Surface roughening by electron beam heating

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The effect of electron beam heating during the preparation of clean silicon surfaces suitable for epitaxial studies in ultrahigh vacuum conditions was investigated using surface chemical characterization techniques and transmission electron microscopy. The electron beam irradiation produced a disordered surface on the incident side of the sample and well-ordered monoatomic steps on the other surface, even at electron energies as low as 3 keV. These results have significant implications for epitaxial thin film growth. © *1997 American Institute of Physics.* [S0003-6951(97)00242-8]

Epitaxial thin films are prepared by a wide variety of techniques. Various growth processes, for example molecular beam epitaxy, require deposition on atomically clean surfaces under ultrahigh vacuum (UHV) conditions. The presence of contaminants, native oxides, or a high concentration of surface defects on the substrate are known to influence the growth mode[1,2] and reduce the quality of the thin films.

One of the most common substrates for epitaxial growth is silicon owing to its extensive use throughout the semiconductor industry. To clean the surface, several techniques are used based on wet chemical etching, ion sputtering, and thermal annealing. Direct thermal radiation,[3,4] passing current through the sample,[5,6] laser annealing,[7,8] galliation,[9] electron-cyclotron-resonance (ECR) assisted hydrogen plasma,[10,11] and direct electron beam annealing[12–15] are only a few of the many thermal techniques currently available. Direct electron beam annealing has an advantage over direct thermal radiation and conventional electrical heating of providing rapid temperature control and lower levels of outgassing. Also, this method does not have the posttreatment contamination problems associated with galliation or ECR plasma techniques.

The cleanliness of the substrate surface after cleaning treatments can be monitored by surface chemical characterization techniques, such as x-ray photoelectron spectroscopy (XPS) and Auger electron spectroscopy (AES), or by observing the strength and sharpness of surface superlattice diffraction spots, such as those arising from the Si(111)-7×7 and Si(001)-2×1 reconstructions.[16]

In the present study, we report atomic scale surface roughening of samples during the preparation of clean silicon surfaces suitable for epitaxial studies in UHV conditions using electron beam heating.

Samples were prepared *ex situ* by traditional transmission electron microscopy (TEM) sample preparation techniques. Three mm disks were cut from *p*-type doped Si(001) wafers, mechanically thinned, dimpled, and polished to roughly 30 μm at the center. After chemical etching ($HF:HNO_3$ 1:9) the samples to perforation, they were introduced into SPEAR, a surface preparation and analysis UHV system. Details of SPEAR and the attached high resolution UHV H-9000 Hitachi transmission electron microscope are available elsewhere.[17,18] Once inside the system, samples were prepared using cycles of low energy, 1 keV, oxygen and argon ion milling followed by direct electron beam annealing to provide clean surfaces on both sides. The annealing was performed at 3 keV and 0.1 mA beam current using a EMG-14 Kimball Physics electron gun. Surface characteristics were monitored with XPS, AES, and TEM. Transmission off-zone electron diffraction (TED) and dark field (DF) were done at room temperature using a beam voltage of 200 kV to limit the microscope electron beam damage effects to the surface during investigations.

A typical clean Si(001) sample displayed a strong 2×1 reconstruction in the TED pattern, low concentration of defects, and well-ordered monoatomic surface steps in DF images. For this experiment we deliberately selected samples with a relatively high density of bulk defects, higher than we normally use, so that the same area could be identified. Figure 1(a) is a DF image of the Si(001) surface taken using a (220) type reflection and showing well-ordered steps. It was observed that the fringes from the surface steps did not overlap. The existence of only noncrossing steps can be attributed to the presence of well-ordered steps on only one side of the specimen, although XPS and AES show both sides as having contamination levels at or near the detection limits of the instruments. Plan-view TEM images show both sides of the sample imaged at the same time, and one cannot distinguish on which surface a particular feature is.

Since the electron inelastic mean free path in silicon for 3 keV electron energy is under 50 Å[19,20] and our TEM samples have a thickness typically greater than 200 Å we inferred that the electron beam irradiation during the thermal treatment is inducing a disordering of the incident sample surface. We investigated this fact by alternate disordering of the specimen surfaces and subsequent observations in the microscope. The reported results are from several such experiments. To simplify the discussion, we will refer to the side of the sample which the electron beam is incident upon during annealing as side A, and the exit as side B.

