Coincidence measurements of secondary ions and scattered ions in collision between slow Ar$^{6+}$ and a GaN(0001) surface

K Motohashi$^1$

$^1$Department of Applied Physics, Tokyo University of Agriculture and Technology, 2-24-16 Nakacho Koganei-shi, Tokyo 184-8588, Japan

E-mail: motohasi@cc.tuat.ac.jp

Abstract. An experimental apparatus has been developed to investigate interactions between highly charged ions (HCIs) and solid surfaces under off-normal incidence. The apparatus consists of a secondary ion mass spectrometer (SIMS) and a low-energy ion scattering spectrometer (LEIS) equipped with a Wien filter ($E \times B$ velocity filter). It allows us to carry out simultaneous SIMS and LEIS analysis. It successfully measured the time-of-flight SIMS spectrum in coincidence with scattered Ar$^0$ and Ar$^+$ in collisions between Ar$^{6+}$ (15.0 keV) and a GaN(0001) surface. Protons and hydrocarbons, N$^+$, Ga$^+$ and GaN$^+$ ions were observed in the SIMS spectra taken in coincidence with the LEIS spectrum of Ar$^0$ and Ar$^+$. The kinetic energy of Ar$^+$ measured in coincidence with protons emitted from the surface was distributed over an energy range from 13.6 to 15.0 keV. This fact directly shows that kinetic processes play a significant role in proton emissions.

1. Introduction

Interactions between slow HCIs and solid surfaces have been attracting much attention because of their possible application to surface nanotechnology. There have been many reports on nanostructure formation as well as efficient secondary particle emission, which make it possible to realize the surface-sensitive chemical analysis. For example, some papers described sputtering yields almost independent of collision energy [1]. Nano-dot volume as a function of the collision energy showed similar energy dependence [2].

However, the details of the reaction processes between slow HCIs and solid surfaces have not been clarified yet because it is very difficult to observe the kinetic and potential processes separately. Secondary-electron emission measured in coincidence with no kinetic energy loss of incident ions was reported by Stöckl et al. [3]. It was the first observation of pure potential emission of secondary electrons induced with HCIs. Though it has been generally recognized that the potential energy of HCIs is very important, especially in slow collisions with solid surfaces, it is very difficult to reduce the kinetic energy enough to suppress kinetic processes in actual experiments.

The off-normal or glancing incidence has two important characteristics that are essentially different from those of normal incidence, especially in HCI collisions with solid surfaces. One is that, a lot of surface atoms positioned along the ion trajectory interact with the HCI in glancing collisions because the electron capture starts from a critical distance, which is almost proportional to the initial charge state of the incident HCI [4], and it continues until just in front of the surface. Thus, sputtering or desorption may also occur at multiple different atomic sites. Another important characteristic is, of
course, a small kinetic energy supplied to the surface normal direction. The kinetic energy $E_\perp$ is given as follows [4],

$$E_\perp = E_0 \sin^2 \theta + E_{im},$$  \hspace{1cm} (1)

where $E_0$ is the initial projectile energy, $\theta$ is the angle between the initial velocity vector and the surface and $E_{im}$ is the image acceleration energy or energy gain. Although $E_{im}$ depends on the charge state of the HCI [5], the potential energy of Ar$^{q+}$ ($q \geq 3$) is greater than the calculated value of $E_\perp$ in collisions with metal surfaces with $E_0 = 2500 \times q$ eV and $\theta \leq 5^\circ$.

A novel experimental apparatus allowing us to achieve multi-coincidence measurements among scattered atoms/ions, secondary electrons and secondary ions has been developed to clarify the sputtering or desorption processes due to electronic excitation induced by slow HCIs [4–7]. This paper will report the first experimental SIMS results measured in coincidence with scattered ions whose charge state and kinetic energy were selected in collisions between HCIs and the solid surfaces.

2. Experimental apparatus and methods

The details of the experimental apparatus were described elsewhere [7], so a brief description is presented here. Figure 1 shows a schematic illustration of the experimental setup. Ar$^{6+}$ ions extracted from a compact electron beam ion source (EBIS) were accelerated by 15.0 keV. After their focus and trajectory are adjusted with an electrostatic lens (L) and four parallel plates ($P_{1 \perp}, P_{1 \parallel}, P_2$ and $P_3$), the ions collide with the target surface at an angle, $\theta \approx 5–10^\circ$, tangential to the surface. The velocity and charge state of the scattered ions or atoms of Ar$^{q+}$ ($0 \leq q' \leq 6$), that capture $(6-q')$ electrons from the surface, were analysed with the Wien filter (WF) and the charge separator (CS), respectively. When each scattered ion or atom hit the detector, three signal waveforms from the position-sensitive anode ($D_1$) were recorded using a digital storage oscilloscope (DSO). One can analyse the position ($y, z$) (see the coordinates in Figure 1) by a simple arithmetic operation on the pulse heights of the waveforms.

