Robust CsBr/Cu photocathodes for the linac coherent light source

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(Received 5 February 2008; published 25 June 2008)

The linac coherent light source (LCLS), an x-ray free-electron laser project presently under construction at SLAC, uses a 2.856 GHz rf photocathode gun with a copper cathode for its electron source. While the copper cathode is performing well for the LCLS project, a cathode material with higher quantum efficiency would reduce the drive laser requirements and allow a greater range of operating conditions. Therefore a robust CsBr/Cu photocathode with greater than 50 times the quantum yield at 257 nm relative to the present LCLS copper cathode has been investigated. Preliminary experiments using a dedicated electron source development test stand at SLAC/SSRL are encouraging and are presented in this paper.

DOI: 10.1103/PhysRevSTAB.11.060702 PACS numbers: 29.25.Bx, 41.60.Dk, 79.60.−i, 82.50.Hp

I. INTRODUCTION

A reliable free-electron-laser–based short wavelength (1.5 to 15 Å) light source like the linac coherent light source (LCLS) [1] requires rugged photoemitters with relatively high quantum efficiency and low emittance. For a given beam current, the quantum efficiency (Q.E., electrons/photon) of the photocathode determines the required light power from the illuminating laser. Increasing the quantum efficiency can increase the operational lifetime of the laser and associated optical components which is limited by radiation damage, as well as reduce the capital equipment and operating costs. A polycrystalline Cu photocathode is currently used in the rf gun for the linac coherent light source (LCLS) injector. Copper photocathodes offer a low thermal emittance and are relatively immune to contamination. However, the quantum efficiency at 257 nm is low (in the 10^{-5} range), and requires relatively high laser pulse energies to meet the requirements of the LCLS.

Photocathodes consisting of CsBr deposited on Cr and GaN substrates have been shown [2–6] to be robust and capable of operation at relatively high current density with lifetimes (1/2 of the initial operating Q.E.) of many hundreds of hours when illuminated with 257 nm (4.8 eV) radiation either in the transmission or reflection modes [4]. Operation below the band gap of CsBr (~ 7.3 eV) was attributed [3–6] to (i) intraband states present in the CsBr layer with an energy about 3.7 eV below the conduction band, and (ii) the direct electron injection from the metal to CsBr layer. Electrons occupying those intraband states can perform transitions to the continuum with an incident photon energy of only 4.8 eV. In the case of CsBr/GaN photocathodes, due to the better alignment of the conduction bands of CsBr and GaN substrate, the direct electron injections from the GaN substrate through the CsBr film are enhanced. Thus CsBr/GaN cathodes offer a large enhancement in the quantum efficiency relative to CsBr/Cr photocathodes [5,6].

The purpose of this paper is to present preliminary experimental results on the performance of CsBr coated Cu photocathodes operating at 257 nm that may find applications in the LCLS at SLAC and at other facilities using photocathode rf guns. The CsBr coated Cu cathode shows more than a 50-fold improvement of quantum efficiency over the Cu cathode, and its quantum efficiency is not greatly affected by a brief air exposure. The LCLS drive laser design requirement for a copper cathode is 250 microjoules per pulse on the cathode. The demonstrated laser output energy exceeds 2.5 mJ. Taking into consideration the losses in the beam transport system, the LCLS drive laser currently delivers 400 μJ on the photocathode. This would be reduced to 5 microjoules for the proposed CsBr/Cu cathode. Results on the electron energy spread of the CsBr coated Cu photocathodes are under investigation and will not be presented in this paper, although an estimate for the thermal emittance is given in Sec. III. However, measurements of CsBr films coated on other substrate materials to be presented elsewhere indicate that thin CsBr films similar to the ones studied in this paper do not affect appreciably the energy spread expected from the uncoated substrate [7].

