Substrate dependence of CsK$_2$Sb photo-cathode performance

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A photo-cathode able to generate a high-performance electron beam with large operability is one of the most important devices in an advanced accelerator. In particular, the CsK$_2$Sb photo-cathode is of interest because it has high robustness and can be driven by visible light. In this study, we performed cathode evaporation on Si(100), Si(111), and GaAs(100) substrates to evaluate the performance dependence on the substrate material and surface state. For each substrate, the cathode performance on the as-received and cleaned substrates were compared. We found that the cathode performance on the cleaned substrate was superior to that on the as-received substrate for all materials. The cathode performance on the cleaned GaAs(100) and Si(100) substrates were similar, but that on the cleaned Si(111) was significantly much lower. This result gives experimental evidence about the substrate surface direction dependence of CsK$_2$Sb photo-cathode performance.

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1. Introduction

In linear-accelerator-based advanced accelerators such as the Energy Recovery Linac (ERL) [1,2], Free Electron Laser (FEL) [3], and Linear Collider (LC) [4], the performance of the accelerated beam strongly depends on the initial beam property. Therefore, the beam performance from the source is important. For example, a high-brightness electron beam with more than 10 mA average current and less than 1.0 $\pi$ mm-mrad [5] emittance is required for the next generation of ERL-based synchrotron radiation (SR) facilities [1]. Since 1980, a photo-cathode has been operated as an advanced electron source in many accelerators [6].

The most popular photo-cathode material is a metal such as Cu [7], Mg [8], or Ag [9]. Due to the large work function of the metal, ultraviolet (UV) light is required for photo-electron emission. There are several techniques used to generate short-pulse UV light; harmonic generation with a short-pulse infrared (IR) or visible light (VL) laser is one of the most popular methods [10]. The desired UV light is obtained from the third or fourth harmonics of the fundamental laser light. The conversion efficiency is usually much less than unity, and the laser system becomes complex, resulting in lower laser stability. Because the quantum efficiency (QE) of photo-electron emission is typically of the order of 0.01%, it requires a high-power laser to generate a high-brightness electron beam.
A Cs$_2$Te cathode [11] requires UV light for the electron emission, but the QE is more than 10%. Due to the high QE, a Cs$_2$Te cathode is one of the candidates for generating a high-brightness electron beam [12].

Multi-alkalis are a group of materials composed from two or more alkali metals. Multi-alkali materials have been used as photo-cathodes in photomultiplier tubes [13]. The multi-alkali cathode is considered to be the strongest candidate as a high-brightness electron source because it can be operated by green light (532 nm) with 10% QE [14,15], which is easily obtained from the second harmonics of a solid-state laser. Moreover, the multi-alkali cathode has a long operational lifetime [16].

A CsK$_2$Sb cathode is fabricated as a thin film on a substrate by evaporation in an ultra-high-vacuum environment. Various materials have been examined as the substrate, e.g. glass(amorphous) [17], Cu(amorphous) [18,19], SUS [19–21], Mo(amorphous) [19,21], Mo(100) [14], Si(100) [15], and GaAs(100) [22], where the numbers in parentheses are the surface direction of the crystalline substrate.

A Cs$_3$Sb cathode has proven to depend strongly on the chemistry of the substrate surface [23]. X-ray photo-electron spectroscopy studies for the CsK$_2$Sb cathode [21,24] suggest that the cathode performance strongly depends on the substrate surface state (oxidation, etc.).

Cathodes fabricated on amorphous substrates (glass, Cu, SUS, and Mo) showed relatively low QE: 1%–5% with 532 nm laser light [17–21]. In contrast, cathodes fabricated on crystalline substrates [Si(100), Mo(100), and GaAs(100)] showed relatively high QE: 7%–10% with 532 nm laser light [14,15,22]. These results suggest that the substrate crystallinity has an impact on the cathode performance.

CsK$_2$Sb crystal direction was studied with X-ray diffraction [25,26]. The quantum efficiency of the cathode in these studies was relatively low (3%), and cathode performance dependence on the surface direction of the substrate or CsK$_2$Sb crystal was not significantly observed. This suggests that the cathode evaporation condition was not fully optimized in these studies.

