Secondary ion emission from a KCl(001) surface by grazing-angle incidence of swift ions

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Abstract. The yield and the mass distribution of secondary ions emitted from a KCl(001) surface were measured, when various MeV ions (0.8 MeV He⁺, 1.34 MeV Li⁺, 2.1 MeV B²⁺, 3 MeV O²⁺, 1.5 MeV Si⁺, 6 MeV Si³⁺) were incident on the surface at grazing angles θi ranging from 1 to 6 mrad. Positive secondary ions such as Cl⁺, K⁺, K₂⁺ and small clusters, K(KCl)ₙ⁺ (n = 1–4) were detected coincidentally with specularly reflected projectile ions. Negative secondary ions, Cl⁻, (KCl)Cl⁻, (KCl)₂Cl⁻, were much fewer by ~100. The secondary-ion production rate P(x) as a function of the distance x between the projectile and the surface atomic plane was derived from the θi-dependence of the yield of the positive secondary ions. P(x) showed the overlinear dependence on the electronic stopping power S(x) for specularly reflected projectile ions. The power index varies from ~1 to ~3 depending on S(x).

1. Introduction
When a solid target is bombarded with high-energy ions, the constituent atoms, molecules, clusters and their ions of the target are emitted from the surface in many cases. This phenomenon is well-known as sputtering. Electronic sputtering, which results from electronic excitation caused by ion bombardment, has been studied for a number of pairs of high-energy ions and targets (oxides, alkali halides, metals) [1-4].

When high-energy ions are incident on an atomically flat surface of a crystal at a grazing angle, ions are scattered at the angle for specular reflection with neither close collisions with the target atoms nor penetration through the surface. In the specular reflection the ions travel through the target electrons outside the surface atomic plane and lose the part of their energy by electron excitations on the surface. These features of the specular reflection of high-energy ions are favorable for the study of electronic sputtering, because only the surface electrons are excited and the contribution of nuclear sputtering is ideally ruled out even with bombardment by no very high-energy ions. Additionally, the trajectories of the specularly reflected ions are well-defined simply with the ion-surface repulsive potential, and so the impact parameter and the density of the consequent interactions (energy deposition, secondary electron emission, etc.) can be controlled by the variation of the angle of incidence of the ions.

In the present study, the yield and the mass distribution of secondary ions emitted from a KCl(001) surface was measured at grazing-angle incidence of various MeV ions. The secondary-ion production rate as a function of the distance between the projectile ion and the surface was estimated for each
kind of projectile ions, and the relation between the production rate and the stopping power was investigated.

2. Experimental
Beams of 0.8 MeV He⁺, 1.34 MeV Li⁺, 2.1 MeV B²⁺, 3 MeV O²⁺, 1.5 MeV Si²⁺, 6 MeV Si³⁺ from a tandem-type accelerator at QSEC in Kyoto University were collimated to less than 0.1×0.1 mm² and to a divergence angle less than 0.1 mrad by a series of 4-jaw slits. The typical beam current was less than 1 pA. The collimated ion beams were incident on a KCl(001) surface at a grazing angle of 1–6 mrad from the surface plane in a ultra-high-vacuum (UHV) scattering chamber (base pressure 4×10⁻⁸ Pa). The surface of KCl(001) was prepared by cleavage in air and cleaned by heating at 250°C for 1 hour in the UHV scattering chamber (~1×10⁻⁷ Pa). The KCl(001) target was at 180°C during the bombardment to keep the surface clean and to avoid macroscopic charging.

The ions reflected specularly from the surface of KCl(001) were selected by a aperture (Ø = 1 mm) placed 411 mm downstream from the target and their energies were analyzed using a 90° sector magnetic spectrometer and a 1-dimensional position-sensitive detector (1D-PSD). The energy analysis of reflected ions only with most probable charge state [5] was performed. For example, only O⁴⁺ ions were energy analyzed for the incidence of 3 MeV O²⁺. This can be excused by the fact that specularly reflected ions already reach their equilibrium charge distribution after many charge exchanges [6].

Positive secondary ions emitted from the surface were collected with electric field between the surface and a negatively biased grid (−1.5 kV) right in front of the surface. The secondary ions passing the grid travel through a drift tube (~0.4 m) and are detected by a micro-channel plate (MCP, effective diameter Ø = 20 mm). Negative secondary ions were also detected only in the case of 3 MeV O²⁺ incidence. The mass to charge ratio (m/z) of the secondary ions is analyzed with the time-of-flight technique, in which the signals from the 1D-PSD (specularly reflected ions) and the MCP (secondary ions) are used as the start and stop signal, respectively.

3. Results and discussion
Figure 1 shows an example of the energy spectrum of projectile ions specularly reflected from the KCl(001) surface. The angle of incidence \( \theta_i \) of O²⁺ ions was 3.0 mrad from the surface plane. The energy spectrum of O⁴⁺ ions through charge exchange with without reflection on the surface is also shown as the primary energy of incident ions. The energy of reflected ions forms a relatively narrow peak at ~2.93 MeV. This peak corresponds to the ions reflected from the surface without surface penetration or a close collision with the target atoms. Thus, this confirms that the secondary ions observed coincidentally with the detection of specularly reflected ions are generated only by electronic sputtering.

