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ORIGINAL PAPER

Characterization of Ion Beam Deposited ^{107}Ag Thin Films on Si(111) Surface by means of Rutherford Backscattering Spectroscopy and Reflection High Energy Electron Diffraction

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The crystalline quality of the ^{107}Ag films on Si(111) surface grown by mass-separated low energy ion beam deposition (IBD) has been studied by means of combined techniques of reflection high energy electron diffraction (RHEED), low energy electron diffraction, and Rutherford backscattering spectroscopy (RBS). Although crystalline quality of the films hardly depends on Ag ion energy in the range of 30–100 eV for RHEED patterns, the RBS-channeling spectra clearly show the difference in the minimum channeling yield χ_{min} and the increasing rate of the channeling yield $d\chi(z)/dz$. The Ag ion energy to obtain a well-crystallized Ag film is found to be 30–50 eV. In addition, it is shown that excellent quality of the Ag films grown at elevated temperature can be obtained by means of IBD.

KEYWORDS: ion beam deposition, isotope, thin films, silver, silicon, growth, morphology, Rutherford backscattering spectroscopy, channeling, electron diffraction, Auger electron spectroscopy

I. Introduction

Ion beam deposition (IBD) is a thin film growth technique using low-energy ion beam. One of the advantages of this technique is to make it possible to fabricate the isotopically purified thin films. However, there is a possibility to generate and/or anneal out a defect by the incident ion beam. The defect concentration depends on the ion beam energy and the substrate temperature. Recently, there are several reports that the crystalline quality of silver films can be improved by high-energy ion irradiation because of the Ag grain growth.[1–3)] Additionally, Thomas *et al.* have been found by means of reflection high-energy electron diffraction (RHEED) that there is no strong energy dependence for the surface crystalline quality of the Ag film in the energy ranges of 25–125 eV for Ag^+ ion.[4)] However, the details of the ion energy dependence and the crystalline quality inside the Ag films are still unknown.

The purpose of the present study is to identify the energy and substrate temperature dependence of crystalline quality for isotopically-purified silver thin films on Si(111) surface by means of RHEED, low energy electron diffraction (LEED), Auger electron spectroscopy (AES), and Rutherford backscattering spectroscopy with channeling (RBS-channeling).

II. Experimental

The low-energy and mass-separated IBD apparatus used in this study consists of an ion source, a beam transport line, a deposition chamber, an analysis chamber and a preparation chamber, as illustrated in **Fig. 1**, and the technical specifications are given in **Table 1**. In this study, Ag^+ ions were generated in Ar gas plasma in arc chamber where AgCl solid was vaporized. After the positive ions were accelerated up to -30 keV, $^{107}Ag^+$ ion beam was extracted at a 90° mass-analyzing sector magnet in the beam transport line. Since the mass resolution of the magnet $M/\Delta M$ is 120, the resolution is high enough to separate $^{107}Ag^+$ and $^{109}Ag^+$ ions, as shown in **Fig. 2**. Mass-separated ion beam is focused by a vertical deflection electrode and an Einzel lens. High-energy neutrals are removed at the 7° deflection electrode. The $^{107}Ag^+$ ion beam was converged and decelerated at three deceleration electrodes just before the deposition chamber down to appropriate energy of 10–200 eV and deposited to a target substrate. The ion beam profile was adjusted by using ion beam profile monitor which equipped 8×8 pin grid array of electrodes. The film thickness was estimated *in-situ* from the gross current of ion beam, and was also examined by RBS analysis. Specimens can be heated up to 1,000°C using a resistance heater.

The specimens used were mirror-polished *p*-type Si(111) wafers, 10–20 Ωcm with a size of $27\times27\times0.5$ mm^3. The Si(111) wafers used had a vicinal angle of 0.5–5%. The specimen was dipped in warmed 5% HF solution for 2 min after ultrasonic cleaning in acetone, and then brought into the deposition chamber whose base pressure was 2×10^{-7} Pa. The surface residual contamination was not detected except carbon which showed the Auger signal less than 1%.

The $^{107}Ag^+$ ion beams of 10–200 eV in the energy range were deposited on Si(111) surface at RT, 200 and 300°C. The pressure during deposition was typically 2×10^{-5} Pa, of which main component was Ar gas. The Ag films deposited were 80–200 nm in thickness and typically ~10 mm in diameter.