II. EXPERIMENTAL DETAILS

The experiments were performed in the source development test stand (SDT) [3] installed at SSRL for photocathode fabrication and evaluations. It consists of a loadlock chamber, a sample preparation chamber, and an analysis chamber. The solid Cu samples (1 cm^2, 1 mm thick) are machined from ASTM class one OFE Cu, then mechanically final polished with 1 μm diamond paste and 0.04 μm colloidal silica suspension. Two wet chemical cleaning procedures are utilized for the Cu samples. Some Cu samples were ultrasonic cleaned in diluted Liquid-Nox [8], followed by deionized (DI) water and methanol rinse, others were treated with HCl (to remove a possible CuO film) followed with DI water rinse.

The cleaned samples are transferred to the loadlock chamber and baked at 140°C prior to transfer to the sample preparation chamber. The Q.E. of all the uncoated Cu samples is measured before the CsBr deposition. For the CsBr coated Cu (CsBr/Cu) samples, an 18 nm thick CsBr film is deposited at 10^{-9} Torr on the Cu substrate at room temperature utilizing an effusion cell operating at approximately 400 °C. The Cu substrates are located about 10 cm from the opening of the effusion cell, and the CsBr thickness is monitored with a calibrated quartz crystal monitor. The coated sample is then transferred under vacuum to the analysis chamber with a base pressure of 5×10^{-10} Torr.

To measure the quantum efficiency of the uncoated Cu samples, they were illuminated in the reflection mode [3] (with the incident laser beam on the same side as the photoelectron collector). A 257 nm Coherent 300 Fred cw laser [9] was utilized with the beam focused on the sample at several power densities.

III. RESULTS AND DISCUSSION

The photocurrent was measured with an optically isolated amplifier with the cathode biased at 1200 V relative to the grounded electron collector (~ 600 V/cm). This electric field provides operation below the space charge limit for all measured photocurrents [10]. Since we observed large variations in the measured quantum efficiency of the uncoated Cu samples, the results presented in this paper compare the quantum efficiency value obtained for the Cu samples coated with CsBr to the maximum Q.E. value obtained in the Cu samples before coating. The observed maximum value is consistent with independent measurements on uncoated Cu cathodes at the SLAC Surface Materials Science Lab [11]. Large variations of the quantum efficiency for a solid Cu sample may be expected at 4.8 eV irradiation, since contamination, copper oxide formation, and mechanical surface stress may change slightly the work function of Cu whose value (~ 4.6 eV) is very close to the 4.8 eV incident photon energy. Therefore, for uncoated Cu samples, quantum efficiency nonuniformities may be expected in large sample areas and from sample to sample due to surface nonhomogeneities. The nonuniformity in Q.E. is expected to be reduced for CsBr coated Cu since many photoelectrons are generated from the CsBr intraband states with a center energy of about 3.7 eV below the vacuum level [3,4]. Figure 1 shows the behavior of the uncoated Cu sample with the maximum measured quantum efficiency at an incident power density of 4.8 W/cm^2. The quantum efficiency initially increases with irradiation, due to UV surface cleaning, and then levels off in about 1 h to a value of about 4.5×10^{-5}.

The results obtained after coating the Cu sample with 18 nm of CsBr are shown in Fig. 2 and compared with the maximum quantum efficiency observed in the uncoated Cu sample under similar conditions. The sample thickness was