![Figure 1. Energy spectrum of O⁴⁺ ions reflected from the KCl(001) surface for a grazing-angle incidence of 3 MeV O²⁺ ions.](image1)

![Figure 2. Mass spectrum of positive secondary ions from the KCl(001) surface bombarded with O²⁺ ions at \( \theta_i = 3.0 \) mrad.](image2)
Figure 2 shows a typical mass spectrum of positive secondary ions from the KCl(001) surface for
the incidence of O\textsuperscript{2+} ions at 3.0 mrad. The abscissa is \( m/z \), where \( m \) is the mass of the secondary ion in
the atomic mass unit (amu) and \( z \) is the magnitude of electric charge of the secondary ion in the unit of
elementary charge (e). The spectrum has several peaks assigned to Cl\textsuperscript{+}, K\textsuperscript{+}, K\textsuperscript{2+} and K(KCl)\textsubscript{n}\textsuperscript{+} (\( n = 1\)–4) ions. Secondary ions with higher charge states or with the larger mass \( (m/z > 400 \text{ amu/e}) \) were
below the detection limit.

Emission of negative secondary ions was also observed for 3.0 MeV O\textsuperscript{2+} incidence. Cl\textsuperscript{−}, (KCl)Cl\textsuperscript{−} and (KCl)\textsubscript{2}Cl\textsuperscript{−} ions were detected, but the total yield of negative secondary ions was only about one-hundredth of
that of positive secondary ions. This dominance of positive secondary ions indicates that the emission of secondary ions should be initiated with local charging due to the emission of hundreds of secondary electrons per projectile [7].
\[ Y(\theta) = Q \int_{x_{\text{tra}}.} P(x) \, dx \]  

where \( P(x) \) is production rate of secondary ions per unit path length of a projectile traveling at the distance \( x \) from the surface atomic plane of KCl(001) and the coordinate \( z \) is along the trajectory of the projectile. \( Q \) is the absolute detection efficiency of MCP. \( Q \) is now assumed to be 0.5. To substitute the equation for trajectory of the projectile to equation (1) gives an Abel-type integral equation, which can be solved if \( V_p(x) < V_p(x_{\text{min}}) \) at any \( x > x_{\text{min}} \) [8] as

\[ P(x) = -\frac{1}{2\pi Q E} \cdot \frac{dV_p(x)}{dx} \left( Y(0) \sqrt{\frac{E}{V_p(x)}} + \int_0^{\pi/2} \frac{dY(\theta)}{d\theta} \left[ \frac{V_p(x)}{E} \sin \theta \right] \, du \right) \]  

where \( E \) is the kinetic energy of the projectile, \( x_{\text{min}} \) the distance of the closest approach of the projectile to the surface and \( V_p(x) \) the surface continuum potential acting on the projectile. Molière potential is used as the ion-atom interaction potential for \( V_p(x) \) and the polynomial expression of the fitting curve (shown by solid lines in Figure 3) is adopted as \( Y(\theta) \), to obtain \( P(x) \) with equation (2). In the same way, the position-dependent stopping power \( S(x) \) for specularly reflected projectile ions were derived from the observed \( \theta \)-dependence of the energy loss \( \Delta E(\theta) \) [9].

The obtained \( P(x) \) and \( S(x) \) decrease almost exponentially with the distance \( x \), but the rate of decrease of \( P(x) \) is larger than that of \( S(x) \). Figure 4 shows \( P(x) \) as a function of \( S(x) \) for each kind of projectile ions. The distance \( x \) as a parameter is limited to the range from \(-0.4 \) to \(-1.4 \) Å, where the closest distance between the projectiles and the surface atomic plane ranges for various angles of incidence. The production rate \( P(x) \) seems to be proportional to power of \( S(x) \) for each. The cross marks connected with dashed curve show the values at a constant distance \( x \) of 0.6 Å. Along this curve, the power index decreases from \(-3 \) to \(-1 \) as the stopping power \( S(x) \) increases from 25 to 180 eV/Å.

### 4. Conclusion

Secondary-ion emission from a KCl(001) surface was observed when various MeV ions are incident on the surface at grazing angles of 1–6 mrad. Positive secondary ions, such as \( \text{Cl}^+ \), \( \text{K}^+ \), \( \text{K}_2^+ \) and \( \text{KCl}_n^+ (n = 1–4) \), are dominant in yield over negative secondary ions, such as \( \text{Cl}^- \), \( \text{KCl}^- \) and \( \text{KCl}_2^- \), which indicates that secondary-ion emission should be initiated with local charging due to secondary-electron emission. The yield of positive secondary ions increases with \( \theta \), while energy loss of the reflected ions is almost independent of \( \theta \). Position-dependent production rate \( P(x) \) for positive secondary ions decreases almost exponentially with the distance \( x \) between the projectile and the surface. \( P(x) \) shows the overlinear dependence on \( S(x) \). The power index for the dependence of \( P(x) \) on \( S(x) \) decreases from \(-3 \) to \(-1 \) as \( S(x) \) increases.

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