

Atomic Structure Analysis of BaO Layers on the Si(100) Surface by Impact-Collision Ion Scattering Spectroscopy

Yeon Hwang*

*Department of Materials Science & Engineering, Seoul National University of Technology,
Seoul 139-743, Korea*

Abstract

BaO layers were formed on the Si(100) surface by thermal evaporation of barium metal with simultaneous oxidation. The atomic structure of BaO layers at the initial stage of the deposition was investigated by the scattering intensity variation of He⁺ ions on time-of-flight (TOF) impact-collision ion scattering (ICISS). The results show that several number of BaO layers are formed on the Si(100) surface with the lattice parameter of bulk phase, and the occupation of oxygen atoms of the BaO layers is on-top site of silicon atoms.

It is known that the adsorption structure of the alkaline earth metals such as barium plays an important role in the epitaxial film growth of perovskite structured or superconducting materials on silicon substrate.¹⁻³⁾ Especially, deposition technology of high dielectric constant materials such as BaTiO₃ on silicon is important to achieve high integrated chip. The atomic structure of the BaTiO₃ film on Si(100) surface has been studied by using transmission electron microscopy (TEM),¹⁾ and reflection high-energy electron diffraction (RHEED),^{2,3)} However, the TEM specimen preparation is difficult and destructive, and the analysis of the RHEED pattern is complicated.

Low-energy ion scattering spectroscopy (ISS) has been established as an effective tool for the structural analysis of solid surfaces.^{5,6)} The excellent surface sensitivity of ISS comes from the efficient ion neutralization process at a surface. Inert-gas ions such as He⁺, Ne⁺, and Ar⁺ are often used as an ion beam because they are readily neutralized via Auger neutralization process, showing no target-element dependence of the neutralization probability.⁷⁾ In ICISS where the scattering angle is nearly 180°, geometrical structures of several layers deep can be analyzed. Also simulations of the scattering trajectories are simplified because the trajectory taken by

the scattered ions is almost identical with that of incident ions and the blocking effects by adjacent atoms can be avoided.

In this study, the atomic structure at the initial growth stage of the BaO layers on the Si(100) surface was examined using ICISS. The crystal structure of BaTiO₃ is composed of alternates of BaO and TiO₂ layers. The lattice constant of BaO and Si is 0.552 nm and 0.543 nm, respectively. This gives a very small lattice mismatch of 1.6%. So the deposition of BaTiO₃ film on silicon should be initiated with the deposition of BaO layer, and it is expected that the analysis of the atomic structure of the interfaces of BaO and Si is important.

BaO layers were grown on the Si(100) surface by metal evaporation and oxidation in the ultrahigh vacuum (UHV) chamber attached with TOF-ICISS. Silicon wafer of which resistivity was 6~12 Ω cm was cut into 15×15×0.5 mm size substrate, and was cleaned with acetone. Silicon substrate fixed to a tantalum holder was introduced into UHV chamber via load-lock system, and the base pressure of the chamber was kept lower than 2×10⁻⁹ Torr. The surface of the silicon substrate was cleaned by heating the tantalum holder at 600°C for 12 h and by subsequent flash heating at 1000°C for several seconds by electron bombardment until sharp RHEED pat-

tems appeared. The temperature of the surface was monitored with infrared optical pyrometer (Minolta, TR-630). BaO layers were deposited on the silicon substrate heated at 400°C by evaporation of the barium dispenser with oxygen gas flowing. During Ba evaporation, oxygen gas was flowed into the chamber keeping the base pressure at 2×10^{-8} Torr.

TOF-ICISS equipment is composed of the ion generator, the transfer line for pulsed ion beam, and the detecting system of scattered particles. The 2 keV He⁺ ions were generated in a discharge type ion source, and they were mass analyzed by a Wien filter. The primary energy of the ions can be changed from 0.5 to 3 keV. Energy of the back-scattered ions or neutral atoms was analyzed by measuring the flight time from the sample to the detector. The ion beam was chopped by the electrostatic deflection plates, which produced a pulsed beam with a full width at half maximum of 40 ns. The scattered particles (ions and neutral atoms) were detected by a microchannel plate (MCP) placed at a distance of 69 cm from the sample. All spectra were measured for 20 s at the sample current of 20 nA by varying the incident angle from the surface by 2° step.