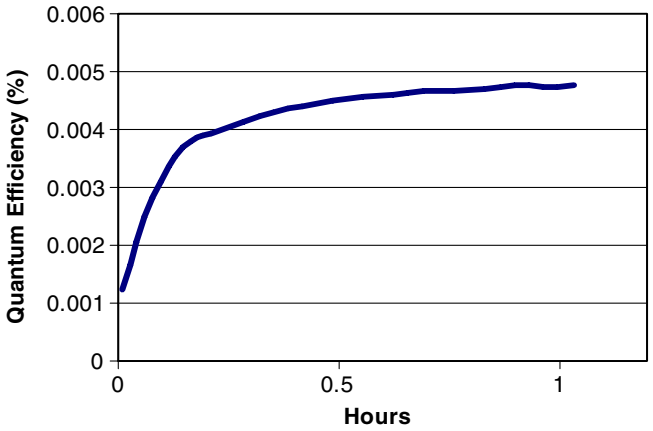

FIG. 1. (Color) Plot of the Q.E. (%) as a function of time for an uncoated Cu target illuminated at 4.8 W/cm^2 with 257 nm laser light. The initial increase in quantum efficiency is due to surface cleaning by the UV irradiation. The 257 nm UV line decomposes ozone and produces high energy O^* (activated oxygen). In addition, the 257 nm radiation is absorbed by most hydrocarbons [16].

chosen following previous work to reduce blanking effects [2,3] in e-beam lithography applications. However, thicker films may be utilized to increase the lifetime and Q.E. in the LCLS short pulse application. An increase in quantum efficiency greater than a factor of 10 is observed for the CsBr/Cu sample shown in Fig. 2. Note that it takes about 12 hours of irradiation at a relative low power density of 4.8 W/cm^2 to reach the maximum quantum efficiency. This initial behavior is expected from the creation of color centers in the material and the transfer of Cs to the surface as described in Refs. [3–6], and differs from the UV surface cleaning shown in Fig. 1. The Q.E. of all the Cu

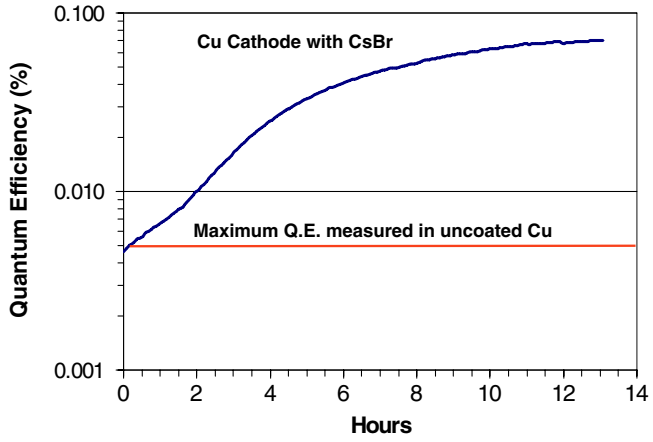

FIG. 2. (Color) Quantum efficiency (%) of CsBr/Cu sample irradiated at 4.8 W/cm^2 versus time before exposure to air. The initial activation (about 12 hours to reach a maximum Q.E.) of the CsBr/Cu photocathode at low power density is shown in the figure. The maximum Q.E. obtained for an uncoated Cu sample is shown for comparison.

samples was measured before coating. However, UV cleaning before CsBr coating is not necessary for Q.E. enhancement. Some Cu samples showed negligible Q.E. before coating and a large value of Q.E. shown in Fig. 2 after coating with CsBr. The initial behavior of the Q.E. can be accelerated at higher power densities as described below.

To demonstrate the robustness of the CsBr/Cu photocathodes, we simulated transferring the sample from the SDT deposition chamber to the LCLS system. For this purpose, the CsBr/Cu sample was exposed to air for 1 min in the loadlock chamber of the SDT system. Subsequently, the sample was pumped down to the 5×10^{-8} Torr range and baked overnight at 140 C in the loadlock chamber before transferring into the SDT analysis chamber at 5×10^{-10} Torr.

The results presented in Fig. 3, after air exposure for 1 min and 140 C baking, indicate about a factor of 10 increase in quantum efficiency for the CsBr/Cu relative to the uncoated Cu sample. Similar results were obtained without the baking step as described below.

