Fluctuations of Electron Beams Emitted from Single-Atom Electron Sources Prepared with Different Techniques

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We compared fluctuations of electron beams emitted from single-atom electron sources, on which the surface Pd metals were deposited by two different techniques, vacuum deposition and electroplating deposition. The similar nano-pyramids were fabricated successfully by both the techniques, but the stabilities of the beam currents were different each other; the electron source prepared by vacuum deposition was far superior to those of electroplating deposition. The possible origin was discussed on the basis of AES analysis. [DOI: 10.1380/ejssnt.2008.11]

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I. INTRODUCTION

Recently, single-atom electron sources of noble-metal covered tungsten (W) tips have attracted a large attention as an electron source of high-performance electron microscopes because of their excellent emission characters such as high brightness, a demountable function, a self-repairing function, and presumably a long lifetime [1–3]. In a previous paper, we demonstrated an extremely high efficiency of the source in the practical electron optics [4]. Because of their importance in practical use, in this work, we have investigated the fluctuations of the currents emitted from these sources prepared by two different techniques.

It is well known that the thermodynamically stable structure of the crystal shows various-truncated shapes depending on the anisotropic surface free energy, and the new electron source discussed in this paper is one of the typical examples. The W {211} surfaces were stabilized largely by noble-metal coating in monolayer thickness, resulting in the formation of facetting structure at elevated temperatures. Therefore, tiny three-faced nano-pyramids grow spontaneously on the W (111) surface as shown in Fig. 1 [5]. Furthermore, the single atom was always situated at the top of the nano-pyramid even at high temperatures; if the atomic structures of nano-pyramid were destroyed by either field evaporation or ion bombardment, it return to the original form only by the heat treatment at 1000 K; namely, self-repairing [1].

The facetting phenomena of the noble metal-covered W surface was reported first by Cetronio and Jones in 1973 [6], and several works were done repeatedly so far by several researchers [7–10]. In these studies, the metal deposition was carried out by a vacuum deposition. In recent years, the electroplating deposition technique was invented by Kuo et al. [11] to reduce the troublesome preparation process in an ultra high vacuum (UHV) and to improve the throughput efficiency, which seems to be very attractive in the practical use. By the electroplating deposition just after the electrochemical etching of the W wire for sharpening, one can neglect two troublesome treatments of cleaning the W surface and vacuum deposition in UHV. Only heating at 1000 K in UHV is enough to fabricate the single-atom electron source in the case of the electroplating-deposition tip, which is the same treatment as that of the conventional FE source. In this work, we have measured the fluctuations of the currents emitted from these sources prepared by the different techniques. On the basis of the AES analysis of the electroplating W (111) surface at various temperatures, the possible origin of the difference in current fluctuation is discussed.

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FIG. 1: A schematic diagram of a single-atom electron source: The collimated electron beam is emitted from the single-atom at the top of a nano-pyramid, which is grown in (111) face of the W tip.
The field emission microscopy (FEM) experiments were done in the vacuum chamber with a base pressure of $5 \times 10^{-9}$ Pa shown in Fig. 2. The extractor electrode and the fluorescent screen were degassed heavily by electron bombardment prior to the FEM observations and current stability measurements. Vacuum deposition was carried out on the clean tip surfaces by using an evaporator of Pd metal installed in the connected UHV chamber. The tip was heated by passing the electric current through a W loop for mounting the tip, and the tip temperatures were measured by an optical pyrometer.

Concerning the preparation of the tips, first, we sharpened one end of a single-crystalline (111)-oriented W wire with a diameter of 0.127 mm by electrochemical etching with KOH (aq). Two kinds of W tips used in this work were prepared by either the vacuum deposition in situ or electroplating deposition in electrolyte as follows.

1. Vacuum deposition technique:

In UHV, we cleaned up the tip surface by flash heating up to $\sim$2000 K, and then deposited palladium (Pd) in a few atomic layers on the tip surface. The Pd-covered W tip was heated at 1000 K in UHV for fabrication of the single-atom electron source, which was confirmed by using FEM and Fowler-Nordheim (FN) plot measurements. As shown in Fig. 3, the FEM pattern of the clean tip exhibited a three-fold rotational symmetry determined mainly by the anisotropic work functions of the hemispherical (111) tip apex. The heat treatment of the Pd-covered tip caused confinement of the emission area owing to the facet formation at the tip apex as shown in Fig. 1; the confinement enhanced the emission current by $\sim$10 as shown by the FN plots in Fig. 3. The observed FEM patterns exhibited either three-fold rotational symmetry or circular one.

