Demountable Single-Atom Electron Source

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We report emission properties of single-atom tips, noble electron sources terminated with a single atom. The repairing function of the sources, which had been already demonstrated by using field ion microscope [Fu, et al. Phys. Rev. B 64, 113401 (2001)], was confirmed by using field emission microscopy, and in addition, the demountable character was demonstrated repeatedly. The brightness of the collimated beam emitted from the single-atom tips was estimated to be an order of $10^{10}$ A/cm²/str. ($E \sim 2$ keV), which is two orders of magnitude higher than those of the conventional field emission sources. There appeared an extra shoulder in the energy spectra, which is strongly correlated with a single-atom ended structure. The fluctuation of the emission current exhibited stepwise and spike-like noises, which were typical features of nano-electron sources. The single-atom sources are promising candidates of the electron sources in high-performance electron-beam instruments. [DOI: 10.1380/ejssnt.2005.412]

Keywords: Demountable source; Single-atom source; High brightness; Coherent beam

I. INTRODUCTION

The coherence lengths of the electron beams are determined by the energy width of the beam and the emission area of the electron tip; the decrease in energy spread increases a temporal coherence length (a longitudinal-coherence length), and the decrease in emission area increases a spatial coherence length (a transverse-coherence length). In 1998, we reported that the energy spread of electrons emitted from a superconducting Nb tip is one order of magnitude lower than those of the conventional field emission (FE) tip [1] and nano-scaled tip of carbon nanotubes exhibits large improvement of the spatial coherence length because of the narrow emission area, $\sim 1$ nm [2]. Since the narrowest electron source is a single-atom electron source, Fink fabricated it by remodeling techniques [3], and Binh et al. developed a surface melting method to construct a nano-protrusion terminated with a single atom [4]. The similar structure was also fabricated by applying high electric fields to the contaminated tungsten ($W$) tip ($111$) [5]. However, those tips were not so stable enough to maintain the reasonable life time, and in addition, the reproducibility of the fabrication was very poor, which prevents from applications to practical use in scientific instruments with electron beams. Recently, Kuo et al. [6] and Rokuta et al. [7, 8] reported the electron emission from the noble single-atom tip, which exhibited not only high reproducibility of fabricating the nano-scaled structure, but also a demountable character and a repairing function. The noble electron source makes it possible to apply the practical use in electron microscopes and fundamental physics experiments for Fermion anti-correlation phenomena. In this paper, we have discussed the present situation of developing the noble single-atom tips based on the emission properties.

II. EXPERIMENTAL

The experiments were performed in ultra high vacuum (UHV) of less than $1 \times 10^{-8}$ Pa, and tip temperature was 50 K. The apparatus for field ion microscope (FIM), field emission microscope (FEM) and field emission spectroscopy (FES) used in this experiment are described in detail elsewhere [2, 5, 7]. The second chamber for electron projection microscope was used to confirm the demountable character of the single-atom tips. The single-crystalline $<111>$-oriented tungsten wire with a diameter of 130 $\mu$m is spot-welded on the bow-shaped tungsten wire. The tip was etched electrochemically. The tips were mounted in a UHV chamber, and the surfaces were cleaned by field evaporation. The detailed fabrication way of single-atom tips are described as related with the reported ones in the next paragraph.

III. THERMODYNAMICALLY STABLE TIP PRODUCED BY FACETING PHENOMENA

In thermodynamically equilibrium, the crystal takes up a shape that corresponds to the minimum of the total surface free energy. Hence, a stable crystal tends to be bounded by crystal faces with the low surface free energy. The equilibrium shape was speculated by minimization of total surface free energy, if the anisotropy of the surface free energy is given. In general, crystals rarely take up this equilibrium shape because they are prepared by non-equilibrium process. However, if a small crystal is heated close to the certain temperature, where atoms have sufficient mobility to rearrange, the crystal may approach the equilibrium shape. On can see the situation at the nano-scaled area at the top of the tip, where the surface
atoms are easily mobile to rearrange the atomic structure even at relatively low temperature in comparison with the melting point. On the hemispherical surface of clean W tip, heating at 700 K produces the growth of the faceting \{211\} faces near the \{111\} faces. When the tip is heated at elevated temperatures together with applying high electric fields, on the other hand, remolding phenomena occur, and as a result, the surface near the specific crystal orientation build up owing to the surface tension enhanced by the high electric fields. The build-up structure in the certain crystal orientations were constructed depending on the conditions of field strength and temperature. Since the nano-scaled structure was not thermodynamically stable, it returns easily to the original one irreversibly by heating or other treatments. The remolding single-atom sources were easily destroyed. The reproducibility of the shape and the crystal direction of the nano-protrusion fabricated were poor. Even the surface contamination causes build-up phenomena only by electron emission at room temperature [5]. As is discussed in the following, the fabrication method in this paper is different from the reported techniques including the remolding.

