Enhancement of photoemission from and postprocessing of K_2CsSb photocathode using excimer laser

Erdong Wang,^{1,*} Triveni Rao,¹ and Ilan Ben-zvi^{1,2}

¹Brookhaven National Laboratory, Upton, New York 11973, USA

²Stony Brook University, Stony Brook, New York 11794, USA

(Received 20 September 2013; published 20 February 2014)

The high quantum efficiency at visible wavelengths of alkali-antimonide photoemissive materials, such as K_2CsSb , makes them excellent potential photocathodes for high-current applications. We have developed a technique of using an ultraviolet laser to clean the cathode's substrate and thus enhance the photoyield of a K_2CsSb photocathode subsequently deposited on the substrate. We have shown that the quantum efficiency of the cathode from the laser-exposed substrate can be at least 50% higher than that of an unexposed surface. We have also formulated a nonthermal technique for completely removing the cathode from the substrate while preserving an ultrahigh vacuum to assure the regrowth of the cathode. The bialkali cathode is dissociated and then removed completely upon 10 s exposure to a 248 nm laser beam with 3.5 mJ/mm^2 of energy density at a 30 Hz repetition frequency. Here, we discuss these experimental results and their potential applications. We also describe applications of this technique to reduce the beam's halo and its emittance.

DOI: 10.1103/PhysRevSTAB.17.023402

PACS numbers: 07.77.Ka, 85.60.Ha, 42.62.Cf, 79.60.-i

I. INTRODUCTION

Several photoinjector applications require photocathodes capable of delivering average currents up to 100 mA with high quantum efficiency (QE) at visible wavelengths, over a prolonged time period [1,2,3]. A high initial QE also will increase the time over which the QE is above the minimum acceptable value, thereby lengthening the machine's operation time. Alkali antimonide cathodes show great promise towards meeting these first two requirements. The high QE is obtained by fabricating the cathode on a clean substrate in a ultrahigh vacuum (UHV) environment and transporting it to the injector through a UHV load-lock system. With conventional vacuum practices, vacuum levels in the range of 10^{-11} torr are obtained routinely. Typically, the substrate is heated to remove the adsorbed gases and oxide layers. A QE of up to 12% at 532 nm was obtained in the Average Power Laser Experiment (APLE) project at Boeing, and have similar values at Cornell University and BNL [4,5,6]. The achievement of the third requirement for the cathode, a long operational lifetime, is dictated by the vacuum contamination and/or ion-back bombardment of the cathode in the gun environment. A K_2CsSb cathode operating in a normal conducting rf gun, delivering 32 mA average

current, had a lifetime of 1–10 hr that was limited by vacuum conditions surrounding the cathode [4,7]. In Cornell University's dc gun, the K_2CsSb cathode delivered up to a 60 mA average current with a 30-hr 1/e lifetime, and the NaKSb photocathode delivered up to 65 mA average current with a 66-hr 1/e lifetime [8,9]. Similarly with the K_2CsSb cathode in a 250 kV dc gun at JLab, average current densities of 166 mA/mm^2 have been generated with no degradation of the QE [10]. At the end of the cathode's life, in the dc gun, the entire substrate, along with the spent cathode, is replaced with a fresh one. In some rf guns, such as the APLE normal-conducting rf injector or in BNL's energy recovery linac gun, the cathode is an integral part of a large insertion section. In the former, the spent cathode was removed by heating the cathode region to 600°C and a new cathode was fabricated on the resulting surface [4]. Heating the substrate has several limitations: (1) At these high temperatures, the surrounding volume is heated by radiation, thus increasing the outgassing surface area, and thereby degrading the vacuum; (2) thermal cycling from such high temperatures to room temperature increases the overall fabrication time; (3) some of the surface oxides cannot be removed by heating; (4) in some gun designs, the cathode is an integral part of a large or massive insertion device and heating it poses significant engineering problems; and 5) the heating element is an integral part of the UHV system so that servicing it is difficult without breaking the vacuum.