Figure 4 shows the behavior at high power density (4×10^5 W/cm^2 with the 257 nm cw laser) of the CsBr/Cu sample previously exposed to air. The sample was previously irradiated at somewhat lower power density for 1 h before the data shown in the figure was taken. We observe from the figure that operating at high current density after 1 h activation, the maximum quantum efficiency can be reached in a few minutes. In a free-electron laser source, activation can be performed with a cw laser previous to pulse operation. Results presented in Ref. [2] indicate that the Q.E. loss after stopping the illumination for a few minutes in an activated CsBr/Cr sample is less than 5%. This behavior was referred to in this reference as the blanking effect, which is important for nanolithography

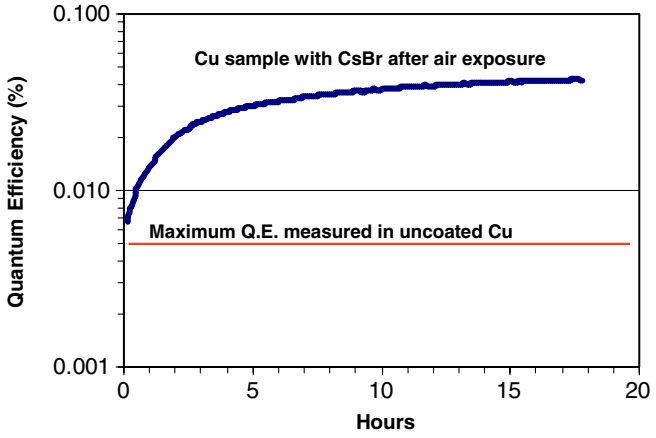

FIG. 3. (Color) Quantum efficiency (%) of CsBr/Cu exposed to air for 1 min and baked at 140 C. The sample was illuminated at 4.8 W/cm^2 with a 257 nm laser. The maximum quantum efficiency observed in an uncoated Cu sample is also shown in the figure.

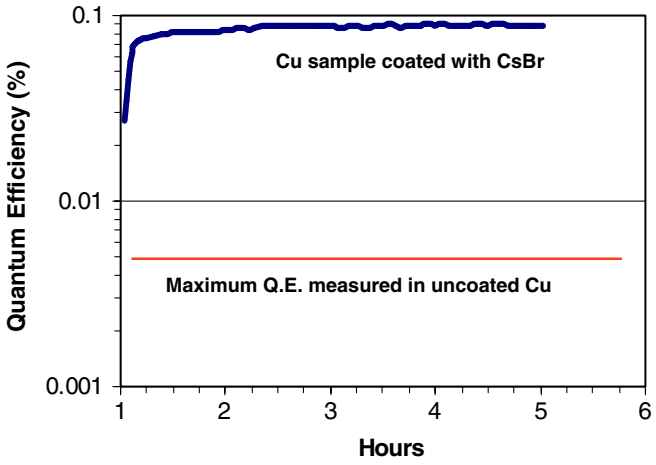

FIG. 4. (Color) Results of quantum efficiency (%) obtained with a copper target coated with an 18 nm thick CsBr film. The sample was exposed to air for 1 min and baked at 140 C overnight before the data was taken. The data shown was obtained at 4×105 W/cm^2 with a 257 nm laser, an accelerating voltage of 1200 V, and a chamber pressure of 5.6×10^{-10} Torr. The maximum Q.E. for the uncoated Cu sample presented in Fig. 1 after surface cleaning by UV exposure at 257 nm is also shown for comparison.

applications where the electron beam must be switched on and off during pattern writing. The results for the CsBr/Cu are compared to the maximum Q.E. observed in the uncoated Cu sample in Fig. 4, indicating almost a 20-times increase in quantum efficiency for the CsBr/Cu sample.

Surface preparation of the Cu substrates before CsBr deposition is important. An example is shown in Fig. 5. In this case, the Cu sample was treated with 50% HCl for a few seconds (to remove any possible CuO film) after the standard cleaning described above, and then washed with DI water. The results obtained after CsBr deposition and subsequent exposure to air for 1 min without the bakeout step are also shown in Fig. 5. The measured quantum efficiency obtained before and after air exposure shown in Fig. 5 increased considerably relative to the data shown in Fig. 4 for the case of surface cleaning without HCl.

