crystal (100) Cu photocathode are shown in Figure 3 and compared to the same sample coated with a 25 angstrom thick CsBr film. Note that the CsBr film broadens the energy spread somewhat as shown in the normalized data of Figure 3B, but increases the *QE* substantially as shown in the Figure 3A (spectra normalized to *QE*). It should be mentioned that when the laser power density is increased by 5X the change in energy spread is less than 10% and operation with a large *QE* enhancement is possible at a current density greater than 100 A/cm^2 .

Two analysis techniques are used to extract the intrinsic emittance from the measurements. In the first method the width of the measured energy spectrum is used to directly obtain the intrinsic

emittance. In the second method, the measured *QE* gives the effective work function which is then used with the three-step model to compute the intrinsic emittance. The results of the two analysis methods are compared with measurements of the energy spectrum and *QE* for bare copper and for CsBr-coated copper cathodes. The experimental intrinsic emittances found using the two methods are in good agreement.

In the energy spectrum analysis the intrinsic emittance is given in terms of the excess energy, E_{excess}, of the emitted electrons

$$\frac{\varepsilon_{\text{intrinsic}}}{\sigma_x} = \sqrt{\frac{E_{excess}}{3mc^2}} \qquad (1)$$

The excess energy is the maximum kinetic energy an electron can have after escaping from the cathode. In the three-step model, the excess energy is the photon energy minus the effective work function, $E_{excess} = \hbar\omega - \phi_{eff}$. The effective work function is the cathode material work function minus the Schottky work function, $\phi_{eff} = \phi_W - \phi_{Schottky}$. The Schottky effect reduces the photoemission barrier when there's an external electric field, *E*, and is given as $\phi_{Schottky} = 3.7947 \times 10^{-5} \sqrt{E(V/m)}$ eV. In this analysis the excess energy is assumed to be equal to the full-width at half maximum of the derivative of the error function like data obtained from the energy spread analyzer shown in Figure 3A and 3B (for CsBr/Cu and 100 single crystal copper). Figure 3 A and 3B show the photoelectron spectra measured for CsBr/Cu and 100 single crystal copper. These spectra show the excess energy spread is 0.77 eV and 0.31 eV for CsBr/Cu and copper, respectively. Using these excess energies in Eqn. (1) the intrinsic emittance for single crystal copper is 0.45+/-0.05 microns/mm-rms and the emittance is 0.71+/-0.05 microns/mm-rms for CsBr/Cu.

The second analysis method uses the quantum efficiency to obtain the effective work function and from it the intrinsic emittance. Since in the three-step model the work function links the *QE* and the emittance, a consistent analysis requires using the same work function for both the *QE* and intrinsic emittance. Here the measured *QE*s are analyzed to obtain the effective work functions for (100) copper and copper with a 25 nm thick layer of CsBr, CsBr/Cu. These work functions are then used to compute the intrinsic emittance.

The effective work function can be obtained from the relation for the *QE* of metal-like photoemitters near threshold [1] assumed valid for UV activated CsBr (since it is no longer an insulator after UV activation).

$$QE = \frac{1-R(\omega)}{1+\frac{\lambda_{opt}}{\lambda_{e-e}}} \frac{\left(\hbar\omega-\phi_{eff}\right)^2}{8\phi_{eff}\left(E_F+\phi_{eff}\right)} \qquad (2)$$

Here the *QE* is given as the number of electrons per incident photon. $R(\omega)$ is the reflectivity for a photon of frequency ω, the 1/e optical absorption depth is λ_{opt} and the electron-electron scattering length is $\lambda_{e\text{-}e}$, the Fermi energy is E_F and the effective work function is ϕ_{eff}. Figure 4 shows a plot of the *QE* for a normal incidence 257 nm (4.826 eV) photon as a function of the effective work function assuming typical values for the reflectivity, the optical absorption length and the e-e scattering length and using 7 eV for the Fermi energy [14]. The *QE* curves correspond to two sets of values for the reflectivity, and the optical and the electron-electron scattering lengths have been used in the first factor of Eqn. (2). These curves are useful for estimating the systematic uncertainty of the effective work function. Specifically, the systematic uncertainty of the effective work function is approximately the horizontal distance between the red and blue lines.