Laser cleaning, in which high-intensity picoseconds laser pulses were used to ablate the metal's surface and to expose a pure metal cathode, has been used successfully

*Corresponding author.
wange@bnl.gov

Published by the American Physical Society under the terms of the *Creative Commons Attribution 3.0 License*. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

with metallic photocathodes, such as copper and magnesium [11,12,13]. In the following sections, we describe our usage of an excimer laser to clean the substrate's surface and enhance the final QE of a K_2CsSb cathode. Then, we describe how we employed the same excimer laser to totally remove this cathode. In both instances, the roughness of the substrate was not increased.

II. EXPERIMENTAL ARRANGEMENT

The cathode-fabrication system, whose arrangement is shown in Fig. 1, consists of four vacuum sections: (a) transfer arm 1, (b) transfer arm 2, (c) deposition chamber, and (d) storage chamber. The storage chamber is a load-lock system, intended for transferring the cathode from the fabrication area to the gun; it can hold up to five samples at a time. There are two viewports in the load-lock section, one of which is used for cleaning the substrate by irradiation with the excimer laser. With transfer arm 1, we transfer the substrate/sample between the storage chamber and the preexisting cathode fabrication system, the latter comprising transfer arm 2 and the deposition chamber. The deposition chamber is equipped with a residual gas analyzer (RGA), a quartz crystal monitor, and multiple viewports for observation and laser irradiation of the cathode. The RGA monitors the partial pressures of the residual gases in the chamber during evaporation. Alkali metal sources (20 mg Alvacat: type s for potassium and type v for cesium) and antimony (99.999% purity pellets from Goodfellow) are used for the deposition.

These sources are inserted through gate valves into the main chamber during evaporation but are isolated from each other and from the main chamber to prevent cross-contamination of the sources. The alkali metals are evaporated through direct resistive heating of the source, while the antimony pellets are kept in a ceramic crucible that we heat by passing a current through a tungsten boat holding the crucible. Transfer arm 2 positions the substrate above each of the sources in the main chamber. During evaporation, the temperature of the substrate can be raised to the required value through the resistive heating of the substrate holder, and hence, the substrate. A crystal monitor is used to establish the evaporation rate of the metal sources during fabrication of the cathode. The QE of the sample in the deposition chamber is measured by irradiating the cathode with either a 5 mW, 532 nm laser or a white light dispersed by a spectrometer. The optical power is measured by a power meter calibrated for the appropriate wavelength. The substrate is electrically isolated from its surrounding and can be biased up to 90 V. The charge leaving the substrate is measured using an electrometer (Keithley Model 6485). The vacuum level of all the four sections is maintained at 10^{-11} Torr by ion and NEG/TS pumps.

The excimer laser used for these experiments was a KrF laser (model MSX-250) that can deliver laser pulses at 248 nm wavelength, with a repetition frequency of up to 300 Hz and pulse duration of 3.5 ns. We selected a 20 cm focal length at the appropriate position from the laser to

FIG. 1. K_2CsSb cathode fabrication system. (a) Transfer arm 1; (b) transfer arm 2; (c) deposition chamber; (d) storage chamber and substrates; (e) sources chamber; (f) crystal monitor; and (g) UV laser.

produce a 1.95×2.86 mm FWHM spot on the substrate for laser cleaning the substrate and 1.1×1.6 mm spot size on the cathode for removing it.

