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A high current density GaN/CsBr heterojunction photocathode with improved photoyield

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A 4 fold improvement in photoyield has been observed in GaN films coated with CsBr films (CsBr/GaN) relative to CsBr/Cr photocathodes reported in previous studies. A model is presented involving photoemission from intraband states in the CsBr film and direct electron injection through the CsBr film from the GaN substrate. The lifetime of the films at high current density >90A/cm^2 is limited by the temperature rise in the GaN films caused by the high photon absorption at 257nm. The lifetime can be improved utilizing a cooled substrate or a high thermal conductivity substrate under the GaN films.

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The need for a high current density, low energy spread and long lifetime electron source in electron beam lithography[1] and microscopy[2] is well established. High current density photocathodes also have very important applications in future light sources such as the Linac Coherent Light Source [3, 4] and Energy Recovery Linacs [5,6] In a previous report [7,8], we have shown that a rugged photocathode can be achieved consisting of a thin CsBr film on a thin Cr substrate which can operate at high current density (>150A/cm^2) when excited with photons of energy of 4.8eV (257nm) which is smaller than the CsBr bandgap of 7.3 eV. A photoyield up to 200nA/mW (~0.1% quantum yield) at 257nm was achieved. In addition, an emission band centered at 330nm was detected utilizing a photoluminescence technique. We argued that this electron emission is from photoelectrons generated from the intragap states in the thin film CsBr and the possible electron injection from the metal film [9].

To achieve a higher quantum yield, we replaced the Cr metal film with a GaN substrate to enhance the light absorption and electron injection. In this study, we fabricated a CsBr/GaN heterostructure photocathode. These CsBr/GaN cathodes have a higher quantum yield up to 800nA/mW (~0.4%). A higher electron current density than CsBr/Cr cathode can be potentially achieved for a given laser power. We can operate this cathode at >90A/cm^2 current density. The cathode lifetime (50% reduction in photoyield) depends mainly on the current density which is proportional to the laser power density on the sample for a given photoyield. Due to the high light absorption of the GaN film, high intensity laser illumination causes a temperature rise at the emission spot. Our simulation results shows a temperature rise close to 80 C in the GaN film (100nm thick) when illuminated at high power density with a 257 nm laser (resulting in an emitted current density >90A/cm^2). This temperature rise causes a faster degradation of the photoyield. Therefore the lifetime of these cathodes at high current density can be limited. In the future, we will optimize the band alignment between the CsBr and III-nitride substrate to achieve higher quantum yields. This will allow operation at less laser power for a given electron current density. In addition, long lifetimes at high

current density may be obtained with either sample cooling or using a more thermally conductive substrate.

As described in the previous report [7, 8], the apparatus consists of two connected vacuum chambers: one for deposition and one analysis, with base pressure in 10^{-10} torr range. The wurtzite GaN (0001) film is p-type (Mg doped 1 x 10^{18} cm^3), 0.1μm thick, and molecular beam epitaxy (MBE) grown on an AlN buffer layer on a c-axis sapphire substrate. The sample was grown by SVT associates. The conductivity of the p- type region is 6 ohms-cm measured with a 4 point probe. As shown in figure 1, the samples are lithographically patterned with a metal grid to improve the surface electrical conductivity. The samples receive a final cleaning in 98% sulfuric acid at 70ºC without affecting the metal pattern, followed by a deionized water rinse. The clean samples are loaded in the load-lock and baked overnight at about 150ºC.

The samples are transferred into the deposition chamber to deposit a 15nm thick CsBr film (figure 1) using an MBE high temperature effusion cell operating at 400°C. The thickness of the CsBr film is monitored in-situ with a crystal oscillator. The cathode is then transferred from the deposition chamber to the analysis chamber under vacuum where the photocurrent can be measured in both transmission and reflection modes. In the transmission mode, a quartz UV lens focuses the expanded laser beam onto the sample substrate to a minimum spot size of about 1.25um utilizing a shear plate. The electron collector (+1kV) is about 1mm away from the sample. Furthermore, the cathode holder is mounted on a piezoelectric driven flexure stage. This enables scanning the cathode surface across the laser beam, thus a photocurrent map (up to 80um x 80um) can be obtained.

