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Study on work function and corresponding electron emission during NEA activation of GaAs surface

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Abstract

We have investigated the relation between the work function and corresponding quantum efficiency during the negative electron affinity (NEA) activation processes of GaAs surfaces qualitatively and determined the regions of positive electron affinity (PEA) surface and NEA surface. We found that the two surface states of NEA and PEA were switched during the alternating supply of Cs and O₂ in the NEA activation sequence. After the supply of O₂ following Cs deposition onto the clean GaAs, the surface still showed the PEA state for a few minutes and changed to the NEA state with increase of electron emissions. It should be noted that once the NEA surface was formed, there were two regions where the quantum efficiency continued increasing, both with and without changing the tendency of spectrum. From the above results, it is considered that the increase in quantum efficiency is closely related to the number of electron emission sites and that there are various electron emission structures.

I. Introduction

A negative electron affinity (NEA) surface in which the vacuum level lies below the conduction band minimum is one candidate for next-generation electron sources because of its large photoelectron current, ultra-short-pulsed operation, high spin polarization, low emittance nature, etc.. Usually, an NEA surface is formed by (Cs,O) layer adsorption on clean p-type III-V semiconductor surfaces. The formation process is called “NEA activation” or “yo-yo” methods, where Cs deposition and subsequent O₂ exposure are repeated several times. Photoemission during the NEA activation shows cyclic ups and downs that correspond to the supply of Cs and O₂, and the maximum value of photocurrent is gradually increased and is finally saturated. The obtained photocurrent curve is “yo-yo curve”. The NEA surface has been studied for more than half a century, and various models have been proposed for the NEA surface structure. However, the detailed processes and mechanisms of actual NEA surfaces are not yet understood. It is essential to clarify the detailed surface process for

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comprehending the NEA surface structures and realizing practical applications toward new functional electron sources. In the present study, we discuss changes of the work function vs. time evolution of quantum efficiency during the NEA activation and determined the regions of the positive electron affinity (PEA) and NEA surfaces. It was found that the quantum efficiency increased with changes of the tendency of the quantum efficiency spectrum and almost without changes of tendency during the NEA surface formation.

II. Experimental procedures

The samples used were epi-ready Zn-doped p-type GaAs (100) ($\sim 1.0 \times 10^{19}/\text{cm}^3$) substrates. The thermal pretreatment of samples and the successive NEA activation with alternating supply of Cs and O₂ were carried out in a UHV chamber. Cs atoms were supplied by sublimation from a Cs dispenser (SAES Getters) and the amount of O₂ (>99.999995 vol.%) was controlled with a variable leak valve. The base pressure of the UHV chamber during NEA activation remained at $2.0\text{--}4.0 \times 10^{-8}$ Pa using a non-evaporable getter (NEG) pump and an ion pump. To measure the emitted photoelectron current, sample were biased by -100 V. The quantum efficiency (QE) was calculated as a simple ratio of the number of emitted photoelectrons to the number of incident photons. The optical system consisted of a Hg–Xe lamp, a spectroscope, an optical aperture, lenses, a viewport, and mirrors to shape and transport the excitation light. A bandpass filter with a 503 nm transmission limit was inserted in the optical path to prevent second-order diffracted light. The excitation light ($h\nu = 1.50\text{eV}$, density $296 \mu\text{W}/\text{cm}^2$) was used to measure the time course of QE in the NEA activation. When measuring the QE spectra, the wavelength was swept from 650 to 1000 nm in 10nm steps. Time for a single scan of the entire wavelength region in spectrum measurement was almost 30 sec. Note that we could not simultaneously measure both the optical QE spectra and the time evolution of QE at 1.50 eV; the measurement of QE should be stopped during the optical QE spectra experiments. Therefore, there were short gaps in the time course curve of QE (see Fig. 1 and 2).