The *QE* for CsBr/Cu gives an effective work function of 3.9+/-0.1eV. Copper's *QE* of 2.8×10^{-5} corresponds to 4.55+/-0.05eV for its effective work function. These work functions along with the photon energy of 4.826 eV are then used in the three-step model expression for the intrinsic emittance [1],

$$\frac{\varepsilon_{\text{intrinsic}}}{\sigma_x} = \sqrt{\frac{\hbar\omega - \phi_{eff}}{3mc^2}} \qquad (3)$$

The experimental parameters and the intrinsic emittances found with these two methods are summarized in Table I. There is good agreement between the emittances measured with the electron spectra and those determined with the *QE*. This result confirms the validity of the three-step model and its rather simple assumptions. The intrinsic emittances are listed for bare copper and CsBr/Cu cathodes as determined by the two methods discussed in the text. The experimental excess energies and the *QE*s are also listed. The intrinsic emittance errors are given using the systematic errors shown in Figure 4.

We have shown that a *QE*=4.5E-4 can be obtained with a CsBr coated copper cathode at 257nm with a very low power density of a few Watts/cm^2. The QE for higher power density is > 10^{-3} as shown in Figure 1 for a laser power density of 4×10^5W/cm^2. Therefore, the QE in a CsBr coated sample is increased by focusing the laser beam or increasing the laser power, an effect which is due to additional activation. Thus it is possible to obtain an order of magnitude QE increase relative to UV cleaned Cu targets since the QE of uncoated clean Cu targets does not change with laser power density. In addition, we have observed that the experimental energy spread for CsBr/Cu does not change with increasing power density. Therefore the increase in QE with power density while the energy spread remains constant appears to be due to an increase in the electron density of states with activation rather than to any further decrease of the work function since the energy spread is unchanged. Thus the increase in QE with power density

without affecting appreciably the energy spread is an interesting phenomenon that deserves more study and will be discussed in a future paper.

The energy spectrum for (100) Cu and CsBr/Cu photocathodes was measured with a simple parallel plate analyzer with a resolution of about .1 eV. The electron energy spectra and the QE data were separately analyzed to obtain their respective intrinsic emittances. The width of the spectra directly gives the intrinsic emittance while the QE analysis requires applying the theoretical QE given by Spicer's three-step photoemission model. The experimental emittances obtained using the two analysis techniques are in good agreement and also agree with the intrinsic emittance derived using the three-step model. The relatively small increase in emittance (μm/mm-rms) obtained for CsBr coated (0.77) relative to uncoated copper single crystals (0.42) is encouraging for FEL's applications in view of the observed large QE enhancement possible at high power density. Experiments with picosecond pulsed sources are now in progress to determine the response time of CsBr/Cu photocathodes.

References

[1] D.H. Dowell and J.F. Schmerge, PRST-AB **12**,074201(2009)

[2] Juan R. Maldonado, Piero Pianetta, David H. Dowell, John Smedley, and Peter Kneisel , Journal of Applied Physics **107,** 013106 (2010)

[3]Juan R. Maldonado**,** Zhi Liu, D. H. Dowell, Robert E. Kirby, Yun Sun, Piero Pianetta, and Fabian Pease , Phys. Rev. ST Accel. Beams **11** (2008).

[4] Juan R. Maldonado, Zhi Liu, Yun Sun, Scott Schuetter, Piero Pianetta and Fabian Pease, J.Vac. Sci. Technol. B 25(6) Nov/Dec 2007.

[5]. Zhi Liu, Juan Maldonado, Scott Schuetter, Yun Sun, Piero Pianetta and R. F. W. Pease, Appl. Phys. Lett. 2007, 90 (23).

[6] Zhi Liu, Juan Maldonado, Yun Sun, Piero Pianetta, and R. F. W. Pease, Appl. Phys. Lett. 89, 111114 (2006).

[7] Coyle, ST; Shamoun, B; Yu, M; Maldonado, J; Thomas, T; Holmgren, D; Chen, X; Scheinfein, MR; DeVore, B; Gesley, M, Journal of Vacuum Science & Technology B (Microelectronics and Nanometer Structures); March 2004; vol.22, no.2, p.501-5.

[8] J. R. Maldonado, S. Coyle, B. Shamoun, M. Yu, M. Gesley and P. Pianetta, , JVST,B 22(6), Nov/ Dec. 2004, pp 3025-31.

[9] J.R. Maldonado, S.T. Coyle, B. Shamoun, M. Yu, T. Thomas, D. Holmgren, X. Chen, B. DeVore, M.R. Scheinfen, and M. Gesley, Invited paper, Nanofabrication Technologies, Proc. SPIE Vol 5220, August 2003, pp 46-51.