III LASER CLEANING OF THE SUBSTRATE

Three high-purity, well-polished molybdenum substrates were cleaned with alcohol and then acetone, in an ultrasonic bath for 15 min each. Then, the substrates were dried in a stream of nitrogen; each substrate was mounted on a copper puck and then inserted into the storage chamber, giving us three samples for measurement. Once the system was baked and pumped to 10^{-11} Torr, sample 1 was moved from the storage chamber to transfer arm 1, the sample, then positioned under the viewport, and the substrate was exposed to the excimer laser. To establish the optimal energy density of the laser for cleaning the substrate, we tried four densities: 1.22 mJ/mm^2 , 0.98 mJ/mm^2 , 0.77 mJ/mm^2 , and 0.36 mJ/mm^2 on four separate spots on the same sample. Each spot was exposed to 20,000 shots at a repetition rate of 300 Hz. During this step, the base pressure in the region increased up to low 10^{-9} Torr, as measured at the storage chamber. However, the pressure dropped down to the base value of 10^{-11} torr within a few seconds after turning the laser off. Figure 2 (left) is a photograph of sample 1. There is no visible variation between the laser exposed (a) and unexposed areas (b). We used this sample for measuring the surface roughness before and after laser exposure. On sample 2, after exposure to the laser as described, we grew an alkali antimonide cathode using the standard K_2CsSb procedure for cathode fabrication: $100 \text{ \AA}$ Sb at a substrate temperature of 80°C , and $200 \text{ \AA}$ K at 140°C , followed by Cs evaporation, during

FIG. 3. Map of photoyield of an entire sample. The peaks of the QE occur at laser-exposed locations. The laser-exposed area is bounded by a rectangular box. The numbered positions are related to different laser-energy densities irradiating this area.

which we monitored the electron yield using a 532 nm, 5 mW laser. We stopped Cs evaporation when the QE reached the maximum value. Immediately afterwards, the QE decayed by 30% but stabilized over 5 hr. The measurements described below were executed in this steady-state condition. We grew the cathode over the entire substrate, covering areas both exposed and unexposed to the laser. The photograph of sample 2 is shown in Fig. 2 (right).

Figure 3 shows the photoemission map of the photoyield from the sample obtained by scanning the entire sample; it

Sample 1

Sample 2

FIG. 2. Photograph of the molybdenum substrate (left: sample 1) and the prepared K_2CsSb photocathode (right: sample 2). On sample 1 at left (a) laser exposed region of molybdenum substrate and (b) nonlaser exposed region of the molybdenum substrate. The darkening seen on sample 1 is due only to the angle of illumination during photography. On sample 2 at right (a) fork holding sample 2, on the copper puck, (b) copper puck, and (c) molybdenum substrate. Both the copper puck and molybdenum substrate are coated with the cathode material. The cathode on the laser-exposed region is clearly visible as a grey area bounded by the rectangular box.

FIG. 4. QE line scan through the center of the laser-exposed spots. All three figures, (a)–(c), present a line cut of QE along the y axis across the center of the laser-exposed spots for energy densities, respectively, of 1.22 mJ/mm², 0.98 mJ/mm², and, 0.77 mJ/mm². The low QE of (a) at 0 mm resulted from the shielding of the sample by a screw in the sample holder during evaporation.

shows that the QE of laser-exposed locations is higher than that of the unexposed locations. The average QE of laser-exposed location is 6%. Here, the QE is defined by the ratio of the number of electrons emitted to the number of incident photons at a given photon energy. Our previous

tests show that the QE of alkali antimonide cathodes typically is around 3%–4% when grown on a molybdenum substrate heated to 200° C for 4 hr. The cathode grown on a substrate exposed to a UV laser gave a 50%–100% higher QE.

FIG. 5. (a) SEM and (b) AFM images of laser-exposed and nonlaser-exposed molybdenum sample. (a) SEM and (b) AFM: images of laser-exposed area. The average roughness in 100 μm^2 ranges from 3.7 to 4 nm. (c) SEM and (d) AFM: images of nonlaser-exposed area. The average roughness in 100 μm^2 is comparable to that of the exposed area.

We deduced the density of laser energy needed for optimizing the cathode's performance from the 1D plots of the QE scan along the y axis of Fig. 4 at x locations designated as 1, 2, and 3. The QE for the area exposed to an energy density of 0.36 mJ/mm^2 is nearly unchanged from that of the unexposed region. The rest of the exposed regions show a dramatically increased QE.

