COLUMNAR STRUCTURE GROWTH BY SILICON MOLECULAR BEAM EPITAXY

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ABSTRACT

Si columnar structures were fabricated using Si MBE on Si substrates with column sizes in the order of ~ 100 Å. The objective is to explore a viable approach to fabricate quantum wire structures. The growth of the structures, which was due to the growth instability, was an excellent example of a self-limiting process. The dependence of column morphology on the critical parameters, e.g., Si molecular beam incident angle, substrate temperature, substrate rotation, speed, etc., were demonstrated. Comparison between the experimental and the computer simulation results demonstrated the importance of the latent heat related atom migration as compared to the normal surface diffusion at low substrate temperatures and several Å/s beam fluxes. A substrate temperature window (≈125°C) was observed which allowed the fabrication of crystalline micro-columns on Si (100) substrates. RHEED studies indicated that the crystalline micro- columns were heavily twined. The twinning phenomenon was also observed in the computer simulation results and interpreted as a result of the reduction in twin formation energy due to the extremely small dimension of the columns. Thermal stability of the columnar structures is discussed. Finally, photoluminescence studies and some potential applications are also discussed.

INTRODUCTION

It has long been recognized that columnar microstructures exist in many types of thin films deposited under various conditions. [1-3] Many macroscopic properties such as the effectiveness of W-Ti-(N) as barrier layers for Al in Si integrated circuit technology[4], and magnetic anisotropy in iron thin films[5], have been associated with such microstructures. We report our recent study on several aspects of the Si columnar structures grown by molecular beam epitaxy (MBE) on Si substrates. The motivation of our study is to explore any potential optoelectronic application of these structures through effects such as quantum confinement, and to understand certain aspect of this film growth mode which will hopefully improve our understanding of other growth modes of Si films on Si substrates.

EXPERIMENTS

We use a Riber Eva-32 Si MBE apparatus with in-situ RHEED capability to deposit the films on Si (100) substrates. The detailed microstructures are imaged using a tilted cross-sectional transmission electron microscopy (TTEM) technique in an under-focused or over-focused condition as illustrated in fig.1. Such a TEM geometry allows us to obtain the dimensions of individual columns, which is difficult to do using the conventional cross-sectional geometry. Our experimental results are compared with our molecular dynamics simulation results. The samples were subjected to various annealing processes including low temperature oxidation and high temperature vacuum furnace annealing to study the thermal stability of the columnar structures. An Ar-ion laser at 514 nm wavelength line and a Spex spectrometer with a cooled Ge detector are used for photoluminescence (PL) measurements.

The molecular dynamics (MD) simulation was carried out by considering the impingement of a beam of Si atoms onto a Si substrate at a 75° angle of incidence. Single atoms were inserted at regular intervals into the computational cell at positions far from the substrate where they had no interactions with atoms in the deposit. The velocities of the inserted atoms were directed at normal incidence to the substrate, and the magnitudes of the velocities correspond to a beam temperature of 2800°K. Atomic trajectories were calculated using Newtonian mechanics and forces based on an empirical potential for Si[6]. The temperature of the substrate was regulated by frequent renormalization of the velocities of a group of atoms in the substrate. The atomic beam intensities that are about nine orders of magnitude greater than those used in practice, but they were maintained below the point where the temperature of the deposit would be increased by more than ≈50°K. Details of the simulations have been given elsewhere[7].

RESULTS

Fig.2A and 2B are conventional cross-sectional TEM micrographs. It is quite obvious that the columns are inclined toward the incident direction of the molecular beam when there is no substrate rotation, and are vertical when there is substrate rotation. The dimensions of each individual column can not be determined from these micrographs because of the overlapping of many columns.

The overlapping problem is alleviated by using the TTEM geometry. Fig.3 is an as-grown Si-on-Si columnar structure. The sample was grown at 150 C (thermocouple measurement). By measuring the individual column sizes, we are able to find an average column size of 130± 30 Å. This should be compared with the column size of another sample grown at room temperature (25°C), where the column sizes are in the order of 40 Å. This difference in column sizes with substrate temperature agrees fairly well with the MD simulation result.

The temperature dependence of the growth modes as characterized using RHEED is shown in fig.4. The temperature is determined by a thermocouple situated behind the center of the substrate. Since this temperature reading could be different from the true surface temperature by as much as 200°C (by comparing with a calibrated optical pyrometer reading at > 500°C), the temperature quoted here should be taken as a relative temperature scale only. We see from fig.4 that the growth mode changes from the two-dimensional (streaky RHEED as in fig.4A) mode to the three-dimensional (spotty RHEED as in fig.4B) mode when the growth temperature, and thus the surface mobility of the Si adatoms are lowered. At low enough temperatures, the films grown become microcrystalline or amorphous (ring RHEED patterns as in fig.4D). This result indicates that there is a temperature "window" for growing crystalline Si columns on Si substrates. As we try to grow samples in this temperature "window", however, we find that the columns first grow in the form of twinned crystals (fig.4C), and then gradually change into polycrystalline form. This exact phenomenon is observed from our MD simulation results (fig.5) for a substrate temperature of 370°C. We believe that the phenomenon can be explained by the small dimension (≈ 100 Å) of the columns which have lower energy barriers for twinned crystal formation, and the intersection of two developing twinning planes (as indicated by the dashed pentagon in fig.5) which causes the subsequent amorphous growth.