Figure 1 shows Ba and Si peak intensity variations in the ICISS spectra as a function of incident angle along the [001] azimuth of the bulk silicon substrate. When the ion beam is projected onto an atom, a region called the shadow cone where projectile ions cannot penetrate because of the Coulomb repulsion is generated toward the opposite direction of the incident beam. The shadow cone can be calculated using the Thomas-Fermi-Moliere (TFM) potential with the screening length proposed by Firsov.⁸⁾ The target atoms residing at the surface of the shadow cone show an extremely large scattering intensity owing to the large flux density focusing on the incident ions at the surface of the shadow cone. This focusing effect produces an intensity variation of the scattered ions according to the incident angle⁵⁾ as shown in Fig. 1. Several intensity peaks from barium at 8°, 18°, 28°, 32°, 58°, 64° and 80° appear when deposition time exceeds 140 min.

The calculated shadow cones of barium and oxygen are shown in Fig. 2 for the rock salt structure

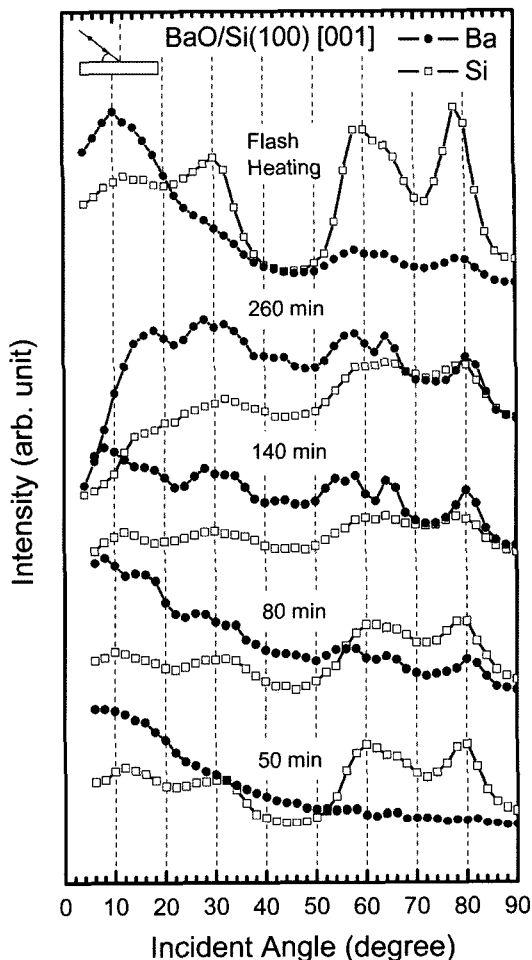


Fig. 1. Polar angle scan of Ba and Si peak intensities in TOF-ICISS at the BaO deposited Si(100) substrate along the bulk [001] azimuth. The incident angle is from the surface.

of BaO along the [001] azimuth. The intensity peaks of 10°, 32°, 59°, 64° and 80° appear from the calculation, and these are well coincident with the measured ones within an experimental error. That the calculation was performed using the lattice parameter of bulk BaO results in the same lattice parameters between BaO layers and bulk phase. When BaO was deposited for 50 min, the focusing effect of Ba appears only at 10° of incident angle and no focusing peaks from deeper layers are shown. Furthermore, the focusing peaks from Si substrate are still shown. This means that BaO is formed in monolayer on the substrate. As the dep-

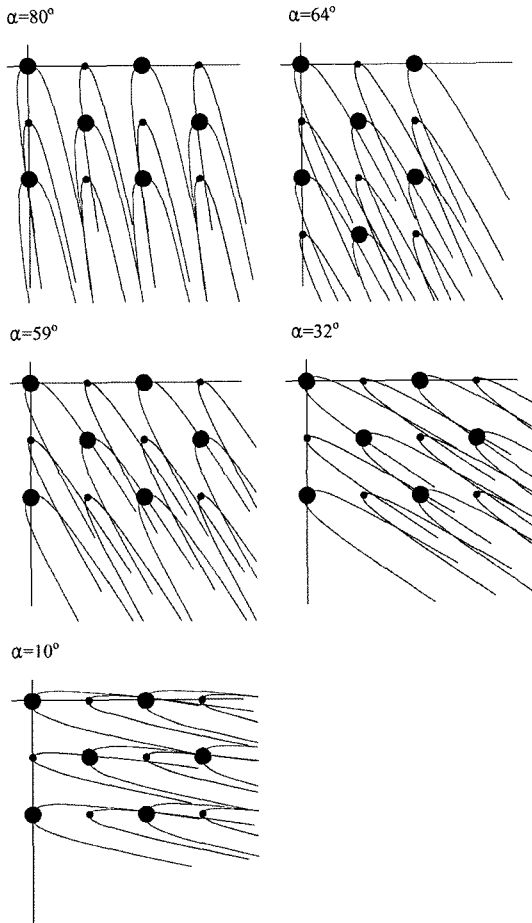


Fig. 2. Schematic view of the shadow cones for 2 keV He⁺ ions impinging on the BaO(100) surface along the [001] azimuth. Small and large circles represent oxygen and barium atoms of rock salt structure, respectively.