By comparing cathodes evaporated with an optimized condition, on substrates in different surface directions, we expect to reveal the dominant factor for cathode performance, whether cleanness, crystallinity, or surface direction of the substrate.

For that purpose, a CsK$_2$Sb cathode was fabricated on Si(100), Si(111), and GaAs(100), and their performance was compared. To observe the dependence significantly, the cathode evaporation condition was first optimized. For each substrate, the as-received and cleaned surfaces were examined. In the next sections, we describe the experiment, and provide the results and discussion.

2. Experiment

Figure 1 shows a 3D CAD drawing of the CsK$_2$Sb evaporation chamber. The vacuum was maintained with an ion pump (ULVAC PST-100 with 100 l s$^{-1}$ pumping speed) [27] and non-evaporative getter (NEG; SAES CapacitTorr-D400, 480 l s$^{-1}$) [28], and was kept at ultra-high vacuum (UHV) at a typical pressure of $1.0 \times 10^{-8}$ Pa. The cathode substrate was fixed on a molybdenum puck and the chamber accommodated up to three pucks for cathode evaporation. The puck could be transferred to and from the chamber through the gate valve to the VSC (vacuum suitcase), which was designed to transport the cathode without breaking the vacuum, because the CsK$_2$Sb cathode is easily damaged by air exposure; the VSC maintains good vacuum quality (a typical pressure is $1.0 \times 10^{-8}$ Pa).

The puck was mounted on the cathode holder during evaporation and electron emission. Three cathode pucks were placed vertically in line on the holder. The holder could be moved vertically...
Fig. 1. Three-dimensional drawing of the CsK$_2$Sb evaporation chamber. Up to three cathode pucks could be stored in the chamber. The cathode could be transferred through the gate valve to another vacuum chamber (vacuum suitcase) for cathode transportation.

to adjust the position of each puck for evaporation, photo-electron emission, and transfer. For each cathode puck, an 8 mm diameter substrate was placed on a molybdenum baseplate. The substrate was soldered on the base with indium and fixed by a tantalum cup. In this study, Si(100) and Si(111) p-type wafers with a resistivity of \( \leq 0.002 \, \Omega \, \text{cm}^{-1} \) and GaAs(100) p-type wafers were employed as the substrates.

The Si wafer was etched, single-side polished, and finally washed with the RCA method by the sales company. The GaAs wafer was also etched, double-side polished, and finally washed with the RCA method. The Si(100) and Si(111) substrates were processed with a 5% HF solution for about five minutes to remove the surface oxidized layer [29]. The GaAs surface was processed with an \( \text{H}_2\text{SO}_4 : \text{H}_2\text{O}_2 : \text{H}_2\text{O} (4 : 1 : 1) \) solution for about five minutes to remove the surface oxidized layer [30].

According to Ref. [29], the surfaces of Si(100) and Si(111) are equivalent except for the surface direction after the HF treatment. This cleaned surface is kept for 3 and 50 hours in air for Si(100) and Si(111), respectively [31]. In our experiment, after the cleaning, the Si sample was kept in a desiccator (typical pressure was 1 Pa); the air exposure duration during the sample transfer was in total less than 30 minutes, and the re-oxidation on the substrates was negligible. Therefore, the Si(100) and Si(111) substrates in our experiment were equivalent except for the surface atomic arrangement.

To examine the effect of the surface oxidation, we also examined the as-received substrates [32,33]. The atomic arrangement of the as-received surface is disturbed by the oxidation layer [34], and it is considered to be amorphous [33,34].

The evaporation sources were mounted on a linear movement mechanism in the chamber. The high-purity (99.9999%) Sb pellets were resistively heated in a tungsten evaporation basket. The K and Cs sources were dispensers provided by SAES Getters Co., Ltd. [35]. The amount of material
on the substrate was monitored with a quartz thickness monitor (INFICON Q-pod Quartz Crystal Monitor). A cross-section of the arrangement around the cathode puck is shown in Fig. 2. The thickness monitor was placed in a symmetrical position relative to the cathode substrate in order to absorb an equivalent amount of vapor. The source-to-substrate distance was maintained at about 12 mm during the evaporation.