Fig.6A and 6B are TTEM pictures of an as-grown and a 300°C annealed (in wet oxygen ambient) samples, respectively. It seems that the columns are fused into ridges when annealed even at such low temperatures. Enhenced surface diffusion is believed to be responsible for the process.

The PL results are shown in fig.7. Despite the small dimensions of the columns, no significant shift of the peak position of the luminescence is observed (as might be expected in the light of the recent luminescence results of porous Si[8]). The oxidized sample does show a shoulder on the higher energy side of the bulk Si peaks. The origin of this shoulder in the PL spectrum, however, is not clear at this point.

Although PL did not show a clear indication of quantum confinement effects from these "quantum wires", we believe this could still be an alternative approach to the quantum confined structures. Controlling the sizes of the columns and passivating defect states are going to be the key to achieving this goal.

SUMMARY

We, have fabricated Si columnar structures on Si (100) substrates under various conditions. The results show that the columns incline toward the molecular beam incident direction when the substrate is not rotated, and the columns become vertical to the substrate surface when the substrate is rotated. A temperature window at $\approx$125°C is found which allows the growth of crystalline columns. In this growth mode, the growth proceeds as twinned crystals, and eventually turns into micropolycrystalline. The column dimensions are determined to range from 40Å to $\approx$ 100 Å. Excellent agreements are observed between the experimental and the MD simulation results. Photoluminescence study did not show a clear indication of quantum confinement effects.

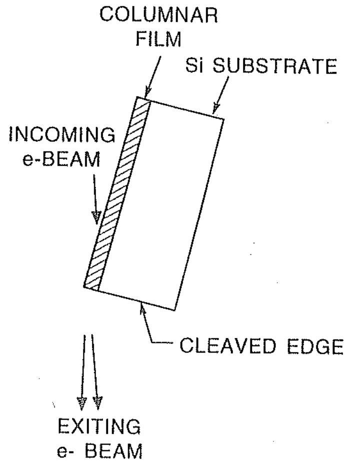

Figure 1. Illustrative geometry of tilted cross-sectional TEM.

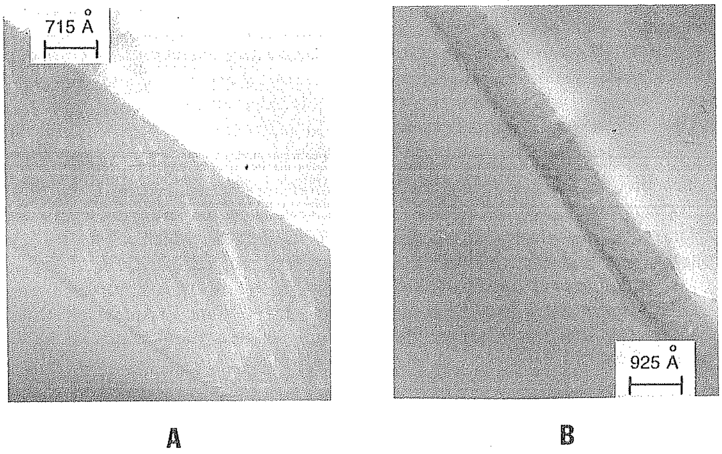

Figure 2. Conventional cross-sectional TEM micrographs of (A) a sample grown at glancing beam incidence without substrate rotation; and (B) a sample grown first without and then with substrate rotation.

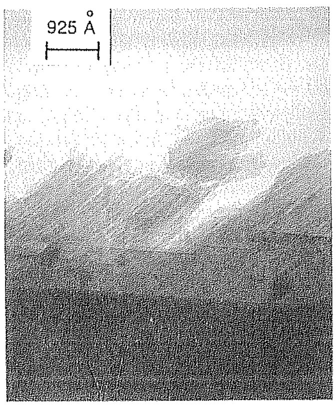

Figure 3. Tilted TEM micrograph of a sample grown at 150°C substrate temperature (measured by thermocouple).