In figure 2, we show a photocurrent curve of a CsBr/GaN photocathode in transmission mode under 9mW of 257nm laser illumination. By scanning the metal grid across the focused laser beam [7], we estimate the laser spot size is less than 2μm. Under such conditions, we can operate the CsBr photocathode with more than 3.6μA (>90A/cm^2) for at least 20 hours. A photoyield up to 800nA/mW (~0.4%) can be achieved.

In previous studies, we observed an emission band centered at 330nm detected utilizing photoluminescence. We argued that the photocurrent was generated from the intraband absorption states about 3.7eV below the CsBr conduction band. In addition to the sub-band gap photoemission from the CsBr thin film, there is also direct electron injection from the Cr film[9]. In the present study, to further enhance this electron injection, we selected a p-type GaN film to replace the Cr thin film. We present a photoemission model in figure 3. We believe that an enhanced direct electron injection from the GaN substrate to the CsBr conduction band contributes to the higher quantum yield observed in figure 2. Considering the CsBr has a band gap plus electron affinity of 8.2eV [10], these intraband absorption states are located at the mid-gap region. The GaN conduction band minimum is almost aligned with the CsBr conduction band minimum. Our measurements at 257nm show that a 100nm GaN film and a 5nm Cr film absorb >80% and <10% of the photons, respectively. Unlike a metal where electron-electron scattering dominates, the conduction electrons generated inside the MBE grown GaN can travel farther due to the weaker electron-phonon scattering. Therefore, more conduction band electrons in GaN can be directly injected into the CsBr conduction band and have enough energy to escape from the surface.

But unlike CsBr/Cr cathodes, which can be operated at a given quantum yield at a high current density for a long period of time, the quantum yield of CsBr/GaN cathodes decreases much faster while operating at high current density. This is the result of the temperature increase on the surface of the CsBr/GaN photocathode. The GaN absorbs >80% of the incoming photons. The steady state temperature in the emission area will be much higher than that of CsBr/Cr photocathodes.

To estimate the steady state temperature, we used a finite element analysis software package, ANSYS, to model the heat diffusion from the laser/emission spot through the GaN region and into the sapphire substrate. Mesh density studies were performed on each model such that the element size no longer affected the temperature outputs. The density, specific heat and thermal conductivity are 6.2g/cm^3, 0.49J/g·K and 1.3W/cm·K for GaN [11, 12] and 3.98g/cm^3, 0.78J /g·K and 0.35W/cm·K for

sapphire at 300K [13] respectively. And the perimeter of the sample is kept at 300K. In the simulations, we used temperature dependent specific heat and thermal conductivity values. For a 1.5 μm and a 2 μm diameter spot, the simulations show that the steady state temperatures reach 379K and 363K at 10mW laser power respectively. The temperature dependent thermal conductivities had a significant impact on the temperature results. The real thermal conductivities of the thin films can be smaller than the bulk values we used here. This can lead to a higher steady temperature. We believe this local temperature increase leads to the faster decay of the photoyield.