The quantum efficiency of the CsBr/Cu sample presented in Fig. 6 was measured for a long time interval $>$250 hours. The data was obtained in open loop without correcting for laser intensity fluctuations and external vibrations that caused jumps in the quantum efficiency due to laser spot motion. No indication of imminent failure or rapid quantum efficiency decay was observed. From our experience on CsBr on other metal substrates (Cr and Mo), a lifetime of hundreds of hours can be obtained from a single spot with >100 A/cm^2 current density. The lifetime of the CsBr/Cu cathode is not expected to be an issue under a lower current density. However, two factors may contribute to the decay of the lifetime: the thermal degradation of the cathode at higher current density (this may be

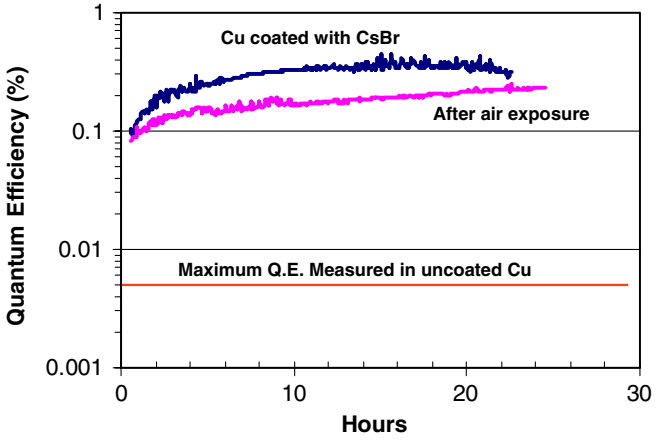

FIG. 5. (Color) Quantum efficiency (%) at 257 nm of the CsBr/Cu sample as deposited and after exposure to air for 1 min and pumped down to low pressure without bakeout. The CsBr/Cu sample was treated with HCl and deionized water prior to CsBr deposition. The sample was irradiated at 1×10^5 W/cm^2 with a 257 nm laser at a pressure of 5×10^{-10} Torr, and shows a greater quantum efficiency than the untreated sample shown in Fig. 4. The quantum efficiency enhancement is more than 50-times relative to the maximum value obtained in the solid Cu target. The maximum Q.E. for the uncoated Cu sample is shown in the figure for comparison to the CsBr/Cu sample.

ameliorated with proper cooling) and the depletion of the CsBr thin film after a long period of operation. Of course, thicker CsBr films may be utilized to extend the lifetime. Alternatively, since the CsBr film is not affected in the unexposed areas [4], a movable photocathode designed to expose a fresh CsBr/Cu area may be utilized to extend the lifetime if needed.

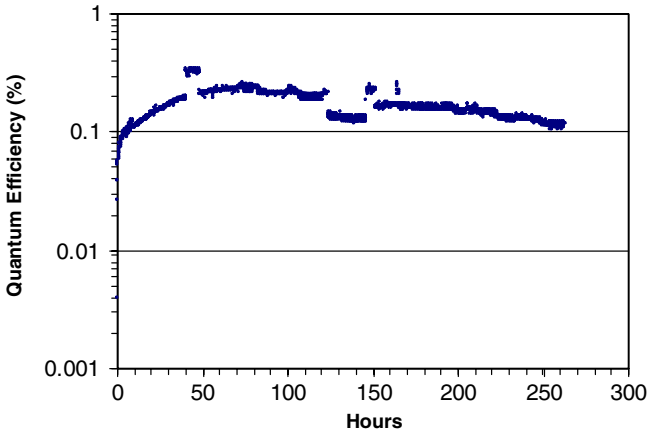

FIG. 6. (Color) The quantum efficiency of a CsBr/Cu photocathode was measured in open loop for a relatively long time >250 hours without great deleterious effects. Spikes and offsets were due to laser fluctuations and external vibrations that caused jumps in the quantum efficiency due to illuminated spot motion. The laser power density was $\sim 5 \times 10^5$ W/cm^2 for a spot size of 1.5 μm diameter. The behavior of the Q.E. with time is explained in Refs. [2–6] in terms of intraband excitations, direct electron injection from metal substrates, Cs migration, and contamination.