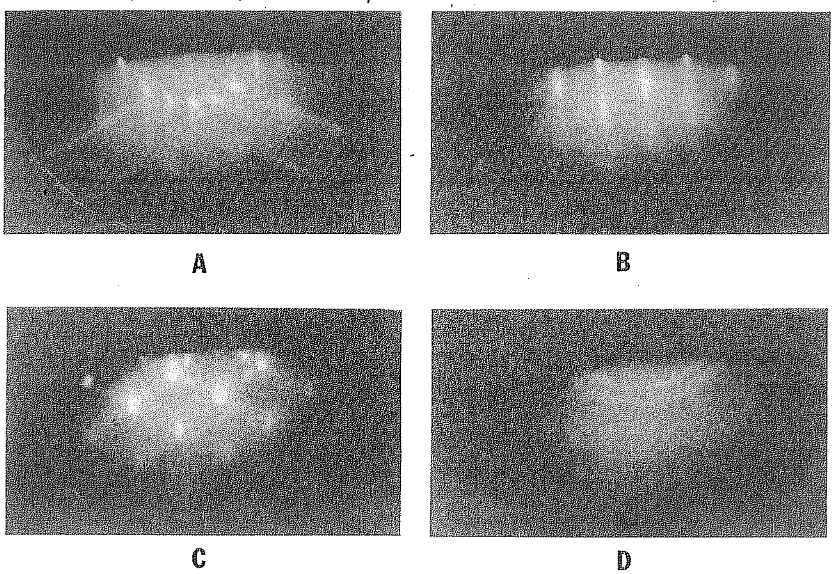

Figure 4. RHEED pictures taken at various stages of the columnar structure growth: (A) 550°C growth, (B) 125°C growth, (C) heavily twinned structure; and(D) 25°C growth, or after growing a thick layer at 125°C.

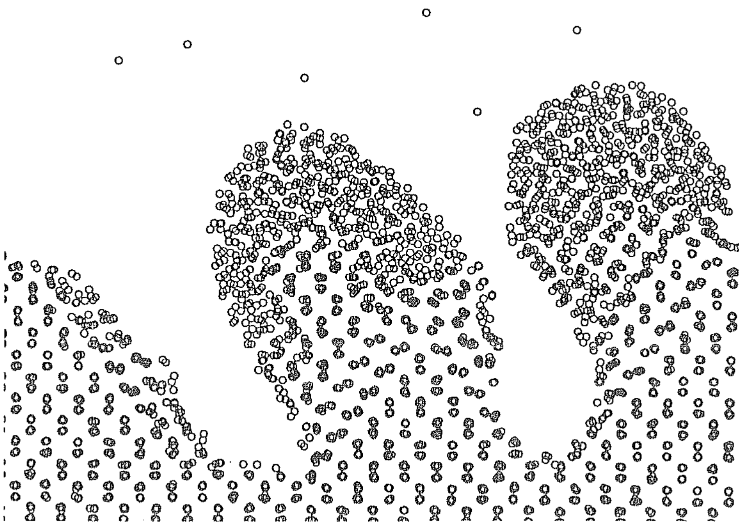

Figure 5. Molecular dynamics simulation result of Si columnar structure growth.

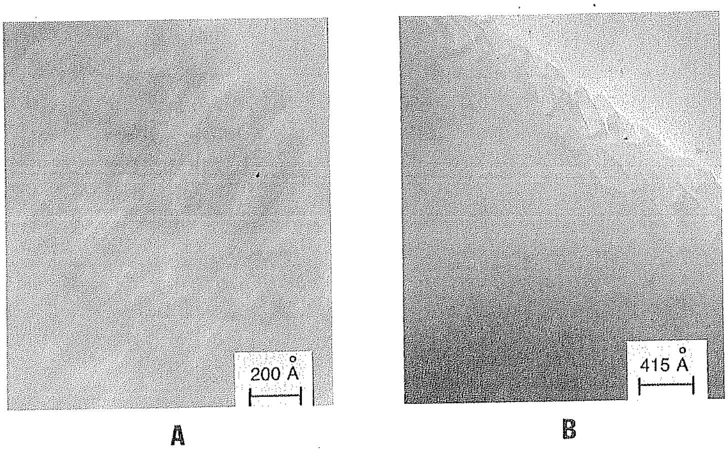

Figure 6. Tilted TEM image of (A) an as-grown columnar structure, and (B) the samp sample annealed at 300°C in wet oxygen.

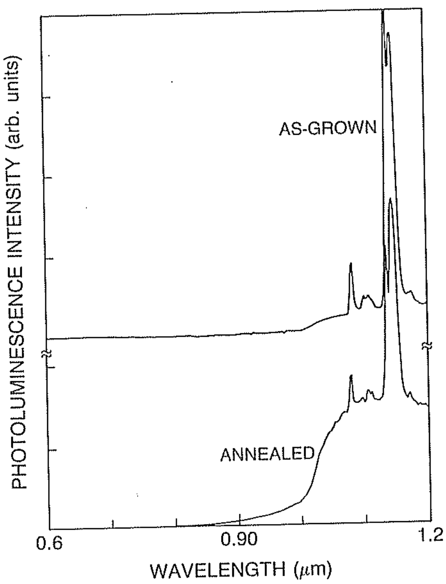

Figure 7. Photoluminescence result of the as-grown and the annealed samples.

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