To control the cathode temperature, a tungsten heater was used. The heater was mounted on the head of the linear mover, which could be inserted behind the cathode puck, as shown in Fig. 2. The cathode puck temperature was measured with a thermocouple.

Typically, the CsK$_2$Sb cathode was formed by sequential evaporation of Sb, K, and Cs on a substrate. The QE of CsK$_2$Sb depends on the evaporation condition even if the substrate is the same [25]. In order to eliminate the effect, we fixed the evaporation condition (Sb thickness and substrate temperature during the evaporation), giving a maximum QE and less QE fluctuation due to condition variation. The amount of K and Cs were automatically determined, giving maximum QE after each evaporation. The maximum QE of CsK$_2$Sb as a function of substrate temperature is shown in Fig. 3 for the Si(100) and Si(111) substrates, respectively. We repeated the evaporation several times for each substrate. The error on the horizontal axis is temperature fluctuation during Cs evaporation, and the error on the vertical axis is statistical error. We found that the QE was maximized around 100 °C on Si(100) and Si(111) with good reproducibility. Figure 4 shows that the QE as a function of Sb thickness showed a weak dependence. We repeated the evaporation several times for each substrate. The errors on the horizontal and vertical axes are statistical errors.

According to these results, the evaporation was performed at 100 °C with 10 nm Sb thickness. The amount of K and Cs were automatically determined to maximize QE after each evaporation, i.e. we stopped the evaporation whenever the QE was saturated. The evaporation procedure is summarized as follows:

1. The substrate was heated to 450–600 °C and held at that temperature for several hours to clean the surface (heat-cleaning process).
Fig. 3. The maximum QE is shown as a function of temperature during the evaporation, with blue squares and red circles for the Si(100) and Si(111) substrates, respectively. The error on the horizontal axis is fluctuation during evaporation and the error on the vertical axis is statistical error from three measurements.

Fig. 4. The maximum QE is shown as a function of Sb thickness, with blue squares and red circles for the Si(100) and Si(111) substrates, respectively. The errors on the horizontal and vertical axes are only statistical.

(2) The temperature was lowered to 100 °C. This temperature was maintained during the evaporation.
(3) A 10 nm-thick layer of Sb was evaporated on the substrate surface.
(4) Then, K was evaporated. Simultaneously, the QE was observed by illuminating the cathode with 532 nm laser light. The evaporation was stopped when the QE was saturated, which usually occurred between 0.3% and 0.8%. The typical thickness of K was 60 nm.
Typical examples of QE evolution in time during evaporation are shown with red circles, blue triangles, and green squares for the cleaned Si(100), Si(111), and GaAs(100) substrates, respectively. QE appears in K evaporation and increases rapidly in Cs evaporation. K evaporation started on 48, 45, and 45 min for the cleaned Si(100), Si(111), and GaAs(100) substrates, respectively. Cs evaporation started on 183, 175, and 135 min for the cleaned Si(100), Si(111), and GaAs(100) substrates, respectively.

(5) The Cs layer was evaporated. The Cs evaporation was controlled in the same manner as the K evaporation. The QE was typically between 1% and 10%, and the thickness was 120 nm.

(6) The substrate was cooled to room temperature.

The QE evolution during the cathode evaporation is shown in Fig. 5 for the cleaned substrates. QE was measured with a 532 nm laser. The cathode was biased at $-100 \text{ V}$, and the photo-current was measured as the current of the bias supplier. The laser power was adjusted to 0.8 mW during the K evaporation and 0.36 mW during the Cs evaporation to avoid the space charge limitation. The laser spot size was 0.5 mm$^2$, and the typical photo-current was 1–3 $\mu$A for K evaporation and 3–9 $\mu$A for Cs evaporation. We examined Si(100), Si(111), and GaAs(100) substrates. The results (maximum QE) are summarized in Table 1, including the results of earlier studies. We repeated the evaporation five times for each substrate. After each evaporation, the substrate was heated to initialize the surface. The error was obtained as the standard deviation of the five measurements, and it is statistical only.

