Nanodots formation with slow highly charged ions

Yasunori Yamazaki

Graduate School of Arts and Sciences, University of Tokyo and RIKEN

We have been developing a compact electron beam ion source with a high-T$_c$ superconductor as a solenoid magnet, which can be operated at liquid nitrogen temperature, and can deliver slow highly-charged ions as high as $q = 42$. With this ion source together with other ion sources, nanodot formation processes were studied for a highly oriented pyrolytic graphite (HOPG) plate as a target. The impact site was observed with both the scanning tunnelling microscope (STM) mode and non-contact atomic force microscope (NCAFM) mode. It was found that protrusion-like dots were observed for both modes at the same position, and one HCI induced one dot. The dot size (diameter) and height were observed to be more or less the same for both modes, i.e. an HCI impact induces topographic modification on the HOPG surface. The dot size and height were measured as functions of the charge state ($q = 8 - 46$) and the kinetic energy ($E = 1 - 300$ keV) of highly-charged ions. It was found that the dot size increased linearly with the charge state, although the dependence on the kinetic energy was very weak if any.

1 Introduction

Interaction of slow highly-charged ions (HCIs) with metal, semiconductor, and insulator surfaces has been intensively studied in the last two decades$^{1-11}$ attracted by its exotic collision dynamics such as the formation and evolution of hollow atoms, "preferential" ionizing desorption of adatoms on the surface with HCIs, nano-size surface deformation/modification$^{12-16}$, etc.

![Fig. 1: A schematic view of atomic processes taking place when a slow highly-charged ions approaches a metal/semiconductor surface.](image-url)
HCIs can be characterized by two parameters, i.e., the charge state $q$, and the potential energy $\frac{Z_q}{q}$, the energy to be released when the HCI is neutralized. For example, $\frac{Z_q}{q}(Kr^{10+})$ is comparable to $\frac{Z_q}{q}(U^{10+})$ although $\frac{Z_q}{q}(Kr^{54+})$ is $\approx 3$ times larger than $\frac{Z_q}{q}(U^{54+})$. When a slow HCI approaches a metal surface, it is accelerated toward the surface due to its image charge, then at a certain distance, starts to capture valence electrons forming molecular orbitals, and eventually dives into the target releasing all its potential energy through emissions of Auger electrons and X-rays, a schematic view of which is drawn in fig. 1.

In this paper, we discuss the characters of nanodots formed on the HOPG surface due to deposition of potential energies of the slow HCl referring to our recent observations together with a compact electron beam ion source with high-T$_c$ superconductor solenoid.

2 Electron Beam Ion Source with a bulk High-T$_c$ Superconductor

An electron beam ion source (EBIS) is widely used as a unique source of highly charged ions (HCIs). Once ions are produced in an EBIS, they are radially trapped by the electric field of the electron beam, which is compressed by a strong magnetic field, and are successively ionized by the electron beam. The charge state evolution in an EBIS is characterized by the so-called ionization factor $J\cdot t$ which is the product of the electron current density $J$ and the trapping time $t$. Because the current density increases approximately in proportion to the magnetic field, a superconducting coil has been commonly used to produce a strong magnetic field. However, such a coil greatly increases the size of the apparatus requiring liquid helium. A compact EBIS, a mini-EBIS, was developed by Okuno, which employed a normal conducting solenoid soaked in liquid nitrogen. In order to develop a compact but high power EBIS with a superconducting solenoid, we have employed a new type of EBIT where a bulk high-T$_c$ superconductor was used as a solenoid, which yielded $\approx 0.8$ T. It is noted that a ring-shaped permanent magnet does not give a strong uniform magnetic field near the axis, because the magnetic flux tends to pass regions with high permeability.

![Diagram of High-T$_c$ EBIS](image)

Fig. 2: The cross section of the High-T$_c$ EBIS.

