X-ray Studies of Si$_1$–$_x$Ge$_x$ Single Crystals

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Abstract—Structural imperfections were studied in Si$_1$–$_x$Ge$_x$ (1–9 at. % Ge) solid-solution single crystals grown using the Czochralski method. The studies were performed using x-ray diffraction topography with laboratory and synchrotron radiation sources, x-ray diffractometry, and synchrotron radiation phase radiography. In all crystals studied, irrespective of the Ge concentration, impurity bands (growth bands) were observed. An increase in the Ge concentration in the range 7–9 at. % was shown to bring about the nucleation and motion of dislocations on a few slip systems and the formation of slip bands. Local block structures were observed in the places where slip bands intersected. The most likely reason for the formation of slip bands is the inhomogeneous distribution of Ge atoms over the ingot diameter and along the growth axis. Therefore, the structure of Si$_1$–$_x$Ge$_x$ solid-solution single crystals can be improved by making them more uniform in composition. © 2005 Pleiades Publishing, Inc.

1. INTRODUCTION

Currently, epitaxial Si$_1$–$_x$Ge$_x$ solid-solution layers grown on silicon substrates are used in electronics, but Si$_1$–$_x$Ge$_x$ single crystals also hold promise. To circumvent technological problems associated with the growth of Si$_1$–$_x$Ge$_x$ solid-solution films, thick relaxed Si$_1$–$_x$Ge$_x$ layers are used, on which, in turn, thin elastically strained silicon layers are grown. These additional operations in the fabrication of semiconductor structures increase their cost. For this reason, the idea of growing elastically strained Si layers on Si$_1$–$_x$Ge$_x$ single-crystal substrates is becoming more and more attractive. However, these substrates have to have a high Ge concentration (of up to 20 at. %) and a low density of structural defects.

Si$_1$–$_x$Ge$_x$ solid solutions are also used as an active element in photoelectric converters (solar cells) due to their sensitivity to radiation in the long-wavelength region of the visible spectrum. The quality of solar cells based on epitaxial layers of gallium arsenide and related III–V compounds is low due to structural imperfections caused by the lattice mismatch between the active layer and the silicon substrate. Replacing the Si substrates by Si$_1$–$_x$Ge$_x$ decreases this mismatch and makes it possible to optimize the fabrication technology of GaAs-based solar cells and enhance their reliability.

Large Si$_1$–$_x$Ge$_x$ single crystals are usually grown using the Czochralski method [1, 2]. In this case, the spatial distribution of germanium is inhomogeneous. Due to segregation of germanium in silicon, the germanium concentration in these crystals varies over a cross section and along the length of an ingot. By controlling the growth conditions, ingots with the desired lattice parameter gradient along the growth axis can be obtained [3]. Si$_1$–$_x$Ge$_x$ crystals with lattice parameter profiles hold promise for use in synchrotron radiation optics. For example, a crystal monochromator in which the interplanar distance varies along the surface enables one to decrease the beam divergence and to increase the reflectance of the monochromator, with the reflected beam remaining monochromatic [3].

The growth of Si$_1$–$_x$Ge$_x$ single crystals with a uniform spatial distribution of germanium poses severe problems, which have not yet been overcome. Impurity bands (growth bands) are always present in Czochralski-grown Si$_1$–$_x$Ge$_x$ crystals [4]. These bands arise due to microscopic fluctuations in the growth rate, which, in turn, are caused by nonsteady-state convective flows in a melt [5].

A change in the Ge distribution in silicon leads to a change in the lattice parameter and favors the formation of structural defects. Since defects have an adverse effect on the parameters of devices and the characteristics of x-ray monochromators based on Si$_1$–$_x$Ge$_x$, it is of importance to develop a technology for producing uniform crystals with desired properties. With this aim, the relation between the growth conditions and the structural perfection of crystals should be investigated.
To date, it has been established that (i) the Ge concentration and the growth front curvature have an effect on the distribution of growth bands and their structure [6], (ii) the amount of dislocations in a crystal and their distribution in horizontal and vertical cross sections of an ingot depend on the Ge concentration and the orientation of the growth axis [1, 3], and (iii) the mobility of dislocations is determined by microscopic inhomogeneities of a solid solution [7, 8]. Micro-inhomogeneities in $Si_{1-x}Ge_x$ and $Ge_{1-x}Si_x$ solid solutions also have an effect on the stress–strain curves of these materials [9].