The QE of the as-received substrate was around 2.5%–5.5% at 532 nm. The cleaned Si(100) and GaAs(100) substrates showed good QE, as high as 10%. These results are similar to those of the cleaned Mo(100) [14] and Si(100) substrates obtained by the Cornell group [15]. In contrast, the QE of the cleaned Si(111) was higher than that of the as-received Si(111), but it is much lower than that of the cleaned Si(100) and GaAs(100) substrates.

According to results shown in Table 1, the cathode performance developed on the cleaned substrate was significantly higher than that on the as-received substrate for all cases. We could not conclude that the substrate crystallinity has an impact on the cathode performance, because the chemical property of the oxidized surface may differ from the clean one, but it can be collateral evidence. The cathode formed on the Si(111) substrate showed the least performance in the tested substrates in both the cleaned and as-received cases. The difference in the cathode performance on the Si(100) and Si(111) substrates provides direct evidence that the cathode performance depends strongly on the substrate
Table 1. Summary of the maximum QE of the CsK₂Sb photo-cathode on Mo(100), Mo(amorphous), Si(100), Si(111), and GaAs(100) substrates at 532 nm.

| Substrate     | Surface treatment | QE (%) at 532 nm |
|---------------|-------------------|------------------|
| Mo(100)       | Polished+sputter  | 10.0 [14]        |
| Mo(amorphous) | Polished+sputter  | 2–5 [19,21]      |
| Si(100)       | As-received       | 4.8 ± 0.6        |
| Si(100)       | 5% HF             | 9.4 ± 0.7        |
| Si(100)       | 5% HF             | 7–10 [15]        |
| Si(111)       | As-received       | 1.6 ± 0.1        |
| Si(111)       | 5% HF             | 2.3 ± 0.3        |
| GaAs(100)     | As-received       | 5.5 ± 0.2        |
| GaAs(100)     | H₂SO₄ : H₂O₂ : H₂O | 10.0 ± 0.2      |

direction, because the material properties of Si(100) and Si(111) are exactly the same otherwise. A similar conclusion was also obtained by comparing the QE of the crystalline Mo(100) [14] and amorphous Mo [19,21], because the physical properties of crystalline and amorphous Mo other than the surface atomic arrangements are almost same. The Mo(100) and Mo(amorphous) substrates in Refs. [14,19,21] are considered to be cleaned because it was polished and sputtered.

The results can be summarized as follows:

- By comparing the as-received and cleaned substrates, the cathode on the cleaned substrates showed better performance.
- By comparing amorphous and crystalline substrates, the cathode on the crystalline substrates showed better performance.
- By comparing Si(100) and Si(111), the cathode on Si(100) showed better performance.

By considering these facts, the cathode performance depends not only on cleanness and crystallinity, but also on the surface direction of the crystalline substrate. This is the first experimental evidence that CsK₂Sb cathode performance depends on the crystal surface direction of the substrate.

3. Discussion

Crystalline CsK₂Sb forms a DO₃ cubic structure [36,37]. The unit cell contains four formula units and is represented by four face-centered sub-lattices shifted by \( a\sqrt{3}/4 \) (where \( a \) is the lattice constant of the primitive translation vector) along the body diagonal [36,38]. The lattice constant \( a \) is 8.61 Å [36, 37,39].

Figure 6 shows the surface atomic arrangements of CsK₂Sb(100), CsK₂Sb(111), Mo(100), GaAs(100), Si(100), and Si(111) surfaces. By considering the matching between the atomic arrangements among CsK₂Sb(100), CsK₂Sb(111), Si(100), Mo(100), and GaAs(100) surfaces, CsK₂Sb is grown in the (100) direction on Si(100), Mo(100), and GaAs(100) surfaces, and in the (111) direction on Si(111).