To fully understand this heating effect, it is worthwhile to revisit the activation process of CsBr based photocathode. As the film is illuminated with the focused UV laser, self-trapped excitons (STE) are created inside alkali halide film[14-16]. For alkali halides with inter-ionic distances greater than one third of the halogen atom diameter, such as NaCl and KCl, the STEs decay into F-H center pairs[14]. For CsBr, the interionic distance is less than one third of the Br atom diameter. The STE decays into a F-center (a negative ion vacancy with on excess electron bound at the vacancy) and a neutral Br atom occupying an interstitial site[14]. The CsBr surface gradually becomes a Cs rich due to the formation of both the F centers and interstitial Br atoms and their subsequent diffusion to the surface. In this process, the F-center reaching the surface will replace a Br ion and neutralize a surface alkali atom; and the Br interstitial reaching the surface thermally desorbs in the form of an atom or a diatomic molecule. Therefore, a Cs metallic surface layer is formed. The energy barrier at the CsBr surface is thus reduced, and eventually, a negative electron affinity surface can be formed at the emission spot. Thus electrons that have been excited or injected into the CsBr conduction band will have a better chance of escaping from the surface. This is why the quantum yield of the CsBr based photocathode increases with time initially until it reaches a steady state, i.e. the so called activation process. It is interesting to note that these color center formation and diffusion processes exist throughout the lifetime of cathode.

As discussed above, the color center diffusion creates the Cs rich surface. It is well known that only one monolayer of Cs can be formed at room temperature due to the weak bonds among Cs atoms. Therefore, the extra Cs atoms will leave the CsBr surface as more Cs is generated at the surface during the cathode operation. Under equilibrium condition, the Cs desorption rate should equal the Cs generation rate at the surface, which is proportional to the color center diffusion rate. This diffusion rate, proportional to $\exp(-E_{barrier}/KT)$, is larger at higher temperatures. For a cathode operated at 400K, the desorption rate is 3.8 times ($e^{4/3}$) of that of a cathode at 300K. As the Cs atoms leave the cathode surface, not only there is less Cs left in the film, but also more defects are generated blocking the electron transport through the CsBr. Therefore, the cathode life time is shorter when it is operated at a higher temperature. We have also observed that the photoyield of CsBr/Cr photocathode decreases when we heat the sample to a higher temperature with a substrate heater. Our simulations also show that this local heating problem can be solved by either cooling the sample or using a more thermally conductive substrate, such as diamond, when operating at a high current density [17].

In conclusion, a CsBr/GaN photocathode with a photoyield up to 0.4% under 257nm laser illumination has been realized. CsBr/GaN can be operated at >90A/cm2 current density. However, compared to a CsBr/Cr photocathode, the photoyield decays faster. This is due to the local heating at the laser spot which is confirmed by our simulation results. Based on these results, we believe that improvements in lifetime and a higher current density can be expected by either cooling the cathode or using a more thermal conductive substrate. We can also utilize a higher band gap material, such as AlGaN, to create better band matching at the interface as compared to the CsBr/GaN cathode. Thus a more efficient electron injection and a higher photoyield can be achieved.

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Captions:

Figure 1. As shown in the insert, a 1mm by 1mm size metal grid is lithographically patterned at the center of the GaN substrate to increase the electrical conductivity of the photocathode. After the sample cleaning, we deposit a 15nm CsBr film on top of the GaN film. We show a schematic drawing of the cross section view of the CsBr/GaN photocathode structure (not in scale). The cathode is mounted on a molybdenum sample holder that is kept at room temperature.

Figure 2. We show a photocurrent curve of a CsBr/GaN photocathode in transmission mode under 9mW 257nm laser illumination. The laser spot size is less than 3μm. The photoyield increases from zero to larger than 800nA/mW (0.4%) in less than 4 hours. This photoyield is about four times of the maximum photoyield of a CsBr/Cr cathode. The photoyield decreases with time, it drops below 400nA/mw after 26 hours. Under such a condition, we can operate the CsBr photocathode with more than >90A/cm^2 (3.6μA) for at least 20 hours.

Figure 3. We show the energy diagram of the CsBr/GaN heterostructure to explain photoemission photocathode. The electron affinity of the CsBr is reduced after the surface becomes Cs rich under 257nm illumination. A possible negative electron affinity can be achieved [7]. Many photoelectrons generated inside the GaN film are injected into the CsBr film. After activation, the energy barrier at the surface is reduced. More electrons can escape from the surface.