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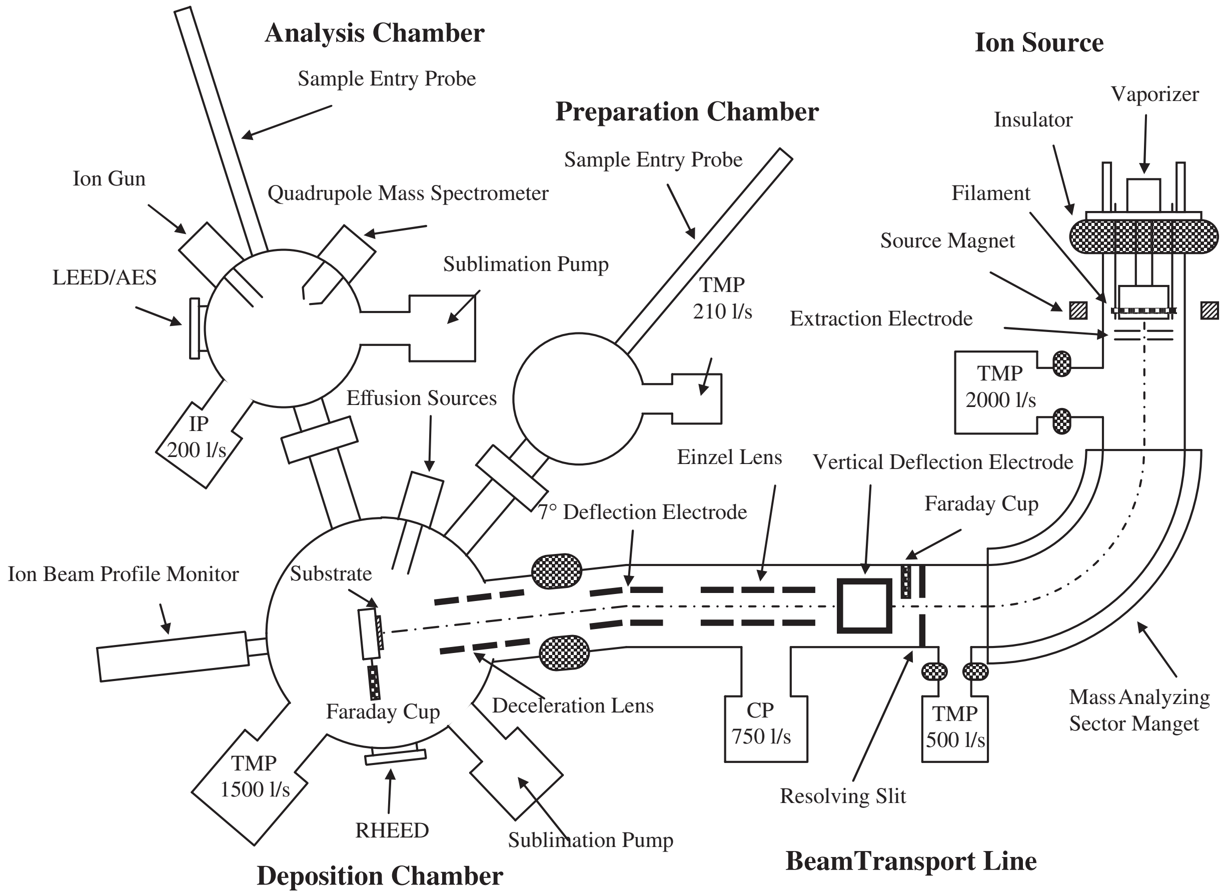

Fig. 1 Schematic illustration of the IBD experimental system

Table 1 Technical specifications of the low-energy ion beam deposition system

Ion source	Freeman type
Acceleration voltage	-30 kV
Analyzing magnet	Deflection angle=90° Maximum field=1.2 T
Mass resolution ($M/\Delta M$)	120
Ion energy	0–1,000 eV
Beam current	100 μA at maximum (Ag^+, 100 eV)
Beam size	≈10 mm in diameter
Deposition chamber pressure	2×10^{-7} Pa base pressure
In-situ analysis techniques	RHEED, LEED, AES, SIMS