Figure 4 displays the QE line scan through the center of the laser-exposed spots. The FWHM laser spot size of the laser beam at 532 nm, used for QE measurement scan, is 0.3 mm. As is evident, the QE of the laser-exposed location is at least 50% higher than the unexposed region. The maximum QE was obtained at a location where the laser's energy density was 0.77 mJ/mm^2 [Fig. 3(c)]. In addition, variations of QE within this exposed area are smaller than in the other two exposed locations. The rms variation of QE for the three exposed areas (a), (b), and (c) is 0.159%, 0.031%, and 0.015%, respectively, calculated over the central 2.5 mm section of the exposed area, and the rms variation is defined by $R_q = \sqrt{\frac{1}{n} \sum_{i=1}^n (y_i - \bar{y})^2}$. A cathode with uniform photoemission can be fabricated by raster scanning the laser over the entire substrate. We confirmed this by growing a 6% QE cathode with rms variation of 0.028% over 70 mm^2 on a laser-cleaned Ta substrate.

The substrate roughness of the photocathode's emission surface plays an important role in the thermal emittance of the photoelectrons [14,15] especially when operated in a high gradient regime. Hence, should laser exposure increase the roughness of the substrate, it will adversely affect the emittance of the electron beam at the cathode. We compared the surface roughness of exposed and unexposed regions of sample 1 using the scanning electron microscope (SEM, JEOL 6500) and atomic force microscopy (AFM, Nanosurf Easyscan 2). Figure 5 shows the findings. The "as received" molybdenum sample has an average roughness ranging from 3.7 to 4 nm in a $100 \text{ }\mu\text{m}^2$ area surveyed by the AFM. Such measurements on the exposed region suggest that laser exposure did not change the substrate's surface roughness, and the topography of the original metal surface was preserved.

Heating the metal substrate for a long time can enhance the diffusion of surface oxygen into the bulk, thereby reducing the oxygen concentration at the sample's surface [16]. However, as mentioned earlier, this technique is time consuming and degrades the UHV. Laser radiation is highly efficient in removing the oxidized surface layer of the substrate and other surface contamination. Typically, for a 1 cm diameter sample, laser radiation will take 20 min, compared to 2–4 hr for a thermal process, not including the time the sample takes to return to room temperature.

IV LASER REMOVAL OF THE CATHODE

We used a similar arrangement as that described above for removing the K_2CsSb photocathode. The laser removal

of the cathode was executed in the deposition chamber. The sample, number 3, was grown with 200 Å thick Sb, 400 Å thick K, and a Cs thickness optimized for maximum QE. A collinear green laser with spot size smaller than that of the UV laser was used to measure the change in QE. Figure 6(a) is a schematic of the arrangement of the lasers and the cathode. During the irradiation, the RGA recorded the partial pressures of the residual gas in the vacuum chamber.

We used various energy densities and repetition frequencies of the excimer laser for removing the cathode from sample 3; the results are shown in Fig. 6(b). With an energy density of 3.5 mJ/mm^2 and repetition rate higher than 30 Hz, the photocurrent decreases to background within 10 sec (total flux of 1×10^{18} photons/ mm^2). With a value of 2.32 mJ/mm^2 , the QE was 12% of the initial value after 100 sec (8×10^{18} photons/ mm^2), indicating the cathode was not completely removed. This flux-dependent behavior can be used to manipulate the profile of the electron beam accurately either by the exposure time and/or the laser's energy density.

As seen in Fig. 6b, the QE versus flux data fits well with to a dual-exponential decay function, one fast, with decay constant of T2, and another slow, with a decay constant of T1, implying that two mechanisms may contribute to removing the cathode. The change in the decay constants with the average power density indicates that both T1 and T2 have similar functional dependence on the laser's average power density (product of energy density and repetition rate) and reach an asymptote at a power density of $\sim 90 \text{ mW/mm}^2$.