The WF works as a velocity-dispersion filter; i.e. the spacing of exit slits extends along the $z$ direction. The displacement along the $z$-axis of the sensed position of a scattered ion with respect to that of a neutral atom reflects its charge state and velocity. Secondary ions emitted from the surface were detected with a multichannel plate (MCP, $D_2$). The difference between the time-of-flight of the secondary ion $T_1$ and that of scattered atom/ion $T_2$, or $t_{12} = T_1 - T_2$, was also recorded with the DSO event by event.

![Figure 1. Schematic illustration of experimental setup](image-url)
Figure 2 shows projectile-beam profile converted from the two-dimensional (2D) position images that were taken with the D1 detector. The origin of the \(z\)-axis was set at the peak position of the 15.00 keV beam profile. The peak position of the 15.20 keV beam was shifted to the +z direction by 0.9 mm to maintain a width of ~2.5 mm (FWHM). The relationship between the energy and the displacement, i.e. the \(E-z\) calibration factor of the WF, was found to be \(2.2 \times 10^2\) eV/mm. The energy resolution was evaluated to be \(5.5 \times 10^2\) eV. The energy resolution was not so good because of the wide (1 mm) entrance slit of the WF. A few structures observed in both spectra may be due to the potential distribution at the ion trap region in the EBIS. Preliminary energy analysis of the primary ion beam will be required to achieve high-resolution measurements.

3. Results and discussion

Figure 3 shows the 2D-position image of \(\text{Ar}^+\) scattered at the GaN(0001) surface upon incidence of \(\text{Ar}^{6+}\) (15.0 keV) at \(\theta \approx 10^\circ\). The vertical axis \(z\) can be converted to the kinetic energy of \(\text{Ar}^+\) passing through the WF, as described above. The horizontal axis \(y\) may be related to the longitudinal scattering angle of \(\text{Ar}^+\). However, the angle cannot be determined at present because its calibration has not been accomplished yet.

Figure 4 shows the kinetic energy distribution of \(\text{Ar}^+\). The distribution, converted from Figure 3, corresponds to the LEIS spectrum. The width (FWHM) of the \(\text{Ar}^+\) spectrum is ~2.6 times wider than that of the primary \(\text{Ar}^{6+}\) one. Thus, significant energy loss, up to ~2.0 keV, was found in the \(\text{Ar}^+\) spectrum, and the loss peak was located around 0.6 keV.

Figure 5(a) shows a 2D-correlation plot between the displacement \(z\) of scattered \(\text{Ar}^{q+}\) (\(q' = 0, 1\) and 2) and the time difference \(t_{12}\) (see section 2) of secondary ions. Figures 5(b) and (c) show the integrated intensity of Figure 5(a) along the vertical and horizontal axis, respectively. Figure 5(b) is almost the same as the usual SIMS spectrum, since \(T_1\) is much longer than \(T_2\). Very efficient proton emission is one of the most important characteristics of the primary \(\text{Ar}^{5+}\) ionization due to electron capture by HCl ions [1, 4–7]. Weak \(\text{N}^+, \text{Ga}^+\) and \(\text{GaN}^+\) peaks originating from substrate atoms were also observed, together with some hydrocarbon-ion peaks originating from contaminants. Figure 5(c) corresponds to the
The charge state distribution of scattered Ar\(^{q^+}\). The populations of the spectra with \(q^' = 0, 1\) and \(2\) were 66\%, 23\% and \(\leq 11\%\), respectively. Figure 5(d) shows the 2D-correlation plot between the time difference \(t_{12}\) of protons and kinetic energy \(E\) of scattered Ar\(^{+}\). The vertical axis of this figure was converted from the displacement \(y\) of Figure 5(a). The scale of the horizontal axis of figure 5(d) is limited to the time range corresponding to the proton peak of Figure 5(a) or (b). It was found that the \(E-t_{12}\) distribution slightly but clearly leaned, as shown by a red line. This fact confirms that \(t_{12}\) of protons changed with the kinetic energy of Ar\(^{+}\), simply because \(T_2\) depends on the kinetic energy. The energy loss of Ar\(^{+}\) measured in coincidence with protons was distributed in the range of 0–2 keV, the same as in the LEIS spectrum of Figure 4. This finding directly shows that some kinetic processes of substrate atoms play a role in proton emission processes.

4. Conclusion
Simultaneous LEIS and SIMS analyses was achieved with a new experimental apparatus using collisions between Ar\(^{q^+}\) and GaN(0001) at an off-normal incidence angle. It was found that some kinetic processes involving surface substrate atoms play a role in proton emission.

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