[10] Juan R. Maldonado, Zhi Liu, D. H. Dowell, Robert E. Kirby, Yun Sun, Piero Pianetta, and Fabian Pease, Microelectron. Engineering 86, 529-531 (2009)

[11]. Juan Maldonado, Zhi Liu, Yun Sun, Piero Pianetta, and R. F. W. Pease ,J Vac Sci Technol Nov/Dec (2006).

[12] Juan R. Maldonado, Yun Sun, Zhi Liu, Sayaka Tanimoto, Piero Pianetta and Fabian Pease, J. Vac. Sci. Technol. B 26(6), Nov/Dec 2008.

[13] William A. Clay, Zhi Liu, Wanli Yang, Jason D. Fabbri, Jeremy E. Dahl, Robert M. K. Carlson, Yun Sun, Piero A. Pianetta, Nicholas Melosh, and Zhi-Xun Shen. Nano Letters. 9(1), 57 (2009)

[14] D. H. Dowell, F. K. King, R. E. Kirby, J. F. Schmerge and J. M. Smedley, PRST-AB **9**,063502(2006)

Figure Captions

Figure 1.- Enhancement of the *QE* of a polycrystalline copper sample measured with a CW 257 nm laser. The *QE* of the uncoated copper substrate increases a factor 50 after depositing a layer of 25 angstroms thick CsBr. The copper sample was exposed to air for one minute to simulate transfer into a vacuum system. The lower curve shows that subsequent exposure to 257 nm radiation recovers the initially lost *QE*. The laser power density used in these measurements is approximately $4x10^5$ W/cm^2.

Figure 2- Parallel plate analyzer with Au mesh. The analyzer measures the photoemission current over a 2π solid angle as a function of the retarding potential to determine the electron energy spectrum.

Figure 3.- Comparison of the energy spread of an uncoated single crystal (100) copper photocathode with the single crystal (100) Cu photocathode coated with a 25 A CsBr film when illuminated with a .8mm CW 257nm laser beam and a power density of ~3W/cm^2. A) energy spread raw data corrected by the ratio of *QE*'s of CsBr/Cu (4.5x10-4) and uncoated Cu 100 crystal (2.8x10-5) B) Same as A) but with peaks normalized to unity. C) error function shaped data obtained with the energy spread analyzer shown in Figure 2. The energy spread of the uncoated copper sample is 0.31 eV and the energy spread of the CsBr/Cu sample is 0.77 eV, both obtained from the FWHM of the derivative curve.

Figure 4: The quantum efficiency as a function of the effective work function for an incident photon of 4.826 eV. The *QE* has been computed with two sets of parameters for the reflectivity, optical absorption length and electron scattering length to estimate the systematic error of the effective work function.

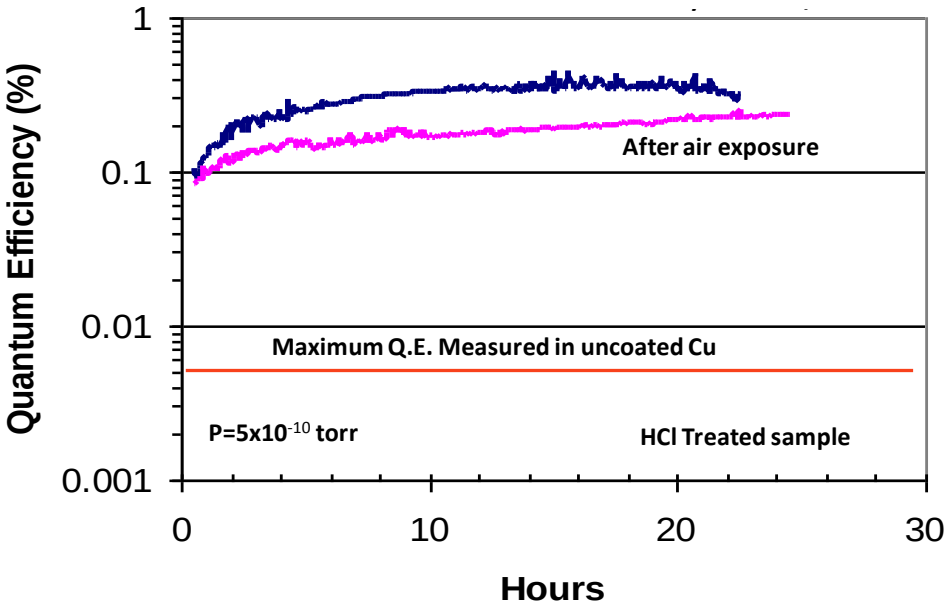
Quantum Efficiency (%)
1
0.1
0.01
0.001
0
10
20
30
Hours
After air exposure
Maximum Q.E. Measured in uncoated Cu
P=5x10-10 torr
HCl Treated sample