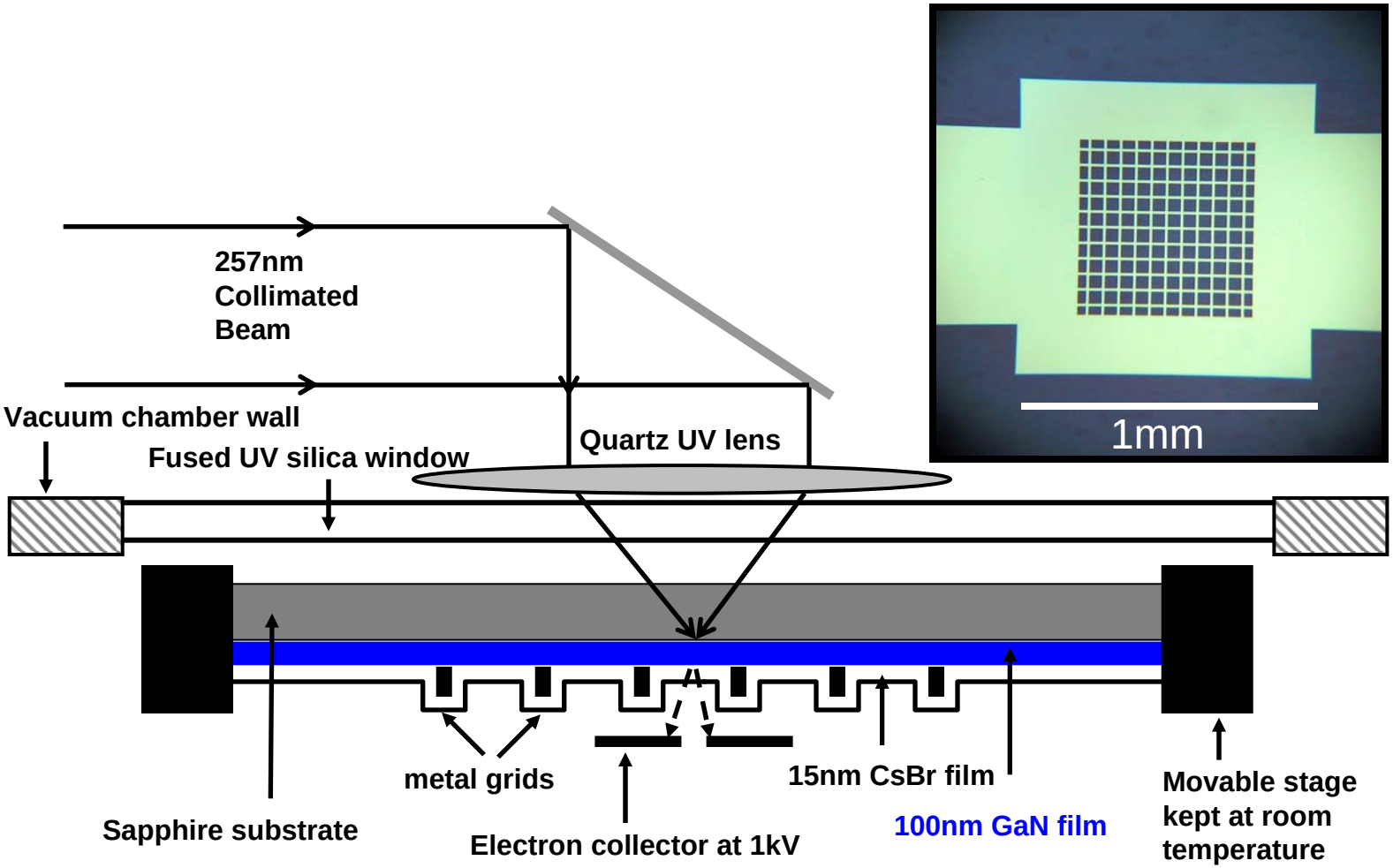
257nm
Collimated
Beam
Vacuum chamber wall
Fused UV silica window
Quartz UV lens
1mm
metal grids
Sapphire substrate
Electron collector at 1kV
15nm CsBr film
100nm GaN film
Movable stage
kept at room
temperature