Figure 2 shows the cross-sectional view of the High-T$_c$ EBIS. The EBIS consists primarily of five parts, an electron gun, an assembly of superconductor rings, a magnetization coil, a drift tube, and a water-cooled electron collector. The electron beam from the electron gun is accelerated toward
the drift tube, compressed by the magnetic field, passes through the drift tube, decelerated to \(\sim 1\) keV, and finally accumulated on the electron collector. The drift tube consists of three successive electrodes and forms an electrostatic well, which axially traps ions in the central part of the drift tube. Ions over-flowed from the trapping region were accelerated by the potential difference between the drift tube and the electron collector, and extracted through the center hole of the collector (a leaky mode operation. The trapped ions can also be extracted actively by dumping the trapping potential quickly, which is called a pulse mode. The main parameters of the High-\(T_c\) EBIS are listed in the Table 1.

| Table 1: Major design parameters of the High-\(T_c\) EBIS |
|----------------------------------------------------------|
| Maximum electron beam energy | 30keV |
| Maximum electron beam current | 100mA |
| Maximum electron current density | 700A/cm\(^2\) |
| Drift tube length | 40mm |
| Ion extraction voltage | 0-5kV |

The design value of the current density is 700 A/cm\(^2\), so that neon-like uranium can be produced in a reasonable time period, such as 5 s or so. In general, the ionization cross section reaches its maximum when the electron energy is several times larger than the ionization energy of the objective ion. The electron energy of 30keV is about three times as large as the ionization energy of sodium-like uranium. In order to extract slow highly-charged ions, the bias voltage of the drift tube should be very low, and accordingly, the electron gun was designed to be raised to -30 kV at the maximum. The electron collector was designed to be biased to decelerate the electron beam to 1-2 keV at the collector, which reduces the heat load to the collector suppressing outgas and also the capacity of the high voltage power supply for the electron gun. The electron gun used is that developed for the Tokyo EBIT \(^{21}\), which has a spherically-shaped barium oxide cathode of 3 mm diameter with its perveance of \(\sim 4.4 \times 10^{-7} A/V^3/2\), i.e., 100 mA at 3.8 kV. The shape and position of the electron collector and the electric field configurations around it were optimized so that the electrons are efficiently decelerated and accumulated on the collector. The collector is cooled by distilled water to absorb a power load as high as 100 W. The inner diameter of the superconducting solenoid was 15 mm, while the outer and the inner diameters of the drift tube were 8 and 3 mm, respectively, with a trap length (i.e., the length of the center electrode of the drift tube) of 40 mm. A hole of 0.5 mm in diameter was drilled through the center electrode to introduce source gas into the trap region. The high-\(T_c\) solenoid was made of three ring-shaped superconductors, with their outer and inner diameters of 51 and 15 mm, respectively each having the thickness of 12 mm. The three rings were packed in a vacuum-tight stainless steel container, which was thermally connected to the inner wall of the liquid nitrogen reservoir via indium sheets. A normal conducting coil to magnetize the superconductor was installed in the liquid nitrogen reservoir as is shown in fig.2. The liquid nitrogen reservoir also works as a cryogenic pump, which helps to improve the vacuum around the drift tube. The capacity of the liquid nitrogen reservoir is about 6l, which is large enough to operate the EBIS continuously for more than 12 h without refilling.

The high-\(T_c\) superconducting material used was YB\(_{\text{a}}\)Cu\(_{\text{3}}\)O\(_y\), which was chosen because of its high critical current density \(^{22}\), larger size, and high machinability. A normal conducting pulse magnet was used to magnetize cooled superconductors (PFM: pulsed field magnetization. The magnetizing coil used in the present high-\(T_c\) EBIS contains 182 turns of a copper wire with a cross section of \(1.4 \times 3.0\) mm\(^2\) with its inner and outer diameters of 74 and 107 mm, respectively, the length of which
is 80 mm. A capacitor bank of 40 mF with a charging voltage as high as 1 kV was used as a power supply, which provides the maximum field of 7 T at the center of the coil for several tens of ms.

Figure 3 shows the charge state distribution of xenon ions, where at least three peaks are recognized for each charge state, corresponding to the natural abundance of major isotopes (129, 131, and 132). The highest charge state observed was 42 (magnesium-like Xe ion) because of relatively poor vacuum due to outgassing during operation. Actually, the combination of the electron beam energy and the current predicts the reduction of neon-like xenon ions. The number of ions measured after the exit aperture (2 mm in diameter) of the analyzing magnet was typically $\sim 10^2$ cps for highest charge states such as $\text{Ar}^{17+}$ and $\text{Xe}^{42+}$, $\sim 10^4$ cps for relatively high charge states such as $\text{Ar}^{14+}$ and $\text{Xe}^{31+}$, and $\sim 10^6$ cps for relatively low charge states such as $\text{Ar}^{8+}$ and $\text{Xe}^{26+}$, after gas pressure optimization.