Figure 1

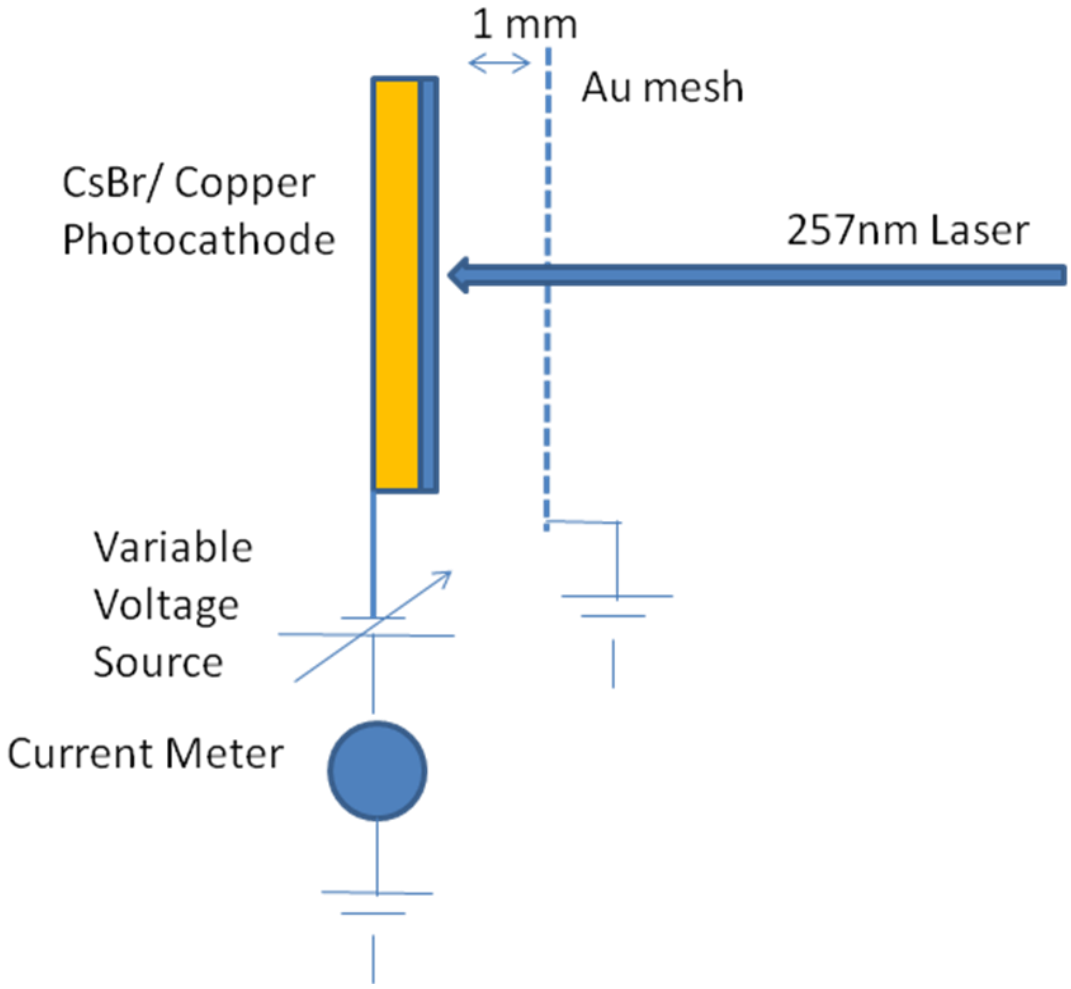
1 mm
Au mesh
CsBr/ Copper
Photocathode
257nm Laser
Variable
Voltage
Source
Current Meter