Adsorption of metallic elements such as Pt, Pd, Au and Rh enhanced the anisotropy of the surface free energy of the W surface. As a result, the \{211\} face of tungsten become stable and it tends to enlarge. The apex of the tip changes from a hemi-sphere shape to polygon-like one covered with the \{211\} facet faces. In 1997, the group of Madey reported that nano-triangle pyramids faced with the \{211\} facet faces grew on the (111) surface observed by STM [9]. By FIM, Fu and Tsong [10] found that the tops of the nano-pyramids were always terminated with the single atoms as shown in Fig. 1, and demonstrated that the pyramid was stable thermodynamically. In addition, they also elucidated that even if they destroyed the atomic structure at the apex by means of field evaporation, they always recovered the original apex with the single-atom termination by only using heating treatments at 1000 K. This repairing function is a promising character of the practical use as the electron source in electron microscope and the other scientific instruments with electron beam. The detailed FIM studies [10] of random walking phenomena of adatoms clarified that the activation energy of the adatom diffusion takes the minimum on the \{211\} face while it yields higher values on the other relevant faces. On the other hand, the potential barriers for the atom to escape from the single-atom site at the top are higher than 2.8 eV. Hence, while the low activation energy allows the adatom to move on the \{211\} face easily, which facilitates the adatom to transfer to the single-atom site, the high potential barrier prohibits the atom from leaving the single-atom site. Eventually, the structure is extremely stable and reproducible, as compared with the remolding and other ones terminated with a single atom.

IV. EMISSION PATTERNS AND ENERGY SPECTRA

Figure 2 shows typical FEM and FIM patterns of clean tungsten \(< 111\> \text{ tip} and single-atom tip of Rh. We have already confirmed the same results for the single-atom tips of Pd and Pt. The single-atom tips were fabricated with the known receipt: after deposition of Rh (or Pd, Pt) atoms in monolayer thickness on the W\(< 111\> \text{ tip} in ultra-high vacuum (UHV), we heated it at 1000 K for a few minutes. The shape of the tip apex was changed drastically. The atomic structure like a hemi-sphere shape changed to a faceting one; only single-atom was observed in the FIM pattern just after the heating as shown in Fig. 2; repeated-field-evaporation procedure clarified the atomic structure of the nano-pyramid. Just below the top single atom, three atoms are located, and the 10 atoms exit at the third topmost layer. The FIM patterns showed clearly that the nano-pyramid terminated with a single atom.
FIG. 3: FEM patterns exhibiting the repairing function of the single atom tip. Only the heating at 1000 K is enough to repair tiny defects such as absorption and desorption. Even if several atoms at the top were removed by field evaporation, the FEM patterns recovered always to the initial one only by heating. (A) the original beam from the virgin tip, (B) the first repaired tip. (C) the third repaired tip.

atom was constructed only by heating.

According to the structural change, electron emission pattern changes drastically. The collimated electron beam is emitted from the single-atom tip. Similar angular confinement was reported for the conventional single-atom tip constructed by remolding techniques [3, 4]. The large difference is the semi-cone angle of the collimated beams, the semi-cone angle of less than 1° (FWHM) of the present tip is much lower than 3−4° of the reported conventional ones [11]. The difference originates presumably from the structure of the pyramids; the {211} facet faces covers the pyramid in the present tip, while the pyramid prepared by remolding are surrounded by the {110} facet faces [11]. Table 1 shows the difference of specific angles between the tip axis, the <111> direction and the directions normal to the facet faces and to the ridge lines. The present tip is blunter than the reported ones, which results in two important features. One is the stability of the pyramids discussed in the next paragraph. The other is the beam diverse. The direction to surface normal to the ledge lines are much close to the <111> direction. Namely, the pyramid emits more collimated beam and realize a longer lifetime in comparison with the remolding one.

Moreover, the present sources are thermodynamically stable. Figure 3 showed the repairing function of the tips; after the removing top atom, only heating at 1000 K is enough to recover to the single-atom ended pyramid. We tried to repair it several times, and succeeded every time, of which the properties were discussed by Fu et al. [10]. On the contrary, the remolding tips were fabricated by using more complicated procedures including the W-atom deposition and heating processes.

In general, the repair function is the necessary requirement of the electron sources in practical use. For instance, the operating temperature at 1800 K of LaB$_6$ thermionic sources keeps the surface low work function to prevent from surface contamination. For Zr-W Schottky electron sources at operating temperatures of 1700 K, surface diffusion of Zr guarantees to maintain the Zr-W structure with the low function. The cold FE tips is heated every 12 h to keep a clean surface in UHV. They are all the suitable treatments to repair the emission site.

The other important advantage of the noble tip is its demountable character; the conventional single-atom tips were used in the same vacuum chamber, in which the fabrication was carried out. Hence, the applications were limited only to the electron projection microscope, not to the other scientific instruments with an electron beam. The situation was the same as the spin-polarized electron source of negative-electron-affinity devices, NEA-GaAs system, because the NEA condition can be maintained only in UHV of 10$^{-8}$ Pa. The source is used in the same UHV chamber, where the NEA surface is prepared. The remolding single-atom tip was so delicate that it should be kept in UHV and not to expose a poor vacuum. The noble tip covered with Pt, Rh, Au or Pd, however, is so inert that it can be exposed to air.