The reason for the reduced performance of the as-received substrates compared to the cleaned substrates could be the lesser quality of the CsK₂Sb crystal. Oxidation distorts the atomic arrangement of the substrate surface resulting in poor matching between the substrate and the CsK₂Sb crystals. The poor matching leads to a lower quality of CsK₂Sb crystal grown on the substrate and reduced performance.
Fig. 6. Atomic arrangement of CsK2Sb(100) and CsK2Sb(111) on the Mo(100), GaAs(100), Si(100), and Si(111) surfaces. The lattice constants of Mo, Si, and GaAs are 3.15 Å, 5.43 Å, and 5.65 Å, respectively [40].

The cathode on the cleaned Si(100) and GaAs(100) substrates showed a good QE, as high as 10%. These results are similar to those on the cleaned Mo(100) and Si(100) substrates obtained by the Cornell group [14]. On the other hand, the cathode performance on Si(111) was less than the others. These results can be explained by the band structure of CsK2Sb. In Refs. [36,37], the bulk band dispersion of CsK2Sb was calculated. The point on the boundary surface of the Brillouin region is called the K, L, and X point in the (110), (111), and (100) directions, respectively. According to the bulk band dispersion, CsK2Sb is a direct transition type at the $\Gamma$ point, with a 1.1 eV bandgap. The bandgap is about 2.1, 3.1, and 1.4 eV at K, L, and X, respectively. If we consider the photo-electron emission in the (100) direction, not only electrons at the $\Gamma$ point, but also electrons at the X point contribute to the emission, because the bandgap at the X point is similar to that at $\Gamma$. On the other hand, for photo-electron emission in the (111) direction, there is no contribution at the L point, because the bandgap energy (3.1 eV) is larger than the laser photon energy (2.3 eV at 532 nm). The photo-electron emission of CsK2Sb in the (111) direction is possible only at the $\Gamma$ point, and this is the reason why the quantum efficiency of the (111) surface is less than that of the (100) surface.

By a similar consideration, the QE of the (110) surface should be less than that of the (100) surface. In the preceding studies [25,26], CsK2Sb was grown in the (200) or (220) directions on the Si(100) substrate, depending on the case, and the cathode performance of (200) and (220) were similar [25]. We consider that the cathode evaporation conditions in these experiments were not fully optimized, because the QE is only 3% for 532 nm light in both cases. That is why the CsK2Sb crystal direction depends on the case, and there was no significant difference in the cathode performance when grown in the (100) and (110) directions in these studies.

In our case, the cathode evaporation condition was carefully optimized and the cathode performance reproducibility was quite good. The cathode performance dependence on the substrate crystallinity and the surface direction was confirmed based on the reliable experiments.

Applications based on linear accelerators (e.g. FEL, ERL, etc.) sometimes require an extremely high-brightness electron beam, and the photo-cathode has to directly provide such a beam. A
high-performance cathode is one of the most important devices in the advanced accelerator in this context. We found that the quantum efficiency depends not only on the substrate material and cleanness (chemical condition), but also on the crystallinity and the surface direction. This fact has an impact on experimental physics with accelerators, because there is some potential to improve thin-film cathode performance by revisiting the substrate crystallinity and surface direction of the substrate. High quantum efficiency has large merit in generating a high-brightness electron beam by relaxing the requirements for the drive laser, which makes the system more reliable and stable. The availability of the system is improved and the effective cost of the project (cost per operation time) becomes less. Our result has the potential of a large impact on accelerator science from this point of view.

4. Summary

We studied the substrate dependence of CsK$_2$Sb photo-cathode performance. We studied GaAs(100), Si(100), and Si(111) as the substrates, and found that the cleaned substrates resulted in higher performance than the as-received substrates for all materials. By comparing cathodes on GaAs(100), Si(100), Si(111), and Mo [14,19,21], we found that the cathodes on GaAs(100), Si(100), and Mo(100) had significantly better performance than on Si(111) and Mo(amorphous). This showed that the cathode performance depends strongly not only on the substrate material and surface state, but also on the crystallinity and the surface direction.

We have obtained the first experimental evidence of the substrate surface direction dependence of CsK$_2$Sb photo-cathode performance. This fact has an impact on accelerator science, because there is some potential to improve thin-film cathode performance by revisiting the substrate crystallinity and surface direction.

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