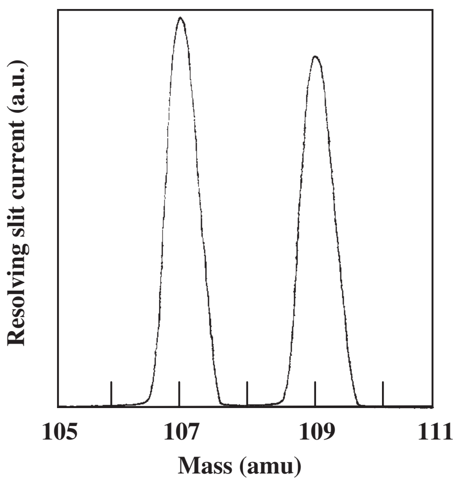

Fig. 2 Mass dependence of the Ag^+ ion beam intensity

The ^{107}Ag films were analyzed *in-situ* by RHEED in the deposition chamber, and then analyzed by LEED and AES in the analysis chamber. The RBS-channeling measurements were carried out in a different UHV chamber that was connected to a beam line of a 1.5 MeV Van de Graaff accelerator. The specimen was mounted on a three-axis rotatable goniometer with two-axis linear motion and an angular resolution of 0.025°. The backscattered He^+ ions were detected with a silicon surface barrier detector at scattering angle of 150°, with an energy resolution of 12 keV in full width at half maximum. The details can be found in Ref. 5).

III. Results

Silver films were formed on Si(111) surface using the $^{107}Ag^+$ ions in the energy ranges of 10–200 eV. The LEED patterns showed Ag(111)1×1 surface, as shown in **Fig. 3**, and the surface contaminations such as O, C, and Si were not detected by AES. On cleaned Si(111) surface, the Ag films grew epitaxially even at the ion energy of 10 eV,

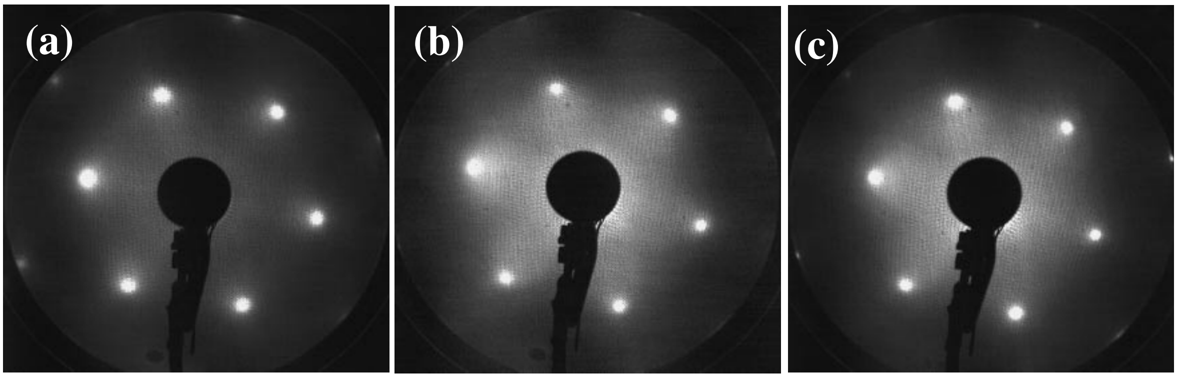

Fig. 3 LEED patterns (E_p=110 eV) for ^{107}Ag film/Si(111) prepared with 30 eV $^{107}Ag^{+}$ ions at the substrate temperature of RT (a), RT with post-annealing at 300°C (b), and 300°C (c)

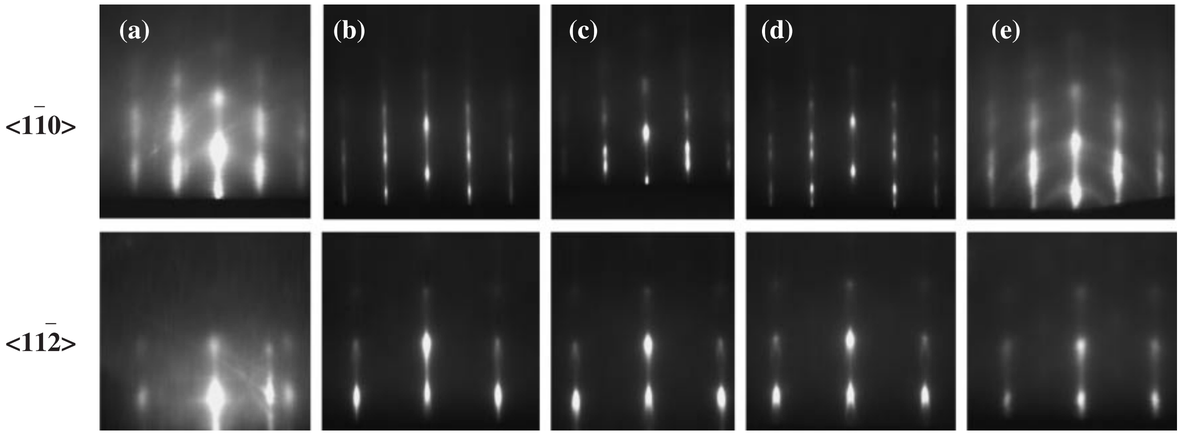

Fig. 4 RHEED patterns for ^{107}Ag/Si(111) prepared with 10 eV (a), 30 eV (b), 50 eV (c), 100 eV (d), and 150 eV (e) ions at the substrate temperature of RT

