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ANALYSIS OF NEA-GaAs SURFACE CONDITION BY TPD

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Temperature programmed desorption (TPD) with a quadrupole mass spectrometer (QMS) were carried out on negative electron affinity (NEA) surfaces formed by alternating supply of Cs and O₂ onto GaAs substrate. A number of Cs¹³³ desorption peaks were observed and the lowest desorption peak appeared at the same temperature range of degradation of quantum efficiency (QE). It was found that intensity of Cs⁺, As⁺, and Ga-oxide peaks varied depending on conditions of thermal treatment and NEA activation sequence.

I. Introduction

Cs and O₂ adsorption on heavily p-doped GaAs surface has an attracted considerable interest, since it produces a negative electron affinity (NEA) surface. In the case of NEA, the vacuum level at the surface lies below the conduction band minimum in the bulk. It is of interest from viewpoints both of fundamental science and practical applications of electron sources. The first NEA nature was demonstrated on vacuum cleaved p-doped GaAs(110) surfaces with Cs adsorption by Scheer and Van Laar¹⁾. Turnbull and Evans reported that the photo-sensitivity were increased by the yo-yo method, where Cs and O₂ are repeatedly supplied on p-doped GaAs(110) surfaces²⁾.

NEA-GaAs for photocathodes are widely used for high quantum efficiency (QE), high spin polarization, low emittance, and short bunched beams. For these remarkable advantages, NEA-GaAs photocathodes have been studied mainly in the field of accelerator physics since 1970's³⁾.

Although NEA-GaAs surface has been investigated for a long time and applied for various fields, its structure is not fully understood, and several models have been proposed, namely, the heterojunction⁴⁻⁸⁾, the dipole⁹⁾, and the cluster models¹⁰⁾. While the mechanisms of NEA-GaAs have been extensively studied, most studies have focused on the ideal surfaces. In the case of photocathodes, GaAs surfaces are repetitively refreshed by thermal treatment, which induced the evaporation of atoms and generation of surface vacancies. In the previous works, we demonstrated that the repeated sequence of the thermal treatments and the NEA activations induced higher QE¹¹⁾. It is probably due to the formation of efficient electron emission sites by the reaction of surface Cs and oxygen with residual species after the repeated sequence.¹¹⁾

In this paper, we discuss reaction processes at NEA-GaAs surfaces by using temperature programmed desorption (TPD) technique which is known to be an effective technique to determine the surface adsorbates for the purpose of deep understanding the relationship between electron emission processes and practical desorption species.

II. Experimental

All experiments were performed in an ultrahigh vacuum system consisting of an install chamber and an activation chamber. Prior to the experiment chambers were evacuated lower than 2×10^{-8} Pa, and the activation chamber is equipped with a

quadrupole mass spectrometer (QMS). The chambers are connected via gate valves, and samples are transferred to the activation chamber by manipulators. Sample was Zn-doped p-type GaAs(100) with $\sim 10^{19} \text{ cm}^{-3}$ of carrier concentration, and was mounted on a molybdenum sample holder by indium soldering. First of all, thermal cleaning of sample was done in the activation chamber prior to the NEA activation sequence. Subsequently, we repeatedly carried out the sequence of thermal treatments and NEA activations for the same GaAs(100) surface in the activation chamber. Details of the sequence have been reported previously¹¹⁾ and explain here briefly. Two types of thermal treatment sequence were carried out for the same sample. One is a high temperature (HT) sequence, in which the sample was heated up to 700 °C during the thermal treatment. The other is a low temperature (LT) sequence in which the sample temperature was kept at 580 °C. In the both treatments, the programming rate was constant at 10 °C/ min, and the treatment time was fixed for 1 hour. After the thermal treatment, the sample was cooled down to the room temperature, and then the NEA activation process was performed. NEA surfaces were formed by means of coadsorption of small amount of Cs and oxygen on the surface, so called yo-yo method. Cs was evaporated from a commercial SAES-getters dispenser and oxygen pressure was controlled with a variable leak valve. During the NEA activation, QE of the sample was measured under illumination of semiconductor laser light ($\lambda = 650 \text{ nm}$, $466 \text{ } \mu\text{W}$). The extracted voltage was -100 V in the QE measurements. During the thermal treatments, TPD measurements were performed at the same time. The sample was faced to the QMS head during the TPD measurements, and the signal intensities at mass numbers m/z of 69(Ga^+), 75(As^+), 133(Cs^+), and 154(Ga_2O^+) were mainly monitored.