One of the advantages of the CsBr based photocathodes relative to other materials like Cs_2Te considered for e-beam lithography tools is the very small ($< 10\%$) spot growth effect [12]. The electron emission spot size changes very little during the lifetime of the cathode for laser illumination spot sizes as small as 300 nm (current density >100 A/cm^2). The Cs diffusion is confined only to the illumination area utilizing relatively low thermal conductivity sapphire substrates. For the LCLS application, with large illumination areas with a high thermal conductivity substrate like Cu, the spot growth effect should be much less than 10%.

Concerning the emission homogeneity and behavior in high rf fields, this is not the first use of CsBr in a rf gun. There have been previous applications in which a CsBr overcoating was applied to Cs_2Te as a protection against vacuum contaminates [13]. In these cases, the uncoated Q.E. of Cs_2Te was reduced approximately a factor of 5, but the lifetime was extended from days to months. Otherwise the Cs_2Te/CsBr coated cathode operation in the rf gun was unchanged. Therefore we expect the CsBr/Cu cathode to have similar or better lifetime performance in an rf gun.

We also expect the time response to be similar to that of copper. Since CsBr/Cu photoemission is the result of direct injection of electrons from the copper into the CsBr layer, the photoemitted electrons should have the same subpicosecond response time as the metal substrate. In any case, the response time should be no different for similar alkali cathodes such as Cs_2Te, which is known to be subpicosecond and used extensively in rf guns [14,15].

While there are plans to measure the thermal emittance, an estimate can be made based upon the photoluminescence spectrum. The spectrum for CsBr/Cr is 0.4 eV wide (FWHM) [3]. The width of the photoluminescence spectrum corresponds to the width of the electron density of states responsible for photoemission and hence the thermal emittance. Therefore we can estimate the thermal emittance for CsBr/Cu by assuming the same width as CsBr/Cr and using this energy spread of 0.4 eV in the relation for the emittance. For emission from copper only, the electron energy spread is simply the difference between the photon energy and the work function. In the case of LCLS, this difference is 4.866 eV − 4.6 eV = 0.27 eV. As the emittance is proportional to the square root of the electron energy spread, the thermal emittance of CsBr/Cu relative to that of copper becomes

$$\frac{\text{CsBr/Cu}}{\text{Cu}} \approx \sqrt{\frac{0.4\ \text{eV}}{0.27\ \text{eV}}} = 1.2.$$

Therefore, the thermal emittance of CsBr/Cu should not be much larger than that of copper alone.

IV. SUMMARY

The results presented in this paper represent a step toward realizing a robust electron source for the LCLS rf gun and related applications, requiring significantly lower laser pulse energy than the uncoated Cu cathodes presently being used. We have demonstrated that CsBr/Cu photocathodes offer higher quantum efficiency ($> 50 \times$) than uncoated Cu cathodes with a relatively long lifetime. These CsBr/Cu photocathodes are robust, allowing them to be exposed to air for a short time during the transfer process from the fabrication chamber to the LCLS system without great changes in performance. There is work still to be done mounting the photocathode in an actual gun to demonstrate the transverse emission homogeneity, time response, and behavior in ultrahigh rf fields. However, due to the very thin (~ 20 nm) CsBr films utilized in the proposed structure and other considerations discussed above, we do not expect large deviations from the uncoated Cu substrate.

ACKNOWLEDGMENTS

The work of the first author of this paper was partially funded by a DARPA Grant No. N66001-07-1-2045 administered by SPAWAR.

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