III. Results and Discussions

Figure 1 shows the time course of the QE under the light illumination of 1.50eV. The horizontal axis represents time after the Cs supply started, the vertical axis shows the QE. The supply of Cs and O₂ is switched at the local maxima and minima during the NEA activation. The duration of Cs and O₂ supply is classified by color; yellow for Cs on and O₂ off, and green for Cs off and O₂ on, respectively. Cs deposition rate was estimated to be close to $0.015\text{ML}/\text{min}^1$. The O₂ partial pressure was estimated

to be $\sim 10^{-9}$ Pa during the supply. The supply of Cs and O₂ continued during the measurements of spectra. The QE at 0–30 min was almost at the background level, and no photoelectron emissions were observed even though the supply of Cs was continuous, and a slight QE peak was then observed at ~ 32 min after the start of Cs supply (from Fig. 2). The QE was again reduced to zero during the continuous supply of Cs; Cs source was completely switched off and O₂ was then introduced, and the QE showed an abrupt increase and saturation. Here, when the O₂ was switched back to Cs, the QE dropped to zero. Repeating this alternating supply of Cs and O₂ gradually increased the saturation value of QE. The average time of O₂ supply was 49min, and the average time of Cs supply except for first supply was 16min. The time of first Cs supply was 50min.

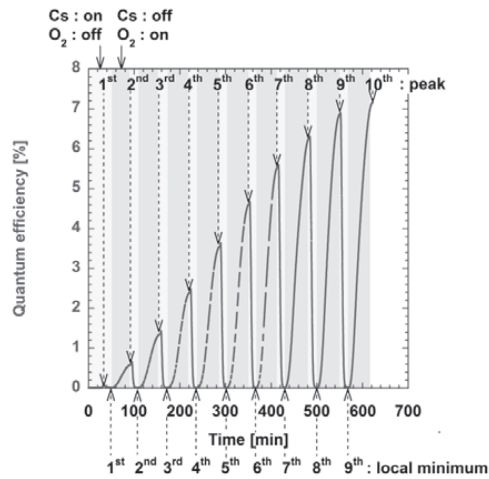


Fig. 1 The time course of the QE at 1.50eV during NEA activation. The yellow and blue background colors represent Cs on and O₂ off, and Cs off and O₂ on, respectively. The peaks and local minima of the yo-yo curve are marked in Fig. 1 to show the points where the QE spectra of Figs. 4 and 6 were measured.

Figure 2 shows the expanded yo-yo curve at 25–95 min. The increase of QE from 50 to 95 min represents the increase process from 1st local minimum to 2nd peak due to O₂ supply. The dashed arrows indicate the points where the QE spectra of Fig. 4 were measured. According to previous works²⁾, when the electron affinity was sufficiently small such as when adsorbing Cs on the clean GaAs surface, electrons excited by light below the vacuum level acquired the necessary energy for emission by phonon absorption. Therefore, it cannot be determined from the threshold of the spectrum alone that the NEA surface was formed. Meanwhile, the escape probability of electrons increased drastically corresponding to decreasing effective electron affinity after the NEA surface was formed^{3,4)}. This result implies that the QE rate of increase

rises rapidly. From the above, we determined that when the QE rate of increase changed from line1 to line2 (i.e., when line1 and line2 crossed), the NEA surface was formed. In other increasing paths by O₂ supply, the NEA region was determined by the same method. Consequently, it was found that the PEA surface was formed at all the local minima of the yo-yo curve, and changed to NEA surface due to the supply of O₂ with increased electron emissions, and the NEA surface was formed at all the local maxima of the yo-yo curve.

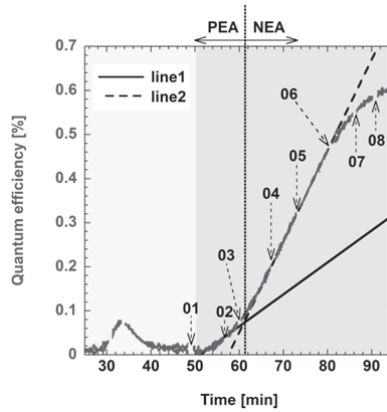


Fig. 2 Close-up of yo-yo curve from 25 min to 95min. The dotted arrows indicate that points where the QE spectra of Fig. 3 were measured. The NEA surface was formed after line1 and line2 crossed.