Figure 2

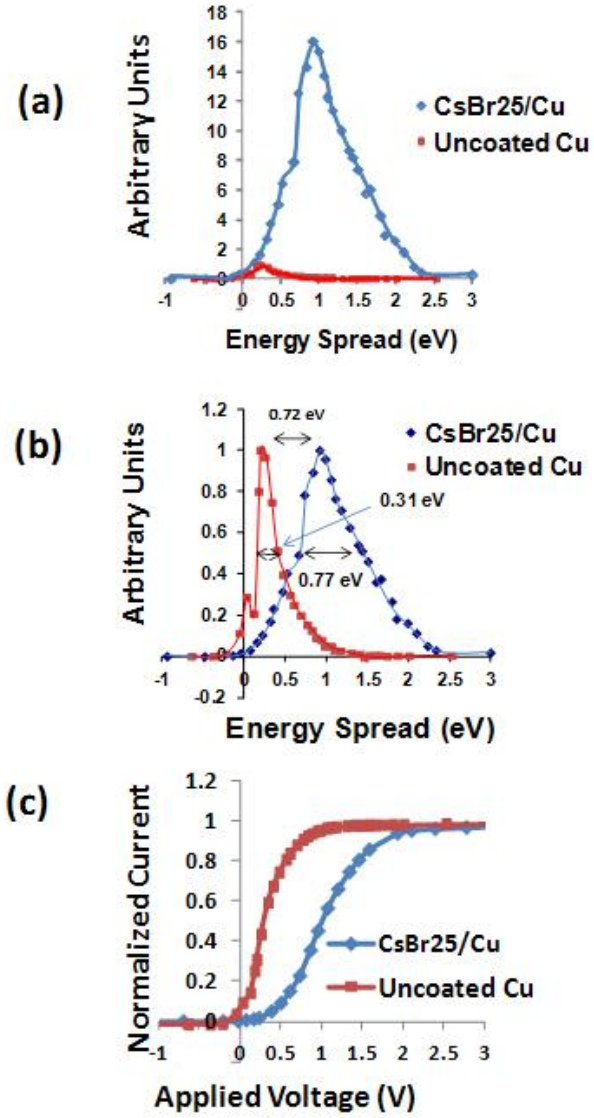
(a)
Arbitrary Units
CsBr25/Cu
Uncoated Cu
Energy Spread (eV)
(b)
Arbitrary Units
0.72 eV
0.31 eV
0.77 eV
CsBr25/Cu
Uncoated Cu
Energy Spread (eV)
(c)
Normalized Current
CsBr25/Cu
Uncoated Cu
Applied Voltage (V)

Figure 3

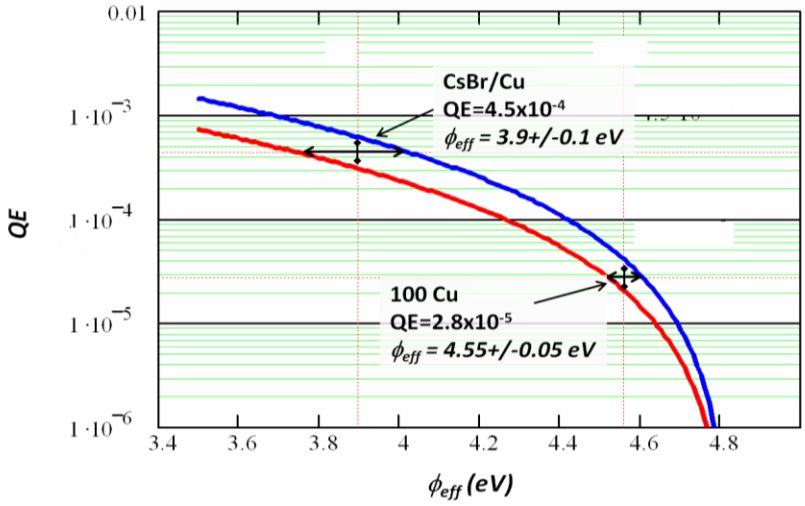
0.01
1·10⁻³
1·10⁻⁴
1·10⁻⁵
1·10⁻⁶
QE
3.4
3.6
3.8
4
4.2
4.4
4.6
4.8
ϕeff (eV)
CsBr/Cu
QE=4.5x10-4
ϕeff = 3.9+/-0.1 eV
100 Cu
QE=2.8x10-5
ϕeff = 4.55+/-0.05 eV

Figure 4

Table I

Cathode	Excess Energy (eV) from electron spectrum	$\sqrt{\frac{E_{excess}}{3mc^2}}$ (μm/ mm-rms from electron spectrum	QE expt.	ϕ_{eff} (eV) From expt. QE	$\sqrt{\frac{\hbar\omega-\phi_{eff}}{3mc^2}}$ (μm/ mm-rms. From expt. QE
Cu	0.31	0.45	2.8x10-5	4.55+/-0.05	0.42+/-0.04
CsBr/Cu	0.77	0.71	4.5x10-4	3.9+/-0.1	0.77+/-0.05