Figure 1

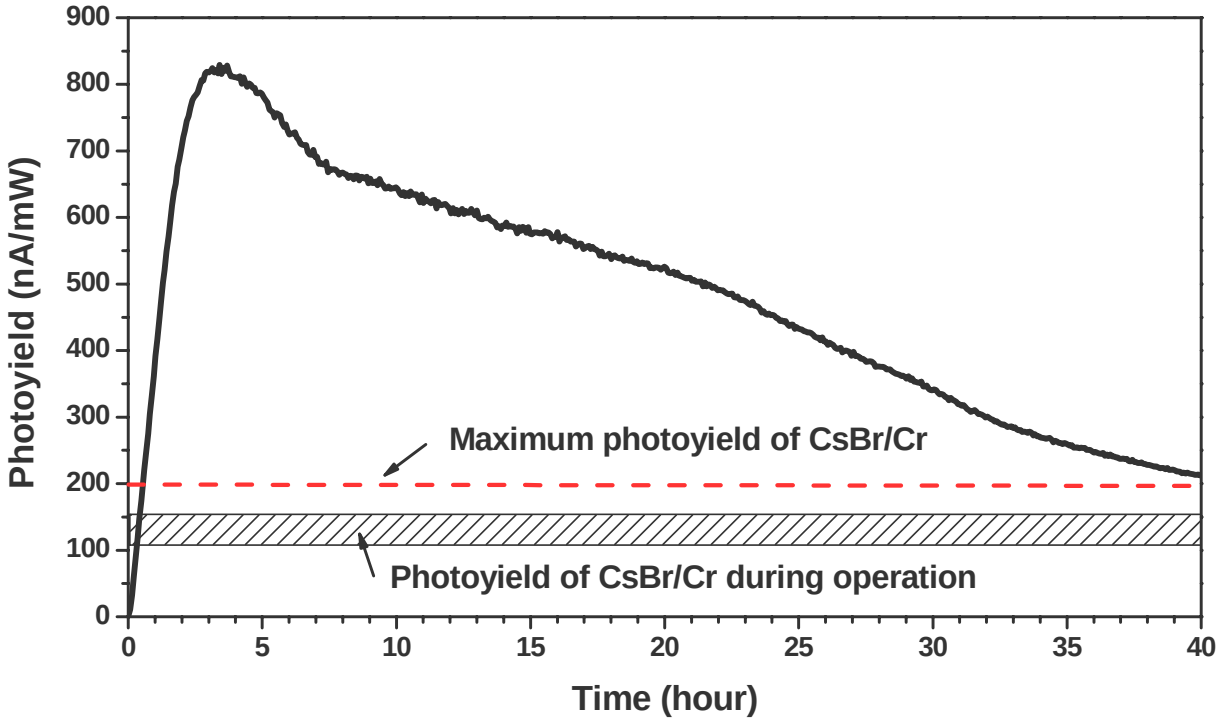
Photoyield (nA/mW)
900
800
700
600
500
400
300
200
100
0
0
5
10
15
20
25
30
35
40
Time (hour)
Maximum photoyield of CsBr/Cr
Photoyield of CsBr/Cr during operation