The objective of this work is to comprehensively study the formation of defects in $Si_{1-x}Ge_x$ crystals depending on the Ge content in the range 1–9 at. % for crystals with various orientations of the growth axis. We used different methods based on x-ray diffraction: x-ray topography, diffracontometry, and synchrotron radiation phase radiography.

2. SAMPLES AND EXPERIMENTAL TECHNIQUES

2.1. $Si_{1-x}Ge_x$ Single Crystals

$Si_{1-x}Ge_x$ crystals ($0.01 \leq x \leq 0.09$) were grown using the Czochralski method at the Crystal Growth Institute (Institut für Kristallzüchtung, Berlin, Germany). Ingots up to 42 mm in diameter had a weak Ge concentration gradient along the growth axis. The growth direction was parallel to $\langle 110 \rangle$, $\langle 111 \rangle$, $\langle 001 \rangle$, or $\langle 122 \rangle$ (the last growth direction arose after twinning of a crystal grown along $\langle 001 \rangle$). Crystals (with n- or p-type conductivity) were lightly doped with phosphorus and boron to a concentration of approximately $10^{15} \text{ cm}^{-3}$ and contained oxygen at a concentration of $6 \times 10^{17} \text{ cm}^{-3}$. We studied samples in the form of ~0.4-mm-thick plates cut perpendicular to the growth axis, with both faces polished using chemical and mechanical methods.

2.2. X-ray Images of Crystals

X-ray images of the plates under study provided most of the information on crystal imperfections. The images were obtained in three different ways: (i) x-ray diffraction topography with a laboratory x-ray source, (ii) Bragg diffraction with a synchrotron source, and (iii) Fresnel diffraction with a synchrotron source. Let us describe each of the above means of obtaining images.

(i) Laboratory x-ray topographs were obtained using the Lang projection method in the Bragg or Laue geometry with commercial equipment (Cu and Mo $K_\alpha$ radiation). The beam divergence was dependent on the radiation wavelength and was of the order of a few minutes of arc. The resolution of the method was a few micrometers. The images were recorded using photographic plates with nuclear emulsion whose resolution corresponded to the highest resolution of the method.

(ii) Synchrotron radiation topographs were obtained using polychromatic radiation with an energy of 10 to 60 keV. The spatial beam divergence was 2 and 5 $\mu$rad in the vertical and horizontal planes, respectively. A beam of radiation with small divergence and with a large cross-sectional area on a sample can only be provided by a sufficiently remote source. A source of synchrotron radiation satisfies these requirements and provides high-intensity beams. When radiation with a continuous spectrum is incident on a single crystal, each set of crystallographic planes “selects” the wavelengths for which the angle between the diffracting planes and the beam satisfies the Bragg condition. As a result, there appear many diffracted beams behind the crystal, with each Laue spot being a high-resolution topograph [10]. The contrast of the images in polychromatic radiation is due to variations in the orientation and extinction. In the former case, the intensity varies from point to point depending on the lattice misorientations. The extinction contrast is due to local variations in the crystal imperfection: in the vicinity of a defect, x rays are scattered in much the same way as in a mosaic crystal and the integrated reflection intensity from this region is higher than that from a more perfect region of the crystal [10].

The detecting device consisted of a 200-μm-thick CdWO₄ crystal scintillator, a lens to magnify the image, and a CCD camera. The field of vision was 8 x 8 mm in size, and the pixel size was 15 μm. The sample–scintillator distance was 8 cm. Experimentally, there was room for only one Laue topograph on the CCD array.

(iii) The images were also obtained using synchrotron radiation phase radiography. The description of the technique for producing x-ray phase images can be found in [11–13]. We employed this technique to detect inhomogeneities (e.g., Ge inclusions) in which the material density differs from that in the matrix. The images were recorded using a method similar to the white-beam method, but the resolution of the CCD array was significantly higher: the pixel size was 0.14 μm and the sensitivity was 16 bit. The highest resolution of the detecting device was 2 μm. The sample–scintillator distance was 8–10 cm.

Synchrotron radiation experiments were carried out in Pohang (Republic of Korea) at Pohang Light Source, station 7B2 (a third-generation synchrotron radiation source).

2.3. X-ray Diffractometry: Measurement of the Lattice Parameter

The lattice parameter was measured using a triple-crystal spectrometer [14]. The beam diameter in the scattering plane was 0.5 mm. The sample could be displaced within ±20 mm in a horizontal direction. The horizontal displacement and rotation of the sample about the axis perpendicular to the sample surface made it possible to measure the Bragg angle at various points and determine the variation in the lattice param-