Figure 5(a) shows the QE spectra when the QE increased from the 1st local minimum to the 2nd peak. The labeled number of each spectrum corresponds to the numbers with dashed arrows in Fig. 2. The QE was indicated in logarithmic scale. As mentioned above, when the QE increased by the supply of O₂, O₂ pressure remained at no more than $\sim 10^{-9}$ Pa. In order to minimize the change of QE during spectral measurements, we carried out experiments with this partial pressure. Therefore, we did not consider that the amount of change of QE when the tendency of spectra in the NEA conditions was compared qualitatively. As seen in Fig. 3(a), the QE increased monotonically in all wavelength regions, and the threshold of the spectrum gradually shifted to lower energy. In 01 and 02 spectra, the photoelectrons were not measured from ~ 1.42 eV (the band gap of GaAs at room temperature). Therefore, the PEA surface was formed at points 01 and 02 as shown in Fig. 2. The photoelectron emission near the band gap of GaAs was observed at point 03. However, from the result of Fig. 2, the PEA surface was formed at point 03 and the NEA surface was formed when spectra 04–08 were measured. As seen in Fig 3(a) and (b), the QE of 04-05 spectra increased with the

change of the tendency of spectra and the QE of 06-08 spectra increased without the change of the tendency. From Fig. 2 and Fig. 3(b), it was found that although the change of the tendency of the spectra was stopped, the QE continued to increase after a short time. It is noted that similar results were obtained for other increase paths. It is considered that the increase of QE with the change of the tendency of spectra has close relation to structures and number of electron emission sites.

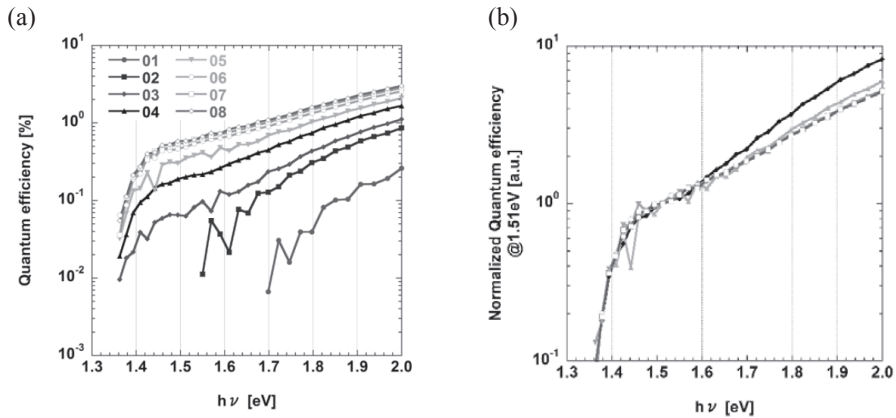


Fig. 3 The evolution of QE spectra (a) and the normalized spectra at 1.51eV (b) when the QE increased from 1st local minimum to 2nd peak. The labeled number of each spectrum corresponds to the labeled number in Fig. 2.

Considering that the Cs deposition rate was about 0.015 ML/min and the total time of Cs supply was about 178 min, the total amount of the Cs deposition was no more than 2.7ML in this study. From the above, the dipole model was applied to this NEA surface⁵). In the case of the dipole model, effective electron affinity decreased corresponding to the increase of QE although the NEA surface was formed. In addition, the escape probability of electrons increased drastically corresponding to decreasing effective electron affinity after the NEA surface was formed, and the QE spectrum depends on not only effective electron affinity but also electron escape probabilities. Therefore, the change of the tendency of the spectra was mainly due to the decreases effective electron affinity because of forming various structures of electron emission sites when the number of (Cs,O) dipole increased. The reason why the tendency of QE spectra stopped changing during the single increasing process was considered to be that the number of the sites formed was limited because the ratio of excessive supply of Cs to amount of O₂ supply was almost constant during this NEA activation. Considering the above, it is estimated that the increase in QE without the change of the tendency of spectra is closely related to the number of electron emission sites.

IV. Conclusions

We have qualitatively investigated behavior in variation of work function and the tendency of QE spectra during NEA activation. It was found that NEA and PEA surfaces were formed at the peaks and at the local minima of the yo-yo curve, respectively. When the photocurrent increased, the PEA surface changed to the NEA surface; electron emission continued to increase, and the characteristics of QE spectra changed once after the NEA surface was formed. At present, it is considered that the increase of electron emission might be closely related to the number of actual emission sites and that there are multiple types of electron emission sites.

In future experiments, we will investigate the adsorbed species in the NEA activation employing surface analysis techniques to discuss the Cs-adsorption states when the PEA surface changes.

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