Fig. 2

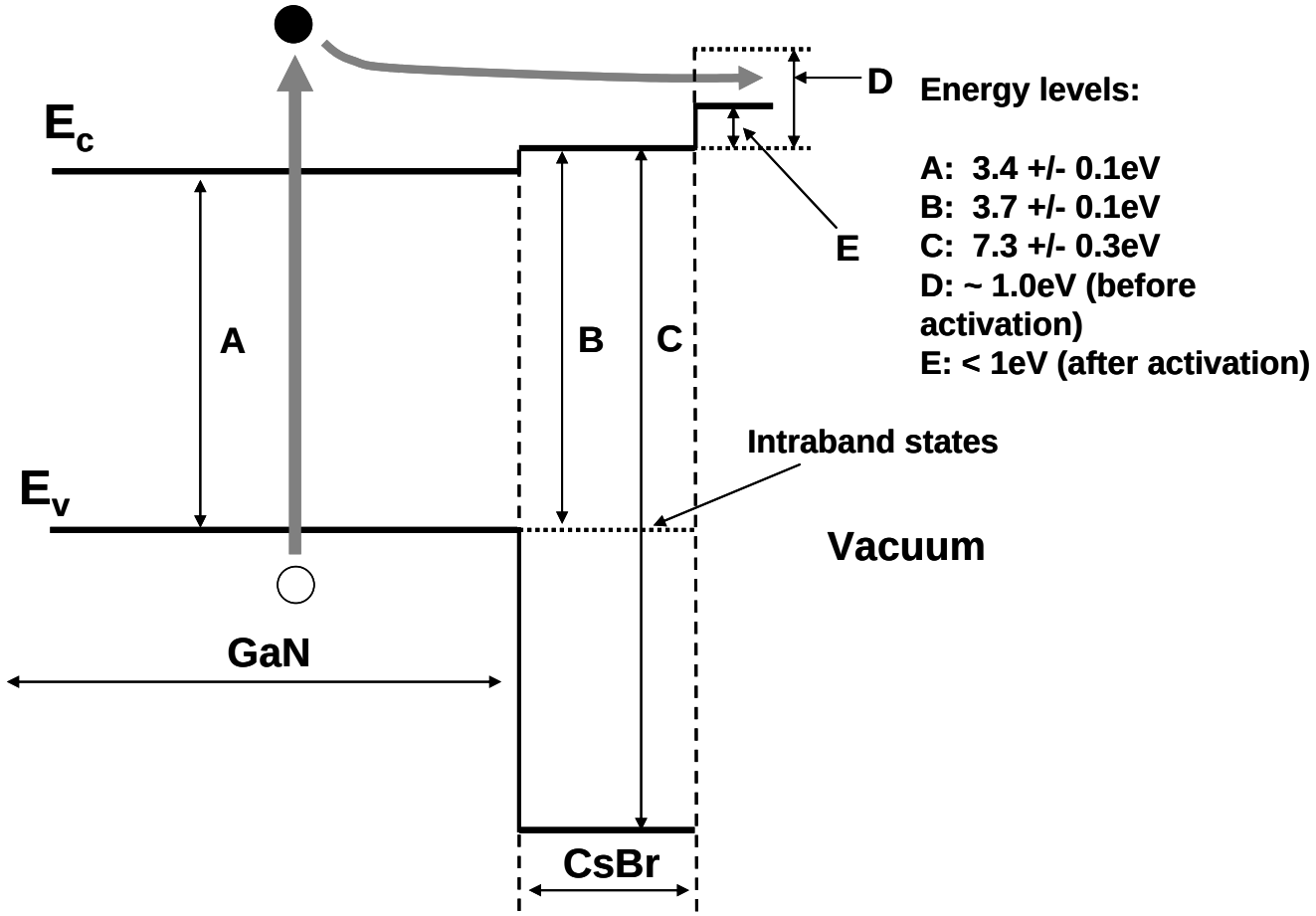
D
Energy levels:
A: 3.4 +/- 0.1eV
B: 3.7 +/- 0.1eV
C: 7.3 +/- 0.3eV
D: ~ 1.0eV (before activation)
E: < 1eV (after activation)
E
E_c
A
B
C
Intraband states
E_v
Vacuum
GaN
CsBr

Fig. 3