Azimuthal directions of incident electron are [1$\bar{1}$0] and [11$\bar{2}$] for upper and lower RHEED images, respectively.

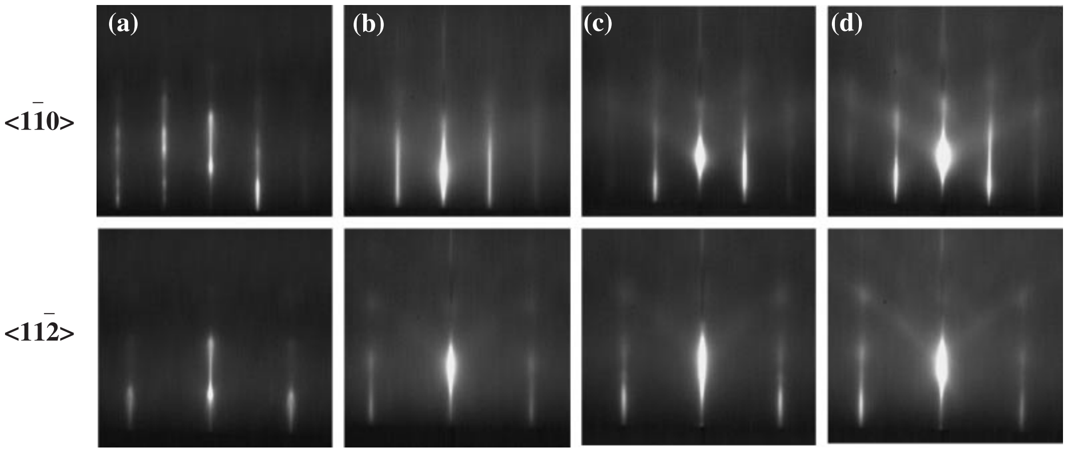

Fig. 5 RHEED patterns for ^{107}Ag/Si(111) prepared with 30 eV Ag ions at the substrate temperature of RT (a), RT with post-annealing at 300°C (b), 200°C (c), and 300°C (d)

Azimuthal directions of incident electron are [1$\bar{1}$0] and [11$\bar{2}$] for upper and lower RHEED images, respectively.

although the ion energy had to be higher than 30 eV for epitaxial growth at the native-oxide covered Si(111) surface. In the case of RT growth, twin spots were clearly seen in the RHEED patterns along [1$\bar{1}$0] and [$\bar{1}$10] direction, as shown in **Fig. 4**. This means that the abundance ratio was comparable with Ag[1$\bar{1}$0](111)//Si[1$\bar{1}$0](111) (so-called A-type) and Ag[$\bar{1}$10](111)//Si[1$\bar{1}$0](111) (B-type), and the twining plane was found to be (11$\bar{2}$). The RHEED patterns prepared with 30–100 eV ions indicated the better crystalline quality, and we found no differences in background intensity and spot

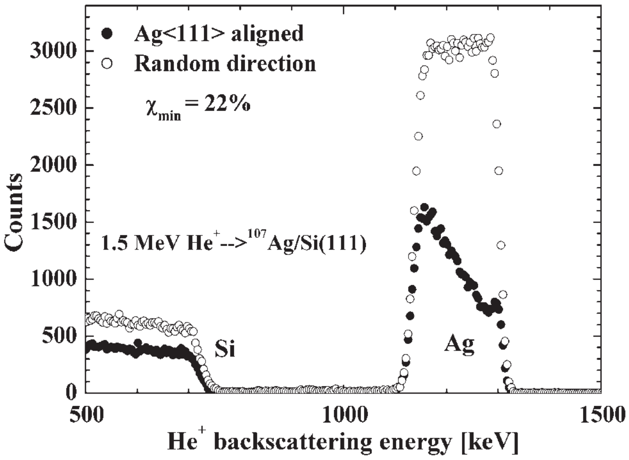

Fig. 6 RBS-channeling spectra of Ag⟨111⟩ aligned and random direction for ^{107}Ag/Si(111)
The film was prepared with 30 eV ^{107}Ag^{+} ions at the substrate temperature of RT.

