Detection System for KeV Proton Beam Scattered from an ESD-surface with Monolayer Pits of Alkali Halide Crystal*

Yuuko FUKAZAWA*1†, Ryoiki NAKAGAWA*1 and Yasufumi SUSUKI*1

*1Division of Science Education, Osaka-Kyoiku University, Kashiwara, Osaka 582-8582

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To observe specular oscillations in ion-surface scattering originating from layer-by-layer desorption of an electron stimulated desorbed (ESD) surface, a 15 keV proton beam was impinged on the surface with an angle of incidence less than 1°. We prepared an experimental system to examine the scattering yields of reflecting protons by conveniently employing a fluorescent screen to view scattering yields (scattering patterns) and a commercial digital camera for yield measurements. Because the present energy of the proton beam was too low to illuminate the screen, a micro-channel plate was inserted in front of the screen. Measurements with electron irradiation show obvious damping specular oscillations; the widths of angular distributions of scattered protons simultaneously oscillated with increasing electron fluence. From the measured periods of the oscillations, the ESD rates of thermal desorption from KBr(001) at 1.5 keV electrons irradiation were obtained for sample temperatures.

1. Introduction

Desorption induced by electronic transition (DIET) in solids is important in not only vacuum technology but also many other scientific fields concerning materials science. Electron stimulated desorption (ESD) of cleaved alkali halide surfaces is one of the most studied DIET systems over the past 40 years. Historically, the study was noticed the sputtering by electrons, which is as a part, complements against luminescence among many radiation effects in ionic solids. The desorption yields and kinetic energies of desorbed atoms have been measured using particle detection systems, such as time-of-flight techniques. It was revealed that the kinetic energies of emitted alkali atoms are thermal energies, whereas those of halogen atoms contain two components: thermal and non-thermal. These two components have been studied from various aspects, including velocity and angular distributions, and their origins were ascertained in the 1980s.

In the 1990s, Hoche et al. examined specular yield oscillations using the He atom scattering technique when the surfaces were irradiated by photons, where the damping oscillations provided evidence of well-known layer-by-layer desorption. At the end of the 1990s, Szymonski et al. observed damping oscillations of the yields of thermally desorbed alkali and halogen atoms (thermal component), which provided evidence of layer-by-layer desorption of the ESD surface. Both Hoche et al. and Szymonski et al. respectively showed that rectangular pits grow on the surfaces by using methods of decoration carbon replica and atomic force microscope measurements. These recent measurements reveal the following scenario:

Numerous F center (an electron at an anion site) and H center (an interstitial molecular halide ion) pairs are created from Frenkel defects by an external electron when the surface of the alkali halide is irradiated by electrons having energy of a few keV. A non-thermal halogen atom is immediately emitted by excitation of the surface excitons; however, it is small fraction of the total halogen atoms, when the current density of electrons is low. A thermal halogen atom is emitted through an H center. Both processes for halogen emission do not cause any change on the surface because their sources contain atoms other than halogen atoms on the topmost surface.

In contrast, F centers recombine with alkali atoms to emit them. For flat surfaces, F centers aggregate at the surface to form stored F centers. Aggregation of F centers in the subsurface layers called X center, which is a hypothetical object and should be able to initiate formation of small pits (initial pits) on the surface. By further irradiation of electrons, more atoms are emitted by the movable F* centers (F centers in their excited state) diffused in the crystal. The F* centers can easily recombine with alkali ions of the low coordinated sites on the topmost layer as compared with those of the topmost normal sites. Halogen atoms are also emitted through H centers when they are not trapped by aggregated F centers or X centers at the surface. By expanding the area of each pit or by increasing the number of pits, removal of the first layer proceeds. Thus, the morphology of the surface develops as the irradiation fluence increases; roughly speaking, this behavior of the pits is repeated during the removal of several layers.

The oscillatory behavior is evident and the scenario seems reasonable, whereas the complex scenario leads us to further questioning and increases our interest. Despite the abundance of published data and articles, reports measuring the absolute yields of emitted atoms per an incident electron are limited. The existing studies only report on a few situations such as sample temperature or...
the electron beam energy. The damping oscillations have also been shown for typical situations but not shown as a systematic compilation.