3 Nanodot Formation on HOPG with Slow Highly Charged Ions

3.1 Experimental Setup

The ion sources used were the high-T$_c$ EBIS described in the previous section and the Tokyo EBIT depending on the charge state necessary for the experiments. The former can produce relatively intense but relatively low charged ions, and was used for charge states up to 40+. On the other hand, the latter can produce highly charged but relatively low intensity ion beam and was used for charge states higher than 40+. An HOPG target cleaved in the air with adhesive tape was irradiated with HCI from one of those ion sources. After irradiation, the HOPG sample was exposed to air, transported and set on a sample holder of the ultra high vacuum SPM (JEOL JSPM-4500A), which can be operated in both the STM and AFM modes. When a conductive cantilever is used, one can observe the same position of a sample both with the STM and AFM modes at atomic resolution without changing the tip.

3.2 Comparison between STM and AFM Images

Figures 4 (a) and (b) show the STM and AFM images of the HOPG target irradiated with 3keV/q Xe$^{46+}$ ions, respectively. The potential energy of Xe$^{46+}$ ion is 66keV. The sizes of the dots in the
Fig. 4: (a)STM and (b)non-contacting AFM images of the same dot observed with the same tip for 3keV/q Xe\(^{46}\) ions on an HOPG target. The right panels show the cross section of along the lines in the left panels.

STM/AFM images were almost the same with those reported by several other groups regarding STM images\(^{13,14,23,24}\). It was confirmed that the nano-dot formed by a slow HCI corresponds to topographical change on the surface\(^{15}\). It is noted that a contact-AFM observation is often not very reliable in discussing topological structures\(^{12,25}\). It is noted on the other hand that the tapping mode AFM was successfully applied to observe protrusions induced with 1keV Ar\(^+\) ions\(^{26}\).

### 3.3 Charge state and kinetic energy dependences

Figure 5 shows the incident charge state dependence of (a)the dot size and (b) the dot height observed with the STM mode\(^ {16}\). The height depends rather weakly on the charge state if any. On the other hand, the dot size increases more or less linearly with the charge states. The dot size and height were also measured as a function of kinetic energies, and did not show any appreciable dependence on the kinetic energy for 1-300keV\(^ {16}\). Consequently, it is concluded that the potential energy or the incident ion charge has more direct and strong influence on the nanodot formation rather than the kinetic energy at least for the kinetic energy range studied here, i.e., collision cascades do not play any important roles. The fact that the dot size does not depend on the kinetic energy but does on the potential energy strongly indicates that the potential energy is quickly released at or near the surface within a fs or so. To pin-down the time range peculiar to the potential energy deposition, an experiment with faster highly charged ions are in progress. It is noted that mica has also been intensively investigated with an SPM. In previous measurements\(^ {12,28}\), protrusion-like dots were observed also on a mica surface. According to their observations, the dot size and height on mica show similar behaviour as those on HOPG in the sense that the dot size was insensitive to the kinetic energy but depended strongly on the incident ion charge, and the dot height was less sensitive to both the kinetic energy and the incident ion charge. These similarities are very interesting because the surface modification mechanism is expected to depend essentially on the charactor of these materials.
Fig. 5: The nanodot size and height as a function of the charge state of highly-charged ions. The open squares, the open diamonds, the open triangles, and the solid squares are from Minniti, Mochiji, Hayderer, and Nakamura, respectively.

Acknowledgements

The work presented here was done in collaboration with N. Nakamura, M. Terada, S. Ohtani, Y. Kanai, A. Endo, and K. Komaki. Vivid and fruitful discussions with the members of Atomic Physics Laboratory are greatly acknowledged. This work is supported by Special Research Projects for Basics Science of RIKEN.

