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Crystallographic Analysis of Thin Film Surfaces Using Micro-Probe Reflexion High-Energy Electron Diffraction

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Micro-probe reflexion high-energy electron diffraction using an electron beam having a 20 nm beam diameter at a beam current of 8 nA, has been developed for performing crystallographic analyses of thin film and bulk crystal surfaces. High spatial resolution and high brightness have made it possible to perform analyses of thin films on substrates having fine structures without such sample preparation as thinning. A dark field imaging method using part of the diffraction spot intensity has also been developed. Using this method, it was found that atomic steps and dislocations on bulk and material-deposited Si surfaces can be observed. This shows the usefulness of the technique for studying crystal growth of thin films with mono-layer depth resolution.

1. Introduction

A key technology for developing new semiconductor devices is the production of good crystalline films on substrates having fine structures through the use of seeded lateral epitaxy, hetero epitaxy or similar techniques.

Microscopic observations of crystalline states of such films have been performed mostly by using trasmission electron microscopy (TEM). However, the thinning processes needed for such observation tend to cause position information in the samples to be lost.

To perform micro-area analyses of thin films without such sample preparation, we developed reflexion high-energy electron diffraction using micro-probe (micro-probe RHEED).[1),2)] Applying this technique to seeded lateral liquid phase epitaxy of Si on SiO_2, its usefulness was demonstrated.[3)] However, the spatial resolution obtained (>0.1 μm) was not sufficient to analyze samples having finer structures.

To compensate for this, we have recently developed a new micro-probe RHEED with high spatial resolution and high detection sensitivity to surface crystallogrophic orientation change.[4)]

This report describes the newly developed micro-probe RHEED and typical application results.

2. Experimental Method

An outline of the micro-probe RHEED is shown in Fig.1. An electron beam with 20 keV kinetic energy is emitted from the field emission tip and focused on sample position A and B. Crystallographic analysis is only performed at position A. Element analysis by Auger electron spectroscopy is performed simultaneously at position B. Beam diameters of 20 nm and 25 nm are

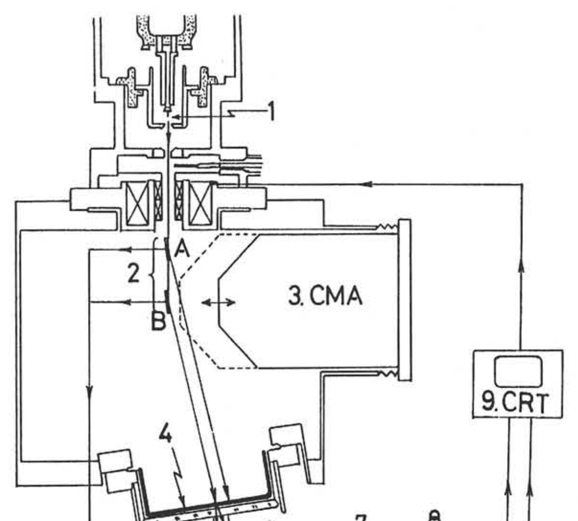

Fig.1: 1: FE tip, 2: samples, 3: cylindrical mirror analyzer, 4: fluorescent screen, 5: optical lens, 6: aperture, 7: optical fiber, 8: photo-multiplier, and 9: cathode ray tube.

obtained at positions A and B, respectively, at a beam current of 8 nA, and beam angular divergence of about 2 mrad. Analytical points are selected using scanning electron microscope (SEM) images produced by the absorption current. Diffraction patterns from these points are observed on a fluorescent screen. A particular diffraction spot is selected using an optical lens and an aperture. A photo-signal of the spot is guided to a photo-multiplier through an optical fiber. A dark field image is then obtained from the SEM image produced by the signal. Areas having specific crystalline states can be observed in this dark field image.

Moreover, it is possible to use part of a particular diffraction spot intensity to obtain a dark field image by using the optical lens and the aperture. This dark field imaging method has high detection sensitivity to crystallographic orientation changes near surfaces. It can show image contrasts caused by an orientation change of ~0.1 mrad which is produced by dislocations or atomic steps on the surfaces.

The vacuum in the sample chamber is usually kept below $5X10^{-10}$ Torr to keep sample surfaces clean.