We recently measured reflected protons at electron-bombarded KBr(001) surfaces with grazing incidence and have observed specular oscillations by the layer-by-layer desorption\(^{12,13}\). In previous works we used 550 keV protons from an accelerator. Decreasing the energy of the incident protons enables us similar measurements over a wide range angle of incidence. In this study, we prepared an experimental setup to observe the oscillation using a 15-keV proton beam from an ion-source and measure the ESD rates of KBr(001). This method is facile compared with the works that employ an accelerator in that respect the transportation path of the proton beam is short.

2. Experimental

2.1 Preparation of the Experimental Apparatus

A schematic diagram of the experimental setup is shown in Fig. 1. The main part of the experimental setup was described elsewhere\(^{14,15}\); thus, only a brief outline and the parts modified for this experiment are discussed.

The ion beam from an RF ion source was transported to an ultra-high vacuum (UHV) chamber, evacuated by an ion pump via a high-vacuum (HV) beam-duct and an HV chamber, evacuated by oil diffusion pumps with alkyl-di-phenyl-ether oil series and via a differential pumping system chamber, evacuated by a turbo-molecular pump. Two sets of X-Y slits were placed upstream of the HV and UHV chambers, respectively. The base pressure of this system was approximately \(1.5 \times 10^{-4} \) Pa at the HV chamber, \(7 \times 10^{-6} \) Pa at the differential chamber, and \(4 \times 10^{-7} \) Pa at the UHV chamber.

A Faraday cup and a simple image intensifier shown in Fig. 2(a) were equipped downstream of the UHV chamber to transport and collimate a 15 keV proton beam from the RF ion source\(^{15}\). The inset shows an illustration of the incident beam spot profile before collimation. In the beam spot profile of \(3 \times 3 \) mm\(^2\), dark lines originated from a stainless mesh, used as a secondary electron emitter, are visible\(^{15}\). To observe the incident beam spot profile after collimation and scattering of protons from the target surface, the image intensifier was replaced by a fluorescent screen made on the ITO glass plate, assembled with a micro-channel-plate (MCP) (F2807; Hamamatsu Photonics), shown in Fig. 2(b).

2.2 Experimental procedure using the collimated beam of protons

After the rough beam collimation to be \(1 \times 1 \) mm\(^2\), sample KBr was inserted into the UHV chamber. The KBr sample \((25 \times 22 \times 4.5 \) mm\(^3\)) was cleaved along the...
(001) surface in air and was mounted on a sample holder with a 4-axis goniometer renewed from the fixed sample heater previously mentioned\(^\text{16}\). The sample holder was a flat Cu plate with two leaf springs to press hold the sample and a thermocouple (Thc) set fasten near the sample. The holder was heated by a crushed flat spiral sheath-heater assembled in it.

To clean the surface, the sample was heated to 425 K and maintained at this temperature for 24 h (simultaneously baking the UHV chamber). It was then heated to 525 K and maintained for 1 h. The surface of the sample was irradiated by 1.5 keV electrons from a gun with an incidence angle of 60° measured from the target surface. At the entrance of the UHV chamber, the electron beam was collimated by a 1.5 × 10 mm\(^2\) slit and horizontally scanned by a 260-Hz magnetic field. The electron beam was also scanned vertically by another 22-Hz magnetic field to produce uniform irradiation (modulated line scan)\(^\text{12,13}\). The electron beam in front of the sample was spread over 30 × 30 mm\(^2\) by the scanning magnetic fields. The current density impinging on the sample surface was 0.02 μA/cm\(^2\), which was monitored using FC with a fluorescent screen operating as an electron suppressor. The temperature of the sample heater was monitored by the Thc on the sample holder and the room temperature. The heater temperatures are shown in this work. Errors in the temperature values were estimated within a few K.

Interrupting the electron irradiation, the 15 keV proton beam collimated to 0.5 × 0.5 mm\(^2\) by two sets of XY slits was impinged on the surface with angles, \(\theta_i\), of 11 ± 3 mrad. The beam current from the XY slits (≈5 nA from the downstream slits) was monitored by a data logger and used for normalization of the incident proton intensity. The incident beam currents remained below 30 pA. The scattering patterns on the fluorescent screen (MCP) were observed by capturing images with a commercial digital camera (Canon EOS 60D). The images were obtained by the summation of more than 10 pictures for each condition. The intensity distributions of the scattered patterns were obtained from B values of the RGB color values of the images. It required 13 min to take each distribution set for the irradiation fluence\(^\text{12,13}\).

Since the numbers of the incident particle to MCP were large compared with the counting rates of MCP that are generally used as particle detection measurements, the bias voltage to the MCP was suppressed. Thus, the light intensity might be under the linear function of the yield of protons.