Table 2 Ion energy dependence of the crystalline quality for ^{107}Ag films on Si(111) surface at the substrate temperature of RT

Ag^{+} ion energy (eV)	30	50	100
Ag⟨111⟩ χ_{min} (%)	22	20	32
$d\chi(z)/dz$ (%/nm)	0.28	0.27	0.30

shape in this energy ranges. At the energy of 150 eV and above, the films were found to be polycrystal. In the cases of the Ag films prepared at above 200°C and at RT with post-annealing above 300°C, the RHEED observations showed sharp streaky patterns, as shown in **Fig. 5**. The LEED spots also became sharper after annealing.

Crystalline quality of the ^{107}Ag films prepared with 30 eV ^{107}Ag ions at the substrate temperature of RT was examined by RBS-channeling. The RBS-channeling spectra showed the minimum channeling yield, which is called χ_{min}, to be 22% (see **Fig. 6**). The ion energy dependence of the χ_{min} is summarized in **Table 2** together with the increasing rate of the channeling yield $d\chi(z)/dz$, which reflects the defects density in the film. Additionally, we examined the χ_{min} of the Ag films grown at RT with post-annealing, and grown at 200–300°C using 30 eV ^{107}Ag^{+} ions. The depth dependence of normalized yield $\chi(z)$ is shown in **Fig. 7**, together with the RT growth using 50 and 100 eV ^{107}Ag^{+} ions for comparison. The χ_{min} of Ag films near surface was found to decrease from 22 to 12% by post-annealing at 400°C. Moreover, the χ_{min} of the Ag films grown at 200–300°C were 10% or less. The temperature dependence of the minimum channeling yield χ_{min} and increasing rate of the channeling yield $d\chi(z)/dz$ are summarized in **Table 3**.

The angular scans of the scattering yields from the Ag films/Si(111) surface grown at RT using 100 eV Ag^{+} ions were measured around the ⟨111⟩ direction to examine the relationship between Ag⟨111⟩ axis and Si⟨111⟩ axis (see **Fig. 8**). It is clearly seen that the direction of the Ag⟨111⟩ axis is different from that of the Si⟨111⟩ axis. In the case

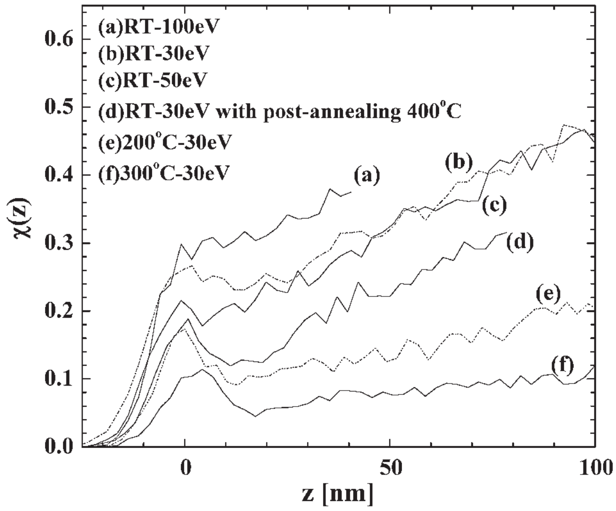

Fig. 7 Normalized yields *vs.* depth for ^{107}Ag/Si(111) prepared with 100 eV ^{107}Ag^{+} ions at the substrate temperature of RT (a), 30 eV at RT (b), 50 eV at RT (c), 30 eV at RT with post-annealing at 400°C (d), 30 eV at 200°C (e), and 30 eV at 300°C (f)

Table 3 Temperature dependence of the crystalline quality for ^{107}Ag films on Si(111) surface prepared with 30 eV Ag ions

Substrate temperature	RT	RT followed by 400°C annealing	200°C	300°C
Ag⟨111⟩ χ_{min} (%)	22	12	10	6
$d\chi(z)/dz$ (%/nm)	0.28	0.27	0.15	0.08

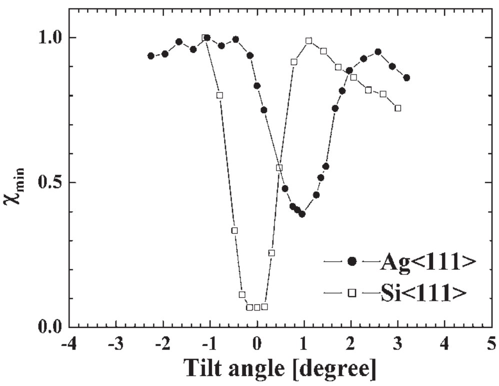

Fig. 8 RBS-channeling angular scans of the backscattering yield obtained around the Si⟨111⟩ and Ag⟨111⟩ axes
The ^{107}Ag/Si(111) was prepared with 100 eV ^{107}Ag^{+} ions at RT.

of vicinal Si(111) surface tilted by 0.5 and 5°, the tilt angle of the Ag film became to be 0.2 and 2° towards the [11$\bar{2}$] direction, respectively.

