論文の内容の要旨

論文題目
High rate and high yield epitaxial silicon film deposition from $SiHCl_3$ under mesoplasma condition
($SiHCl_3$を用いたメゾプラズマCVDによる高速・高収率エピタキシャルSi薄膜堆積)
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"Wafer-equivalent" crystal Si thin-film solar cell has been considered to be a promising approach to realize the low-cost PV power generation because the advantages of the stability and high conversion efficiency potential of crystalline silicon solar cell with the low material utilization of the thin-film solar cell can be combined. The primary requirements to realize this approach would be a development of an epitaxial technology for Si films with high deposition rate and high production efficiency of the gas precursors.

Among various technologies for Si epiaxy, the thermal chemical vapor deposition (TCVD) employing trichlorosilane ($SiHCl_3$: TCS) and hydrogen as reaction gases has been the major and mature technology for the fast rate epitaxial Si film deposition. However, due to its deposition process is basically controlled by the thermodynamic equilibrium condition on the substrate surface, the production yield of Si is practically limited as low as 30 %. Clearly, therefore, a new concept is required to surpass the efficiency.

We have identified a novel process with mesoplasma that provides unique plasma environment, which seemingly fits well to the requirements for the fast rate and high yield epitaxy. This plasma is also anticipated to provide a high atomic hydrogen flux that can suppress the formation of silicon-chlorides. In view of these circumstances, with an aim to develop a novel production route of "wafer-equivalent" crystal Si thin films solar cells, this study focuses on an enhancement of TCS reduction and a direct deposition of high quality epitaxial Si thin films with high deposition rate from TCS under mesoplasma condition.

The investigation of this study has been implemented through understanding the effects of the chemical species and also the effective use the atomic hydrogen that is believed to be the core aspect to reach the aims in this study. The results obtained in this study have demonstrated the superiority of the mesoplasma CVD technology for the simultaneous attainment of the high quality epitaxial Si films with high rate and high yield. In this study, epitaxial Si films having

deposition rate as high as about 700 nm/s and production yield of more than 50% have been successfully achieved with Hall mobility as high as approximately 250 $cm^2 V^{-1} s^{-1}$ with a hole carrier concentron on the order of 10^{15} cm^{-3}. This deposition rate is about one order higher than that of the typical TCVD technology, the production yield is about 2 times of that of TCVD and the Hall mobility is about 70% of that for the commercial p-type silicon wafer. This makes the mesoplasma CVD as a promissing technique for the low cost "wafer-equivalent" Si thin films solar cells production. Study of the deposition process has revealed the nanocluster-assisted characteristic with a high atomic hydrogen density is the primary advantage of mesoplasma CVD, which uniquely distinguishes this technique from other processes.

This thesis consists of 6 chapters, as is described below.

In chapter 1, advantages of the crystal Si thin-film solar cells and its potential to reduce the PV power-generation cost are explained. The development of a technique with high rate and high yield for the epitaxial Si film deposition required for the crystal Si thin-film solar cells application is motivated. The technologies used for the epitaxial Si film deposition are simply presented and the challenges for the high rate epitaxy are analyzed. The objectives of this study by using mesoplasma are given.

In chapter 2, the concept of mesoplasma is given through the comparison with low-pressure plasma and thermal plasma. The characteristics of the mesoplasma CVD and its advantages are presented. The basic deposition process of epitaxial Si films from SiH_4 in mesoplasma CVD and its results for the high rate and low temperature epitaxy are reviewed. Preliminary studies of the epitaxial Si film deposition from TCS are implemented using two power generators with rf frequencies of 5 MHz and 13.56 MHz. The effects of the initial substrate temperature, input power and plasma flow rate are investigated. The feasibility of the Si epitaxy from TCS is revealed and the achieved epitaxial Si deposition rate of about 250 nm/s is already more than 2 times higher than that by conventional TCVD.

In chapter 3, a cavity ring-down spectroscopy (CRDS) system is designed and installed for the in-situ characterization of the chemical aspect of the mesoplasma CVD. Measurement of the atomic hydrogen density is implemented by CRDS through a line-of-sight absorption measurement of H(n=2) atoms (via the n=2 ← n=4 Balmer-b transition) in the plasma plume. Through the investigation of the H(n=2) column density at different experimental conditions, the mechanism of the H(n=2) formation is analyzed to be due to the Ar^+ ion attachment reactions, i.e., the reaction of H_2 with Ar^+, followed by dissociative recombination of an electron to ArH^+. A high atomic hydrogen environment with H(n=2) column density as high as on the order of 10^{11} cm^{-2} is revealed in mesoplasma CVD condition.

Chapter 4 deals with the experimental deposition of the high quality epitaxial Si films with high rate and high yield. The effects of H_2/TCS ratios, TCS flow rates and input powers on the films structure, deposition efficiency, impurity concentration and electrical properties are investigated in this chapter. Investigation of H_2 is focused on the improvement of the production yield and also looking into the effect of H_2 on the Si film deposition from TCS in mesoplasma CVD. Increases in TCS flow rate and input power are attempted to best use the high temperature gases to increase the deposition efficiency (deposition rate and production yield). An epitaxial Si film with a super high rate of about 700 nm/s and high yield of more than 50 % is obtained simultaneously with a Hall mobility of as high as 250 $cm^2 V^{-1} s^{-1}$.

In chapter 5, the thermal boundary layer characteristics is employed to explain the film deposition process, by which a nanocluster-assisted process is demonstrated for the Si film deposition from TCS in mesoplasma CVD. The variations of the deposition efficiency and impurity concentration are discussed through the chemical species consideration. The importance of the atomic hydrogen in the high rate and high yield epitaxial Si film deposition in this process is revealed through the study of the tendencies of the H(n=2) column density with that of the deposition efficiency (rate). A nanocluster-assisted process with a high atomic hydrogen density is suggested to be the major characteristic of the mesoplasma CVD to realize the simultaneous achievement of the high quality epitaxial Si film with high deposition rate and high yield.

In the last chapter 6, the major experimental achievements and also the understandings of the mesoplasma CVD for the fast rate deposition of the epitaxial Si films from TCS with high yield and high quality are summarized. The suggestions for the future work and its potential applications by using mesoplasma CVD are also given.