Figure 4 showed the FEM patterns of the tips with and without heating in UHV just after air exposure. The tips without heating showed complex FEM pattern owing to the surface contamination, but they recovered to the original FEM pattern exhibiting the collimated beam af-

### TABLE I: Specific angles of the directions normal to facet faces and ridge lines, which are referred to the $<111>$ direction.

|                | Facet face       | Ridge line       |
|----------------|------------------|------------------|
| Noble tips     | 19.5° {211} faces| 10.0°, the $<113>$ ridge direction |
| Remolding tips | 35.3° {110} faces| 19.5°, the $<11\overline{1}T>$ ridge direction |

FIG. 4: FEM patterns exhibiting the demountable character of the single-atom tip. FEM patterns obtained from the tip without and with heating at 1000 K after air exposure. Since the tip surface was covered with Rh (or Pd, Pt), the surface is relatively inert for reaction with various gases in air. The FEM pattern recovered to the typical one only by heating in UHV. (A) Just after air-exposure, and (B) after heating at 1000 K.
FIG. 5: Fowler-Nordheim plots of the same tip in different vacuum chambers. Although the geometric configuration of the single-atom tip and anode is different each other, the same collimated beam was obtained and similar FN plots were obtained.

FIG. 6: Typical energy spectra of the single atom tip for different applied voltages. The dotted line is the spectrum of the electrons from a clean W<111> tip.

V. STABILITY OF THE EMISSION CURRENT, BRIGHTNESS AND COHERENCE LENGTH OF THE COLLIMATED BEAM

In general, the fluctuation of the FE current originates from the adsorption of residual gas or/and ion sputtering of the residual gases. Hence, it changes largely depending on the conditions of tip surface, anode surface and vacuum pressure. In this experiments, the anode is MCP (a micro-channel plate), and the pressure of the chamber is about $10^{-8}$ Pa. Figure 7 shows the typical fluctuation of the emission current of the clean $<111>$ tip and single-atom tip of Rh. By the adsorption of the residual gas on the emission area of the clean W<111> tip, the emission current decreases monotonically as the time evolved. On the other hand, the emission from the single-atom tip is almost constant. This seems to be related with the high electric fields at the emission area; field evaporation prevents from the adsorption of residual gases. In Fig. 7, the noise of the single atom tip is step-like and spike-noises, which is typical emission noise feature of nano-tips [4]. The repaired tips exhibit the same noise character together with the same FEM pattern as those of the original.

FIG. 7: Typical changes in emission currents; (a) the clean W<111> tip, (b) a single-atom tip of Rh and (c) the single-atom tip, which was exposed to air and heated at 1000 K in UHV. The single-atom tips exhibit typical noise character of the nano-tips.
single-atom tip. From the parameters obtained in this experiment, we estimated the brightness of the collimated beam. The typical parameters were as follows, total current; \( I = 10 \) nA, the radius of an emission area; \( r = 0.1 \) nm and the semi-cone angles; \( \alpha = 1^\circ \) (FWHM) at \( 0.5 \times I_{\text{max}} \). Here, \( I_{\text{max}} \) means the maximum intensity. Under the assumption of a Graussian distribution, the brightness of the collimated beam was estimated to be \( 2 \times 10^{10} \) A/cm\(^2\)/str, where the accelerating voltage was \( \sim 2 \) kV. The brightness of a standard \( 150 \) kV FE microscope is on the order of \( 10^8 \) A/cm\(^2\)/str. The maximum Brightness \( B_{\text{max}} \) for Fermi gas is given by Silverman as follows \[12\],

\[
B_{\text{max}} = \frac{2meE\Delta E}{h^3}. \tag{1}
\]

By introducing the values, \( E = 2 \) keV, \( \Delta E = 0.2 \) eV, rest mass \( m \), electron charge \( e \) and Plank constant \( h \), \( B_{\text{max}} = 4 \times 10^{13} \) A/cm\(^2\)/str. Hence, the degeneracy, \( B/B_{\text{max}} \) is about \( 5 \times 10^{-4} \), which is the comparable to the reported values of the conventional single-atom tips \[13\].

Hence, one can expect much more improvement of brightness in conventional TEM and SEM. Concerning the coherence length, the improvement of spatial coherency is expected by a factor of \( 10^{-50} \) because of the narrow emission area in comparison with those of the conventional FE tips. The precise experiment by means of electron interference is also in progress.

VI. CONCLUSIONS

In summary, we showed peculiar emission properties of the demountable single-atom electron sources. The brightness was extremely high, and the spatial coherency of the beam is expected to be excellent. The demountable single-atom electron source is promising one used in the electron beam instruments such as transmission electron microscopy, scanning electron microscope, electron projection microscopy and electron diffraction microscopy.

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