III. Results and Discussion

As mentioned above, some results in different thermal condition were reported¹¹⁾, and in the present research it was confirmed that the HT sequence provided stable QE values of typically 8.4 % after the yo-yo method. When we carried out the LT sequence successively after the HT sequence, the QE value remarkably increased by the factor of 1.25. However, repetition of the LT sequence induced abrupt degradation of QE, and QE value decreased almost zero after several times of LT sequence. These results are consistent with our previous report, and TPD measurements were done for each sequence.

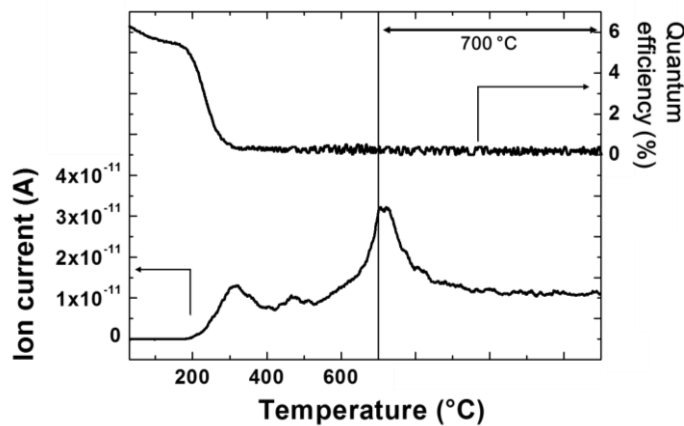


Fig. 1 TPD spectrum of $^{133}\text{Cs}^+$ (lower curve) and the corresponding QE (upper curve) during TPD after HT sequence. The sample was heated at a rate of 10 °C/ min up to 700 °C, and the temperature was kept for 1 hour. Several Cs related peaks were observed and Cs desorption was not completed after the temperature reaches 700 °C. The appearance of 1st peak of Cs and QE degradation were observed at almost the same temperature.

Figure 1 shows the relation between TPD spectrum of $133(\text{Cs}^+)$ and QE evolution of GaAs sample after the HT sequence. Note that sample temperature was constantly increased up to 700°C and was kept at 700°C for 1 hour. Several peaks of $133(\text{Cs}^+)$ were observed and Cs desorption was observed after the temperature reaches to 700°C . This result indicates the 700°C is insufficient to completely remove Cs remaining at surface after NEA activation. It is clear that the QE degradation was started almost at 200°C with the 1st peak of Cs. We consider that a part of Cs relating to the 1st peak plays an important role for electron emission in NEA condition. After appearance of the 1st peak, several peaks of Cs were observed without change in QE during TPD.

Figure 2 shows comparison of TPD spectra of $75(\text{As}^+)$, $133(\text{Cs}^+)$ and $154(\text{Ga}_2\text{O}^+)$ up to 700°C after HT sequence. The TPD spectrum of $133(\text{Cs}^+)$ is same as shown in Fig. 1. Similar tendency was observed both in Cs^+ and As^+ curves at around 450°C . However Ga_2O^+ was not identical to other two elements and no peak was observed at 450°C .

Figure 3 shows TPD spectra after the LT sequence that was successively done after the HT sequence. At this condition, QE obtained was increased to 10.3%, and showed a good agreement with our previous study¹¹⁾. In contrast to Fig. 2, there is no clear desorption peak of As^+ and Ga_2O^+ .