3. Application Results

3.1 Silicon on Insulator

Observation results for seeded lateral solid phase epitaxy (SPE) of amorphous Si film on SiO_2 substrates are shown in Fig.2. The mechanism for seeded lateral SPE has already studied in detail by Yamamoto et al. using TEM.[5]

The sample used was made as follows. Si film about 300 nm thick was evaporated under ultra-high vacuum conditions on Si(001) 4°off-wafer with SiO_2 stripes about 50 nm thick at a substrate temperature of 500°C. Then, the Si film was amorphized by Si ion implantation. The wafer was then annealed at 600°C for 7 hours in a furnace.

The bright contrast area in the dark field image obtained using a 008 spot shown in Fig.2(a) represents the crystallized area. RHEED patterns from analytical points in the Si/SiO_2 and Si/Si bright contrast areas, indicate that the seeded lateral SPE occurs in the [100] direction for about 2 microns. However, the brightness in the

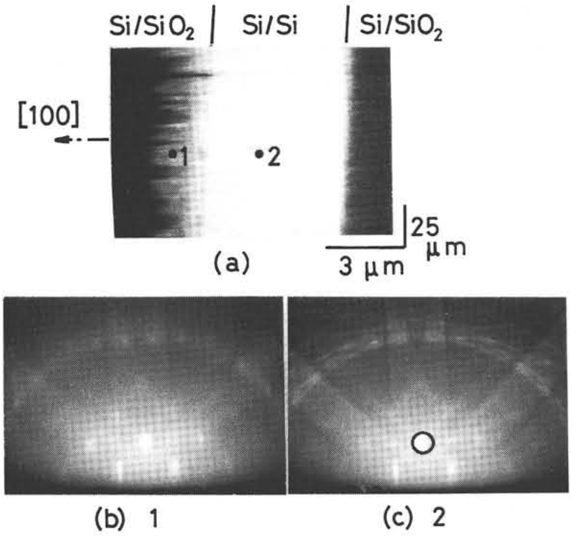

Fig.2: (a) Dark field image of an SPE sample using the 008 spot enclosed by the circle; (b) and (c) RHEED patterns from analytical points 1 and 2.

Si/SiO_2 area is weaker than that in the Si/Si area. This shows that crystalline quality of the film in the Si/SiO_2 area is poorer than that in the Si/Si area. This can also be confirmed by the sharpness of the Kikuchi patterns observed in Figs.2(b) and (c).

3.2 Silicon Implanted by Focused Ion Beam

Focused ion beam (FIB) implantation into semiconductors is a promising technology for maskless ion implantation. Investigation of the crystalline quality of implanted micro-areas is important in determining electrical activation

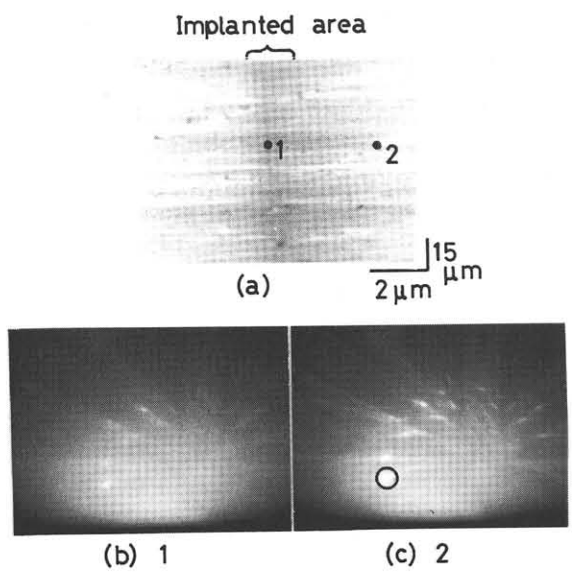

Fig.3: (a) Dark field image of an FIB sample using the spot enclosed by the circle; (b) and (c) RHEED patterns from analytical points 1 and 2.

when annealing is performed.[6)]

Observation results of a Si(001) 4°off-wafer implanted by $^{11}B^{+}$ focused ion beam[7)] are shown in Fig.3. Raster-scanned 16 keV ion implantation was carried out with a beam diameter of about 2 μm at a $2X10^{15}$ ions/cm^2 dose rate and a beam scan speed of $3X10^{-3}$ cm/sec. The dark contrast area in (a) shows the implanted area. RHEED patterns from the implanted and non-implanted areas shown in (b) and (c), respectively, indicate that the implanted area was partially amorphized with a specific spatial distribution.