2. Electroplating deposition technique:

Metal deposition was carried out in a few atomic layers thickness on the W(111) tip surface by electroplating in electrolyte just after the electrochemical etching. Then, the metal-covered tip was mounted in the vacuum chamber, and heated to fabricate the single-atom electron source. Two FEM patterns in Fig. 4 were obtained either at the halfway to the single-atom tip or at the final single-atom tip; until the confinement of the emission area was complete, the FEM patterns were changed with regard to the number and position of spot by heat treatments, which differed from the case of the vacuum deposition. Once the confinement was realized, however, the emission patterns showed either circular one or three-rotational symmetries. The changes in FN plots during heating were not also simple as shown in Fig. 4. The figure at each FN-plot line represents an order of heat treatments at 1000K. In the early stage of the heating, the bright spots in FEM were random oriented, which corresponds to the lines 1, and 2. With repeating the heat treatments at 1000K, the large reduction of emission currents occurred as indicated by the lines 3, 4 and 5. Finally, the FN plots lines came back slightly as indicated by the lines 6, 7 and 8. In the final stage of the heating, one bright spot appeared in the (111) directions. The line 8 was thermodynamically stable, because no changes in both the FN plots and the FEM patterns were detected by further heat treatments.
III. RESULTS AND DISCUSSIONS

Figure 5 shows typical fluctuations of the different currents emitted from the single-atom tip prepared by the vacuum deposition technique. The current fluctuation in emission was plotted against the operating time. The currents of 1, 10, and 20 nA are almost constant being independent of the time. It is well known that conventional clean W tips showed the monotonous decrease in emission current owing to the chemisorption of residual gases even in UHV. The almost constant currents in Fig. 5 clearly indicate there is little influence of the chemisorption on emission. If we observe carefully the emission curves of 1, 10, 20 nA, however, we notice sometimes tiny fluctuations of stepwise or spike-like in those curves, which is a characteristic feature of the FE current from nano-tips [12]. The weak chemisorption might occur near the emission area.

In the case of 30 nA, the emission current decreased drastically stepwise. The relatively large emission such as 30 nA destroyed the surface structure, which were clearly indicated by changes in FEM pattern as shown by inserted pictures in Fig. 5: the circular bright spot changed to the triangle one that was frequently observed at the halfway of the fabrication process of the pyramid. The total emission of 30 nA corresponds to the huge current density, $3 \times 10^8 \text{ A/cm}^2$ at the topmost single atom; either heating or momentum transfer might kick off the topmost single atom. The self-repairing function made it possible to recover the initial single-atom termination, but the re-emission of 30 nA destroyed the topmost structure again. The cycles were confirmed for several times in this experiment.

We used more than 10 tips prepared by the electroplating deposition to measure the emission currents. These FEM patterns are similar to those of the vacuum-deposition combined with the field evaporation, we observed similar pyramids as that of the vacuum-deposition tips as follows; behind the topmost single atom, three atoms were situated, and ten atoms existed at the third layers. Below this structure, a triangle pyramid faced with three \{211\} faces was clearly observed as shown schematically in Fig. 1. Such a fundamental structure of the pyramid was the same as the vacuum-deposited one. However, not perfect symmetrical patterns presumably due to the surface defects were found more frequently as compared with the single-atom electron sources prepared by vacuum deposition.

Figure 6 shows the typical fluctuations of two emission currents of 1 and 10 nA from the single-atom tip prepared with electroplating deposition technique. As shown in Fig. 6, steps and spike noises appeared in all the observed data more frequently in the emission current than those of the vacuum-deposition ones. We did not succeed to find stable emission of the currents above a few nA, which differed from the stability of the vacuum deposition tips. The large difference between two techniques originates presumably from the circumstance of the metal deposition; namely, in electrolyte or vacuum. Hence, the chemical composition at the pyramids seems to be different between two kinds of pyramids. In order to clarify the chemical composition of the single-atom tip prepared with the electroplating technique we applied the LEED and AES analysis as a function of heating temperature as follows: We deposited the Pd metals of a few atomic layer thickness on a W(111) disk surface with a diameter of 10 mm, and heated it up to 1000 K in UHV. The treatment was the same as those of the electroplating tips. At the end of the heating, in fact, the faceting structure grew on the surface, which was confirmed by LEED observations as shown in Fig. 7. With increasing the incident electron beam energy, a part of the LEED spots move in the specular directions of the \{211\} facet faces as shown by the arrows in the LEED pattern. The surface chemical compositions were analyzed by using the AES analysis as a function of heating temperature. Figure 7 shows the results. We detected impurities of sulfur, carbon and oxygen; in particular, the sulfur increased at the final stage of the heating temperatures above 900 K. The sulfur atoms came from a nail polish, insulating layers, which was used to select the deposition area of Pd metal. Hence, those impurities were the most possible candidates for the origin of the large fluctuation of emission current in spite of no knowledge of the mechanism. We should remove those impurities from the tip apex, and it is in progress.
FIG. 7: AES analysis result of Pd-deposited W ⟨111⟩ surface, and superposed LEED pattern which was acquired from a series of 11 LEED patterns taken from 40 eV to 90 eV, 5 eV increment. With increasing the electron energy, a part of the LEED spots moved to the specular directions of the {211} facet faces as shown by the arrows in the LEED pattern. The peak height ratios of the surface elements to that of W are plotted as a function of heating temperature.

IV. CONCLUSIONS

We compared fluctuations in electron beams emitted from two different single-atom electron sources of Pd-covered W ⟨111⟩ tips in detail. While the stable emission up to 20 nA of the vacuum-deposition tip was confirmed, the emission currents from the electroplating ones were unstable even at 1 nA. The surface impurities such as sulfur, carbon and oxygen were detected in the AES spectra of the electroplating ones.

Acknowledgments

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