It was found that the TPD spectrum with the similar tendency as Fig. 3 was obtained

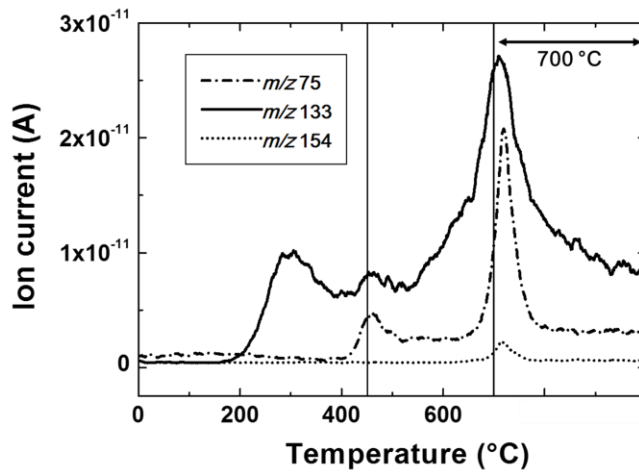


Fig. 2 TPD spectra of $75(\text{As}^+)$, $133(\text{Cs}^+)$ and $154(\text{Ga}_2\text{O}^+)$ up to 700°C after HT sequence. When this condition, QE was 8.4 %. Broken, solid and dotted lines indicate m/z of $75(\text{As}^+)$, $133(\text{Cs}^+)$ and $154(\text{Ga}_2\text{O}^+)$, respectively. Similar tendency was observed both in Cs^+ and As^+ curves at around 450°C , while Ga_2O^+ was not identical to other two elements and no peak was observed at 450°C .

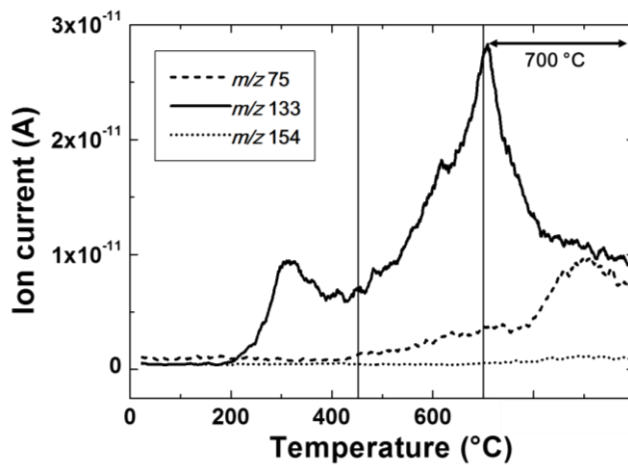


Fig. 3 TPD spectrum of $75(\text{As}^+)$, $133(\text{Cs}^+)$ and $154(\text{Ga}_2\text{O}^+)$ up to 700°C after HT and LT sequence. When this condition, QE was 10.3 %. Broken line, solid line and dotted line indicate m/z of $75(\text{As}^+)$, $133(\text{Cs}^+)$ and $154(\text{Ga}_2\text{O}^+)$ respectively. In contrast to Fig. 2, some of peaks were not observed. There is no clear desorption peak of As^+ and Ga_2O^+ .

by repeating the LT sequence. On the other hand, when we carried out the HT sequence after repetition of LT sequence, QE value was recovered to typical value, and TPD spectrum after HT sequence were same as shown in Fig. 2.

These results suggest that all the adsorbed Cs in various forms do not contribute to electron emission process. TPD spectra were different depending on sequence, especially at 450 °C and at around 600-700 °C. After the LT sequence, no As^+ or Ga_2O^+ peak was observed up to 700 °C. Though detailed surface processes are under consideration, desorption and oxidation of surface atoms (Ga or As) of substrates may take place in the formation of NEA surfaces.

IV. Conclusions

We discussed TPD of NEA-GaAs with several thermal treatment conditions. We confirmed that Cs forms various structures on surface, and that only some of these structures contribute electron emission. Details concerning the dependence of TPD spectra on NEA-GaAs will be reported elsewhere.

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