3.3 Mono-Layer Thick Films on Si Surface

Observation results for a clean Si(111) surface used as a substrate for mono-layer range film are shown in Fig.4. The clean surface was made by several heatings at 1200°C for 3 sec under ultra-high vacuum. The dark field image in (b) was obtained using only the higher angle part of the 444 spot intensity, as shown in the figure. In that dark field image, dark/bright pair contrast (indicated by the arrow) as well as specific steps are observed. The dark/bright pair contrast is typical of that produced by a screw dislocation at the surface. This was previously observed by Osakabe et al. using ultra-high vacuum electron microscopy.[8)] One of these steps terminates in the dislocation, which indicates that these steps are of atomic hight (~0.3 nm) as illustrated in (c).

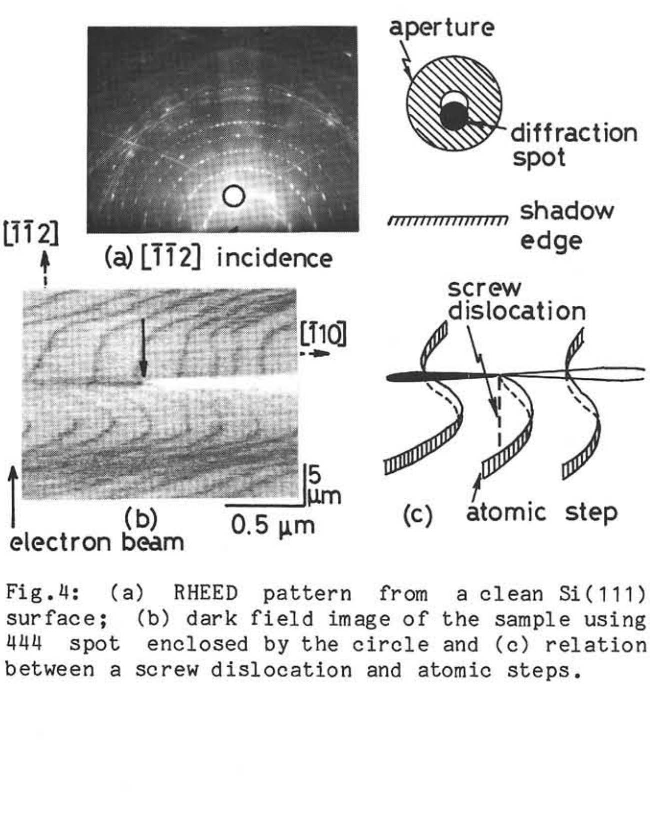

Fig.4: (a) RHEED pattern from a clean Si(111) surface; (b) dark field image of the sample using 444 spot enclosed by the circle and (c) relation between a screw dislocation and atomic steps.

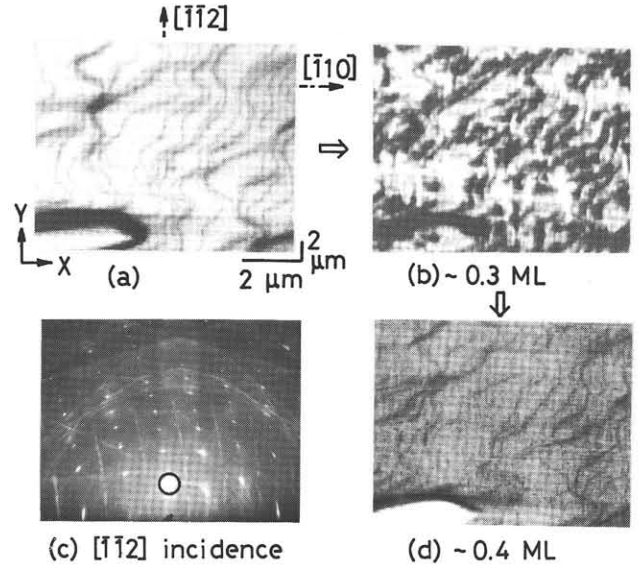

Fig.5: (a) Dark field image of a clean Si(111) surface; (b) dark field image of Au deposited surface at 800°C; (c) RHEED pattern from the dark contrast areas in (b), and (d) dark field image of the Au deposited surface.