2.3 Scattering pattern and section profiles

Figure 3(a) shows an example of the portraits measured by the digital camera, and Fig. 3(b) and (c) show its section profiles along the axis perpendicular and parallel to the surface, respectively.

Although the scattering beam was observed with bias voltages of 850 and 1700 V for an MCP and secondary electron acceleration to the fluorescent screen, the incident beam was observed by decreasing the voltage to 600 and 1200 V, respectively. Although the incident beam was measured within a very short time (1/30th of the scattered one), the portrait in Fig. 3(a) was composed of those of the incident and scattering beams. The angle of incidence was measured to be 11.5 mrad, which is half of the separation between the two beam profiles.

3. Results and discussion

Figure 4 shows the yields of specularly reflected protons (specular yields) on the KBr(001) ESD surface. The present data shown in Fig. 4(a) are total scattering proton yields normalized by the slit currents to suppress sway by beam fluctuation. The data in Fig. 4(b) are specular yields normalized by the total scattering yields (specular yields/total scattering yields shown in Fig. 4(a)).
Fig. 4 Examples of the yield oscillations with the electron fluence for a few target temperatures. (a) Total scattering yields. (b) Specular yields normalized by the total scattering yields.

4(a)). Both yields oscillate with increasing electron fluence and show that layer-by-layer mode desorption proceeds. Thus, the monolayer pits created on the KBr surface by the ESD decrease the total scattering yields and increase the angular distribution. This broadening of angular distribution is also observed as in Fig. 5(a), where the width increases with the number of desorbed atoms (area of pits) and decreases again for the flat surface. The broadening can be observed along the direction perpendicular to the surface; however, it cannot be observed along the direction parallel to the surface as shown in Fig. 5(b). This angular broadening is new information for inspecting the surface morphology of the ESD surface. The oscillations in Figs. 4 and 5 are damping, and desorption rates can be obtained from the periods of oscillations. That is, the inverse of periods of oscillation corresponds to the desorption rate.

Desorption rates obtained from the oscillations are shown in Fig. 6 as the numbers of desorbed atoms averaged in a period of oscillation per incident electron. The number of halogen atoms desorbed by the non-thermal desorption, constitutes an insignificant fraction and they are emitted from the subsurface layers. The alkali atoms desorbed by the thermal desorption are from the topmost surface only. Although the source of halogen atoms desorbed by the thermal desorption is not the topmost surface, a halogen and alkali di-vacancy is left behind on the topmost surface by the recombination of an F* center with an alkali atom (the scenario mentioned in the Introduction)8–11). The desorption rates obtained from the number of atoms on the topmost surface and the number of incident electrons in a period correspond
to the thermal desorption component independently of non-thermal desorption.

These yields have been compared with those obtained by other groups. The desorption rate at 400 K is interpolated from our measured rates to be $11 \pm 3$ atoms/electron, which is high compared with Such et al.\cite{8,9}, who obtained $7.3 \pm 0.7$ atoms/electron under 1 keV electron irradiation with an incidence angle 45°. Although our data have comparatively large error bars, the total number of desorbed atoms seemed to increase with electron energy in this region. In contrast, a previous study reported that the relative halogen yields slightly decrease with the increasing energy of incident electrons from 1 to 2 keV\cite{17}. However, the cited results appeared to be scattered, the angle of incidence of electrons was 45°, halogens were measured at 90° with respect to the surface, and the temperature of the surface was not shown except an information that it was above 90°C\cite{17}. The present comparison of the total number of desorbed atoms does not support the decreasing dependence; however, further research is needed for measuring the electron energy dependence of the total desorbed atoms (thermal component).

4. Conclusion

To observe specular oscillations originated from the layer-by-layer desorption of a KBr(001) ESD surface, a 15 keV proton beam was impinged on the surface with a small angle of incidence. A detection system employing a fluorescent screen and digital camera was prepared to observe the scattering yield of reflected protons. The measurements show obvious oscillations in specular yields and widths of scattering angle distributions perpendicular to the surface. From the measured periods of oscillations, the ESD rates of thermal desorption per 1.5 keV incident electron were obtained for a few target temperatures. The convenience our system provides is the use of a fluorescent screen to observe the scattering patterns and a commercial digital camera to obtain yield measurements. In this experiment using keV proton beam, the beam current is very stable as compared with that of sub-MeV proton beam from an accelerator\cite{12,13}. This can be attributed to small beam losses during transport in this system.

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