osition time of BaO layers is prolonged, several intensity peaks from deep layers of BaO such as at 32°, 58°, 64° and 80° appear. The focusing peaks at 18° and 28° in the Fig. 1 can be explained as has arisen from steps of the BaO layer. Figure 3 shows the calculated shadow cones when step exists on the surface. The barium atoms at the edge of the step have caused the focusing effect on the other barium atoms at the lower terrace at 18° and 27°. This implies that the BaO layers on the Si(100) surface have many steps.

The focusing peaks from the Si substrate do not disappear even at the longest deposition time in Fig.

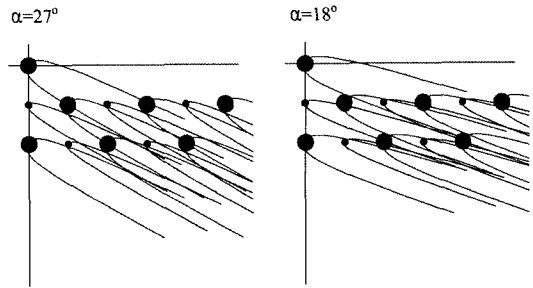


Fig. 3. Schematic view of the shadow cones showing the focusing peak at the steps of BaO(100).

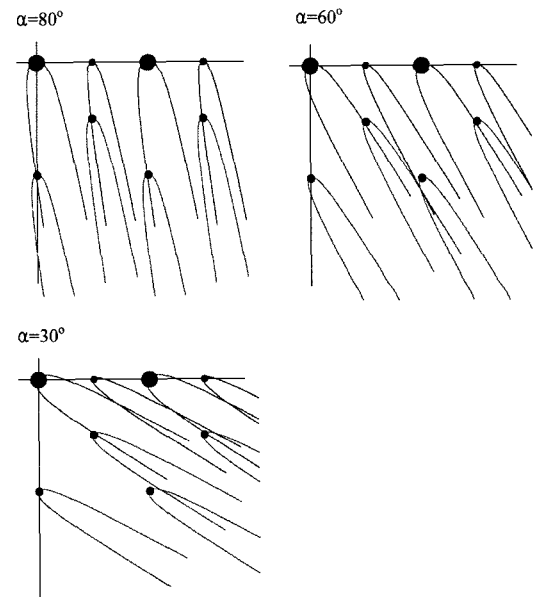


Fig. 4. Schematic view of the shadow cones showing the focusing effect on the silicon substrate. Small and large circles on the top layer represent oxygen and barium atoms of rock salt structure, respectively. The circles on the deep layers represent silicon.

1. Peaks especially from deep Si layers such as at 60° and 80° are well discernable. As ICISS can see deep layers, the atoms of the substrate can be detected if the thickness of the film is very small. Figure 4 shows the calculation for the structure of monolayer of BaO on Si. In the figure, it must be noted that atoms at the top layer are Ba and O, and those at the second and third layers are Si. The shadow cones from barium atoms on the topmost layer are focused on the silicon atoms at 30°, 60°

and 80° . Therefore, we can see that the structure of BaO layers on the Si(100) substrate is composed of various number of layers with steps. One important conclusion from the Fig. 4 is that the deposition site of oxygen is on-top position of the Si atoms and barium atoms occupy the vacant site, which is a similar result to the barium occupancy at valley-bridged or silicon-vacancy sites in Ba-Si(100) structure confirmed by scanning tunneling microscopy.⁹⁾ Furthermore, the (2×1) dimmer structure of Si(100) surface seems to be reduced to (1×1) structure because the simulation of the shadow cones using the bulk structure of silicon coincide with the experimental results.

It is notable that BaO layers are not removed completely after flash heating at 1200°C as the large focusing peak at 10° from Ba remains in Fig. 1. BaO is a refractory material with very high melting point, and the flash heating temperature is too low to eliminate the BaO layers. This characteristic implies us that the BaO layers are formed in complete rock salt structure with properties of bulk.

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