Observation results for a Si surface on which Au was deposited at a substrate temperature of 800°C are shown in Fig.5. Dark field images of the Si and Au-deposited surfaces using a 444 spot are shown in (a), (b) and (d). These images were taken using a beam scanning method with magnification in the Y-direction about six times larger than that in the X-direction to compensate for image foreshortening.[1)] Many atomic steps are observed on the Si surface. When Au was deposited at about 0.3 mono-layer (ML), dark contrast areas with a Si(111) 5X1 Au structure having 5 times the period in the $[\bar{1}10]$ direction against the substrate (which was disclosed by the RHEED pattern in (c)) grew along the atomic steps. Continuing the deposition (~0.4 ML) caused almost the whole area to be covered with a 5X1 Au structure having the above direction.

Since a Si(111) surface generally has three-fold symmetry, three kinds of 5X1 Au structure are expected to appear equally on the surface. However, this was not the case, which indicates that production of the 5X1 Au structure was strongly affected by atomic steps running nearly in the $[\bar{1}\bar{1}2]$ direction. This phenomenon may be considered to be mono-atomic layer level "graphoepitaxy[9)]".

Observation results of a surface deposited by Al at a substrate temperature of about 700°C are

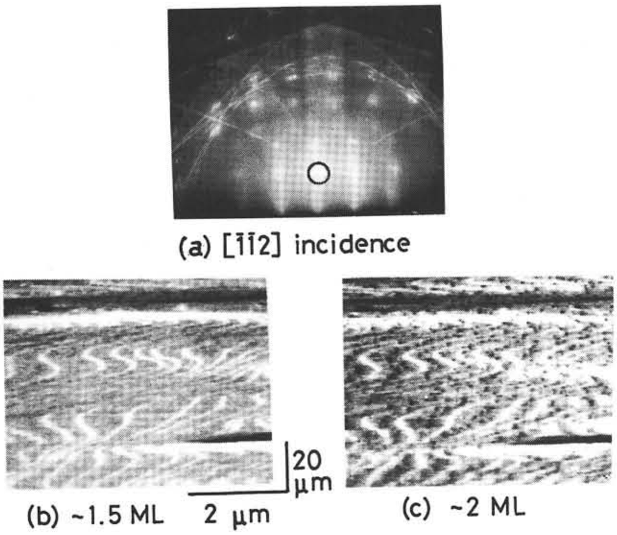

Fig.6: (a) RHEED pattern from an Al-deposited Si surface at 700°C; (b) and (c) dark field images of the surface using the 444 spot enclosed by the circle.

shown in Fig.6. When Al deposition of less than 1 ML was performed, a Si(111)$\sqrt{3}\times\sqrt{3}$-R30° Al structure was firstly produced at the surface. Continuing deposition (~1.5 ML) caused the Al film to grow epitaxially with a {(111)Al//(111)Si, [$\bar{1}\bar{1}2$]Al//[$\bar{1}\bar{1}2$]Si} orientation with the lattice constant of Si (0.54 nm), (not Al (0.4 nm)), as shown by the RHEED pattern in (a). In the dark field image of the surface in (b), bright contrast areas along the atomic steps are observed. Such contrast shows that crystalline quality of the Al film in this area is higher than that of other areas. With further deposition (~2 ML), small Al particles (the many dark dots in (c)) having random orienations were produced on the surface.

4. Conclusions

(1) The micro-probe RHEED has certain weak points in that the electron beam is easily interupted by unevenesses of surfaces and spatial resolution becomes poor in the incident direction due to grazing incidence. However, applications to SPE and FIB samples show that the technique is very useful for crystallographic analysis of micro-fabricated areas without special sample preparation if proper incident directions are selected.

(2) It was also found that a Si film on a Si substrate grows by connecting with atomic steps on the surface. Applications to Au, Al and Si films indicate that this technique will be very useful for in situ microscopic observations, with mono-layer depth resolution, of crystal growth in films for molecular beam epitaxial devices.

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