Surface B was disordered using 600 eV Ar ions for 5 s, 50° from surface normal. This ion energy was the minimum attainable with our ion gun and was found to be sufficient to disorder the surface steps. Figure 1(b) shows the same area as Fig. 1(a), after ion sputtering, but the steps are no longer present. Common features in the images, i.e., bulk defects, were used to identify the same area. The initial state of the sample was recovered after reannealing and Fig. 2(a) is similar to Fig. 1(a) showing only a different area. Using the same conditions for disordering, surface A was Ar ion milled. Fig-

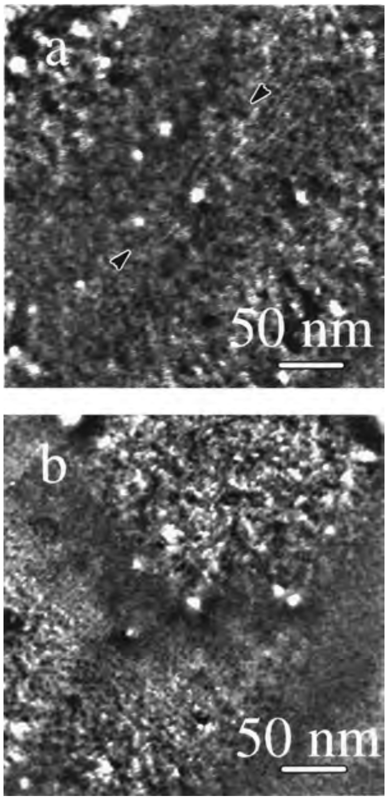

FIG. 1. Dark field image of the Si(001) surface using the Si(220) reflection, (a) prior to ion beam disordering of surface B, nonexposed to direct electron beam during annealing, showing well-ordered steps, (b) after the ion beam roughening, the surface steps are no longer present. Identical bulk defects can be observed in (a) and (b). A step has been arrowed for reference.

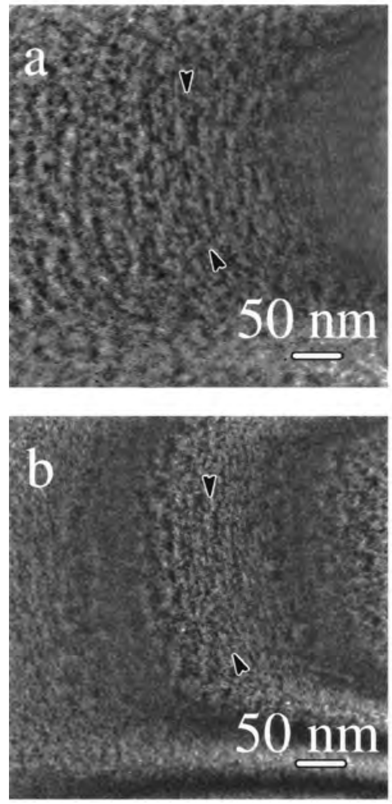

FIG. 2. Dark field image of the Si(001) surface showing the presence of well-ordered steps, (a) prior to and (b) after the ion beam disordering of the sample surface A (incident to electron beam during annealing). A step has been arrowed for reference.

ure 2(b) shows the same region as Fig. 2(a), but in this case the steps are still present. The mottled contrast also present in the image suggests an additional slight disordering. Therefore, it can be assumed that the electron beam annealing produced a disordered surface with nonuniformly distributed steps on the incident side of the sample to the electron beam, and well-ordered monoatomic steps on the exit surface. Reversal of the current direction during resistive heating of the reconstructed Si(001)-2$\times$1 surface has been shown to cause the rearrangement of surface atomic steps.[21,22] This suggests that random surface currents lead to the atomic scale disordering during annealing.

Similar observations were made during a recent study[23] by the authors, of Au thin film deposition on Si(001). Surface steps were no longer present in DF images after 8 Å of Au epitaxial deposition on surface B due to the disordering induced by the presence of gold.

Low energy electron diffraction (LEED) or reflection high-energy electron diffraction (RHEED) techniques could have also been used to detect surface roughness on an atomic scale[24] of silicon samples annealed with a different electron gun, but to the authors' best knowledge there is no such published study.

Isikawa *et al.*[25] have reported recently improved properties, suitable for nanofabrication, of the oxide layer used as resist mask on a GaAs substrate. Their study suggested that the use of an epitaxial surface with non-uniformly distributed atomic steps led to degraded characteristics of the film, this being an example of the important role played by the monoatomic steps. Fabrication of epitaxial thin films on silicon substrates[26] may be similarly influenced by the relative order or disorder of the surface steps.

The application of direct electron beam annealing to thermal treatments has been shown to disorder the incident surface of Si(001) substrates even at energies as low as 3 keV. In spite of successfully cleaning the incident area, the secondary effect, disordering, will potentially reduce the quality of epitaxial thin films grown thereupon. The results have important significance for the use of electron beam annealing for thin films growth and processing.

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