We examined the laser-irradiated sample using AFM and SEM energy-dispersive x-ray spectroscopy (EDX). The typical average roughness in the area where the cathode was removed was 3.7–4.1 nm in $100 \text{ }\mu\text{m}^2$, similar to a fresh molybdenum substrate. Figure 7 shows the chemical composition on laser-exposed and nonexposed areas. In EDX, we used 5 keV electron beam to determine the surface's elemental composition. The thickness of the alkali antimonide cathode layer was approximately 1000 nm. The electron beam is energetic enough to penetrate into the substrate and provide an elemental signal from the entire cathode layer.

After laser exposure, the peaks for Sb, K, and Cs completely disappeared from the exposed region indicating that the alkali antimonide cathode was removed completely by the excimer laser. The EDX spectrum at the edge of the exposed areas shows the presence of only Sb, and not K or Cs, leading to two conclusions: (1) the excimer laser dissociates the molecule K_2CsSb to its constituent elements, and (2) the alkali metals leave the substrate completely while a fraction of Sb is redeposited at the edge of the exposed area. This dissociation theory is supported by the emergence of signals for K, Cs, and Sb in the RGA at the initial stages of laser exposure. The

FIG. 6. (a) Schematic of arrangement of lasers and cathode for removing the cathode; (b) change in the photocurrent as a function of cumulative photon flux for different laser-energy densities and repetition rates. The data are fitted into dual-exponential decay functions. The decay constants are labeled in the legends for the plots.

FIG. 7. The SEM image (left), and EDX signal (right) of exposed (*) and unexposed (+) areas of the cathode. The rectangles in the picture on the left correspond to two overlapping locations on the cathode exposed to the laser. In the EDX on right, the red curve is taken from spot after laser exposure (*) and the blue curve is taken at an unexposed location (+). The characteristic x-ray locations of elements such as Mo, Sb, K, Cs, and O are marked on EDX spectrum.

dissociation energies of the alkali-antimonide bond are between 1.28 and 1.52 eV [17]. Complete dissociation to constituent elements requires 3 times this laser energy [18]. Hence, the probability is very high for dissociating the molecular bonds by an excimer laser whose photon energy is 5 eV. The unexposed region still retains its high QE indicating that the alkali antimonide cathode remains in the unexposed region. Because the dissociated metals are ejected in the direction perpendicular to the sample's surface under laser ablation [19], they are not expected to be redeposited on the rest of sample. This is supported by the similarity of the EDX traces very close to, and farther away from, the laser exposed area.

The compositions of exposed molybdenum are the same as that of the fresh polished molybdenum sample. The boundary of laser exposure is clean where two laser spots overlap. So a larger cathode area could be removed by raster scanning the laser over the area to be cleaned. Although this method was tested using an excimer laser, we feel that other UV laser beams, for example, frequency-quadrupled radiation from Yttrium-based lasers, will be equally effective. However, we plan to investigate the wavelength dependence of the process in future studies.

This technique can be used to resolve several problems associated with alkali antimonide cathodes in high current photoinjectors. Once the QE of the cathode decays below the design value, by irradiating it with the UV laser, we can clean the substrate and remove the cathode simultaneously before fabricating a new cathode on the same substrate without any degradation of the UHV system. Also, in the photoinjector, the halo of the laser beam or scattered laser beam will generate an electron-beam halo that reduces the beam's quality and raises pressure in the gun considerably [20]. By customizing the cathode area exposed to the UV laser, the high QE cathode area could be exactly matched to the size of the electron beam. Since the halo of the laser beam illuminates the low QE substrate (even the non-emitting part), unwanted electron beams are eliminated. Furthermore, complicated laser patterns can be imposed on the cathode to remove selected sections, thereby providing spatially shaped electron beams that can be used either in conjunction with a transversely shaped laser beam or instead of shaping the transverse laser.

V. SUMMARY

We showed that by irradiating a substrate by an excimer UV laser we improve significantly the quantum efficiency of an alkali antimonide photocathode deposited on the substrate. We also demonstrated that we can remove this cathode with the excimer laser in seconds to minutes (depending on the area). This technique is faster than high-temperature treatment, less demanding on cathode design, and less disruptive to the vacuum conditions. Both predeposition cleaning and photocathode material removal preserved the substrates' initial roughness. We studied the

required energy densities and repetition rate of the excimer laser for cleaning the substrate and for removing the photocathode material. This technique can be employed to reduce the electron beam's halo and emittance.