References

1) J. P. Briand, L. de Billy, P. Charles, S. Essabaa, P. Briand, R. Geller, J. P. Desclaux, S. Bliman, and C. Ristori, Phys. Rev. Lett. 65 (1990) 159.
2) H. Winter, Europhys. Lett. 18 (1992) 207.
3) H. Kurz et al., Phys. Rev. Lett. 69 (1992) 1140.
4) J. Das and R. Morgenstern, Phys. Rev. A 47 (1993) R755.
5) F. W. Meyer et al., Phys. Rev. A 48 (1993) 4479.
6) M. Grether, A. Spieler, R. Koerbrueck, and N. Stolterfoht, Phys. Rev. A 52 (1995) 426.
7) J. Burgdoerfer, P. Lerner, and F. W. Meyer, Phys. Rev. A 44 (1991) 5674.
8) S. Ninomiya, Y. Yamazaki, F. Koiki, H. Masuda, T. Azuma, K. Komaki, K. Kuroki, and M. Sekiguchi, Phys. Rev. Lett. 78 (1997) 4557.
9) K. Kakutani, T. Azuma, Y. Yamazaki, K. Komaki, and K. Kuroki, Jpn. J. Appl. Phys. 34 (1995) L580.
10) M. Sporn, G. Libiseller, T. Neidhart, M. Schmid, F. Aumayr, H. Winter, and P. Varga, Phys. Rev. Lett. 79 (1997) 945.
11) G. Hayderer, S. Cermsca, M. Schmid, P. Varga, H. Winter, F. Aumayr, D. Niemann, V. Hoffmann, N. Stolterfoht, C. Lemmell, L. Wirtz, and J. Burgdoerfer, Phys. Rev. Lett. 86 (2001) 3530.
12) D. C. Parks, R. Bastasz, R. W. Schmieder, M. Stoeckli, J. Vac. Sci. Technol. B 13 (3) (1995) 941.
13) K. Mochiji, S. Yamamoto, H. Shimizu, S. Ohtani, T. Seguchi, N. Kobayashi, J. Appl. Phys. 82 (1997) 6037.
14) T. Meguro, A. Hida, M. Suzuki, Y. Koguchi, H. Takai, Y. Yamamoto, K. Maeda, Y. Aoyagi, Appl. Phys. Lett. 23 (2001) 3866.
15) M. Terada, N. Nakamura, Y. Nakai, Y. Kanai, S. Ohtani, K. Komaki, Y. Yamazaki, Nucl. Instr. and Meth. B235 (2005) 452.
16) N. Nakamura, M. Terada, Y. Nakai, Y. Kanai, S. Ohtani, K. Komaki, Y. Yamazaki, Nucl. Instrum. Methods B232 (2005) 261.
17) T. Schenkel, et al., Phys. Rev. Lett. 83 (1999) 4273.
18) E. D. Donets and V. P. Ovseyannikov, Sov. Phys. JETP 53, 466 (1981).
19) K. Okuno, Jpn. J. Appl. Phys. 28, 1124 (1989).
20) N. Nakamura, A. Endo, Y. Nakai, Y. Kanai, K. Komaki, Y. Yamazaki, Rev. Sci. Instr. 75 (2004) 3034.
21) N. Nakamura, J. Asada, F.J. Currell, T. Fukami, K. Motohashi, T. Nagata, E. Nojikawa, S. Ohtani, K. Okazaki, M. Sakurai, H. Shiraishi, S. Tsurubuchi, H. Watanabe, Phys. Scr. T 73 (1997) 362.
22) M. Morita, M. Sawamura, S. Takebayashi, K. Kimura, H. Teshima, M. Tanaka, K. Miyamoto, and M. Hashimoto, Physica C 235/240, 209 (1994).
23) I. Gebescher, S. Cernusca, F. Aunayr, H. Winter, Int. J. Mass Spectrom. 229 (2003) 27.
24) R. Minniti, L.P. Ratliff, J. Gillaspy, Phys. Scr. T 92 (2001) 22.
25) Y. Sugawara, H. Ueyama, T. Uchihashi, S. Morita, M. Suzuki, S. Mishima, Appl. Surf. Sci. 113-114 (1997) 364.
26) B. An, S. Fukuyama, K. Yokogawa, M. Yoshimura, Jpn. J. Appl. Phys. 39 (2000) 3732.
27) G. Hayderer, S. Cernusca, M. Schmid, P. Varga, H. Winter, F. Aunayr, Phys. Scr. T92 (2001) 156.
28) C. Ruehlcke, M.A. Briere, D. Schneider, Nucl. Instr. and Meth. B 99(1995) 528.