IV. Discussion

Surface crystalline quality and roughness of Ag films have been shown by RHEED patterns. In the case of the RT growth, RHEED spots became sharper in the energy range of 30–100 eV and hardly depended on the ion energy. This is consistent with the previous RHEED study.[4] However, in the cases of the films prepared at the temperature of 200–400°C and at RT with post-annealing at 300°C, RHEED observations showed the sharp streaky patterns and the LEED patterns showed sharper Ag(111)1×1 spots. It means that morphology of Ag film surface became flat.

The RBS-channeling experiments have been shown to examine the defects in the Ag films derived by dislocation, boundary, *etc*. Here, the minimum channeling yields $\chi_{\min}$ and the increasing rate of the channeling yield $d\chi(z)/dz$ are used to evaluate the crystalline quality. In Table 2, RBS-channeling analysis revealed the differences of the crystalline quality of the Ag films in the Ag^+ ion energy range of 30–100 eV. In the case of RT growth, the Ag films grown by 30–50 eV $^{107}Ag^+$ ions have fewer defects than those grown by 100 eV $^{107}Ag^+$ ions; defects density becomes minimum about 50 eV. The minimum value of $Ag\langle 111\rangle\chi_{\min}$ prepared by IBD at RT was 20%, which is much lower than those prepared by molecular beam epitaxy (MBE) at RT.[2,3,6] For the ion energy of 30–50 eV, the ion range is typically several angstroms and displacement energy is ~30 eV in the Ag crystal.[7] These facts indicate that not only the coming ions at the surface are easy to move, but surface- and subsurface-atoms are also energized by the incident ions. That is to say, the Ag^+ ions with 30–50 eV may cause local heating and anneal out some of the defects.

In Table 3, the $\chi_{\min}$ of the Ag films grown at 300°C by IBD is found to be 6%, which is a bit better than the $\chi_{\min}$ of 7–10% for the Ag films deposited at 350°C by MBE and is very close to theoretical value of $Ag\langle 111\rangle\chi_{\min}$.[8,9] Since it is considered that the film was twinned, the twin boundaries will not cause dechanneling in the channeling axis of the $\langle 111\rangle$ direction. The $\chi_{\min}$ of the Ag films grown at RT with post-annealing at 400°C is very close to those grown at 200°C, as is seen in Table 3. Nevertheless, there is a significant difference in the defects density in the films ($d\chi/dz$). In other words, the crystalline quality inside the films grown at 200°C is much better than those grown at RT with post-annealing at 400°C. It means the defects can be annealed out only at the surface and subsurface layers. Therefore, elevated temperature growth is found to be essential to make a highly crystallized Ag film.

Channeling angular scans around $Si\langle 111\rangle$ and $Ag\langle 111\rangle$ indicated that the orientation of silver epitaxial films is tilted slightly towards the $[11\bar{2}]$ direction (see Fig. 8). The tilted angle of the Ag films is always smaller than the vicinal angle of the Si(111) surface. Since these orientation can be also seen for the Ag films on vicinal Si(111) surface prepared by MBE,[6] it is considered that origin of these orientation tilt of the films came from the Ag/Si(111) interface mismatching.

V. Summary

The crystalline quality of the ^{107}Ag films on Si(111) surface grown by ion beam deposition (IBD) in the cases of RT growth, RT growth with post-annealing, and elevated temperature growth have been studied by means of combined techniques of RHEED, LEED, and RBS-channeling. In the case of RT growth, RHEED patterns hardly depended on Ag^+ ion energy, but RBS-channeling analysis revealed the difference of crystalline quality of the films; the Ag films grown by 30–50 eV Ag^+ ions have the fewest defects. It is also found that post-annealing can anneal out the defects only at the surface and subsurface layers of the Ag films; therefore, elevated temperature growth during IBD is found to be effective to make a highly crystallized Ag film.

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