ACKNOWLEDGMENTS

This work was carried out at Brookhaven Science Associates, LLC under Contract No. DE-AC02-98CH10886 with the U.S. DOE.

- [1] B. M. Dunham *et al.*, in *Proceedings of the 16th IEEE International Pulsed Power Conference, Albuquerque, NM* (IEEE, New York, 2007), pp. 1224.
- [2] V. N. Litvinenko *et al.*, in *Proceedings of the 21st Particle Accelerator Conference, Knoxville, TN, 2005* (IEEE, Piscataway, NJ, 2005), pp. 2242.
- [3] L. B. Jones *et al.*, *AIP Conf. Proc.* **1149**, 1084 (2009).
- [4] D. H. Dowell, S. Z. Bethel, and K. D. Friddell, *Nucl. Instrum. Methods Phys. Res., Sect. A* **356**, 167 (1995).
- [5] L. Cultrera *et al.*, *Phys. Rev. ST Accel. Beams* **14**, 120101 (2011).
- [6] T. Rao *et al.*, in *Proceedings of the Free Electron Laser Conference, New York, 2013* [<http://accelconf.web.cern.ch/AccelConf/fel2013/papers/tupso63.pdf>].
- [7] D. H. Dowell *et al.*, *Appl. Phys. Lett.* **63**, 2035 (1993).
- [8] B. Dunham *et al.*, *Appl. Phys. Lett.* **102**, 034105 (2013).
- [9] L. Cultrera, S. Karkare, B. Lillard, A. Bartnik, I. Bazarov, B. Dunham, W. Schaff, and K. Smolenski, *Appl. Phys. Lett.* **103**, 103504 (2013).
- [10] T. Rao *et al.*, *Accelerator Detector R&D meeting, Annapolis, MD, 2011*. [http://science.energy.gov/~media/bes/suff/pdf/2011_Accelerator_Detector_RD_PI_Meeting_files/Poster_Presentations/Multi_Alkali_Cathode_Rao.pdf]
- [11] T. Rao, J. Fischer, and T. Tsang, *J. Appl. Phys.* **69**, 3291 (1991).
- [12] L. Cultrera, G. Gatti, F. Tazzioli, A. Perrone, P. Miglietta, C. Ristoscu, S. Orlanducci, and A. Fiori, *Appl. Surf. Sci.* **253**, 6531 (2007).
- [13] F. Zhou, A. Brachmann, F.-J. Decker, P. Emma, S. Gilevich, R. Iverson, P. Stefan, and J. Turner, *Phys. Rev. ST Accel. Beams* **15**, 090703 (2012).
- [14] T. Vecchione, I. Ben-Zvi, D. H. Dowell, J. Feng, T. Rao, J. Smedley, W. Wan, and H. A. Padmore, *Appl. Phys. Lett.* **99**, 034103 (2011).
- [15] S. Karkare and I. Bazarov, *Appl. Phys. Lett.* **98**, 094104 (2011).
- [16] G. Ciovati, *Appl. Phys. Lett.* **89**, 022507 (2006).
- [17] P. Dolizy and F. Groliere, *J. Phys. D* **19**, 687 (1986).
- [18] J. Clayton and W. Bates, "Basic Studies of High Performance Multialkali Photocathodes," *Stanford University, Department of Materials Science and Engineering*, 1979, pp. 87.
- [19] K. Chen, J. Leboeuf, R. Wood, D. Geohegan, J. Donato, C. Liu, and A. Puretzky, *Phys. Rev. Lett.* **75**, 4706 (1995).
- [20] C. Hernandez-Garcia *et al.*, *Proceedings of the 21st Particle Accelerator Conference, Knoxville, TN, 2005* (IEEE, Piscataway, NJ, 2005), pp. 3117.