Title

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ABSTRACT

Silicon thin films were vacuum deposited onto fused quartz at 600°C with a prior coating of ultra-thin aluminum layer (ca 500Å). Significant increase in crystallinity was observed from both X-ray diffraction and transmission electron microscopy studies. The problem of handling aluminum film at this temperature in a vacuum of 1×10^{-6} Torr is discussed, and distribution of aluminum in the silicon film is studied using Auger spectroscopy.

Recently, there have been studies aimed at producing high crystallinity silicon and germanium films at rather low temperatures.[1] Various metals were used in these studies to help recrystallize silicon and germanium. Here we report the use of an ultra-thin aluminum coating (ca 500Å) prior to depositing silicon thin film onto fused quartz held at 600°C. Silicon films thus produced showed much increased crystallinity compared to films directly deposited onto a quartz substrate. The process used is similar to the work of Filby and Nielson[2] using gold except that our work was done at a much lower temperature. Also, aluminum is of particular interest because it gives p-type doping in silicon suitable for solar cells. The problem of handling aluminum film on quartz at high temperature in a vacuum of 1×10^{-6} Torr is also discussed.

Both aluminum and silicon were vacuum deposited onto fused quartz using conventional electron beam heating. A two-source copper crucible was used which allowed successive evaporation of these two materials without breaking the vacuum. The deposition rate was monitored with an evaporation rate monitor from Allen Jones Electronics with precalibrated deposition rates for each of the materials evaporated. Fused quartz substrates were mounted onto a tantalum substrate holder which can be heated by electron bombardment. The substrate temperature was measured with a chromel-alumel thermocouple. The deposition rate was typically ca 40 Å/min for silicon and ca 100 Å/min for aluminum. 5-N purity aluminum and silicon were used for this work, and fused quartz substrate (1×1/2 in.) were cleaned with soap and degreased with isopropyl alcohol before use. Before deposition, the substrates were preheated to ca 800°C for 1 hr for outgassing. Film thickness was measured with a Varian Å-scope interferometer. Auger spectroscopic analysis of aluminum distribution in silicon film was performed using LEED-Auger spectrometer system from Physical Electronics which has a double-focusing cylindrical mirror analyzer and is equipped with an ion sputtering gun. Ion sputtering using argon was performed at an argon pressure of 5×10^{-5} Torr with sputtering rates of ca 30-40 Å/min for the silicon film.

Before we applied the aluminum coating method to silicon film deposition, we found that at the vacuum of 1×10^{-6} Torr which we used, aluminum film was unstable at 600°C either deposited directly onto the quartz substrate at this temperature or deposited at room temperature and heated to 600°C for 1 hr.[3] Both oxidation and reaction with quartz[4] could be responsible for this observation. To overcome this difficulty a Si-Al-Si sandwich coating was used at a composition of ca 100Å-500Å-100Å; the inner silicon layer was to prevent aluminum from reacting with quartz and the outer one from oxidation. This Si-Al-Si coating was deposited onto quartz at room temperature. Upon heat treatment of this sandwich film at 660°C signs of melting were visually observable and the film turned into pink color. X-ray diffraction showed highly preferred (111)-oriented structures for both Si and Al. For comparison, no diffraction pattern were observed for either aluminum or silicon deposited at room temperature, silicon films deposited at 600°C showed only powder diffraction pattern and aluminum film deposited at room temperature and heat treated at 500°C for 2 hrs showed highly preferred (111) orientation.

These comparisons thus clearly indicated melting of silicon with aluminum in the Si-Al-Si sandwich film described above.

Next, silicon was deposited on top of the Si-Al-Si coating at 600°C. After depositing ca 2000-5000Å thick silicon film the substrate was heated to 660°C for 1 hr to allow better eutectic melting between silicon and aluminum.[5] The silicon film thus produced showed highly preferred (111) orientation with the relative intensity of (220)/(111) being 10-20% compared with the value of 60% for the powder diffraction pattern. No diffraction pattern was observed for aluminum. Transmission electron microscopy showed spot diffraction pattern, as shown in Fig. 1. Also shown for comparison is that of the pure silicon film deposited at 600°C as described earlier. Large increase in crystallinity of the silicon film is clearly seen from Fig. 1. From the diffraction pattern observed, grain size of at least 5μ can be estimated.

Auger spectroscopic analysis of the silicon film indicated that aluminum concentration was higher near the surface than that in the bulk film, the latter being on the average less than 1%. Quantitative determination of the aluminum concentration on the surface was difficult because of the overlapping bands of silicon in the oxidized form. The higher aluminum content near the surface thus indicated that silicon atoms eutectically melted with aluminum upon arriving on the substrate, crystallized into large grains and precipitated from the melt. Some of the aluminum also dissolved in and precipitated with silicon although most aluminum stayed on the top of the melt. The thus "diluted" aluminum also accounted for the earlier mentioned fact that no aluminum diffraction pattern was observed for these films. Also consistant with this observation was the resistivity measurement of these films. Using the four-point probe technique, the silicon films deposited using the Si-Al-Si coating had measured resistivity of 5-8 Ω-cm which increased almost 60 times after the films had been etched in Keller's solution.

In summary, we have demonstrated that an ultra thin layer of aluminum coated on fused quartz helped significantly to increase the crystallinity of silicon films deposited at fairly low temperatures. Work is also in progress to find the temperature dependence of the crystallinity increase as well as the aluminum content in the silicon film. Other p-type and n-type metals will also be tested using this technique before it can be combined with that of chemical vapor deposition using, for example silane, for large scale production of silicon thin film solar cells.

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FIGURE CAPTION

Fig. 1. Electron diffraction micrographs of (a) silicon film deposited on quartz at 600°C; (b) silicon film deposited on quartz at 600°C with a prior coating of Si-Al-Si layers.

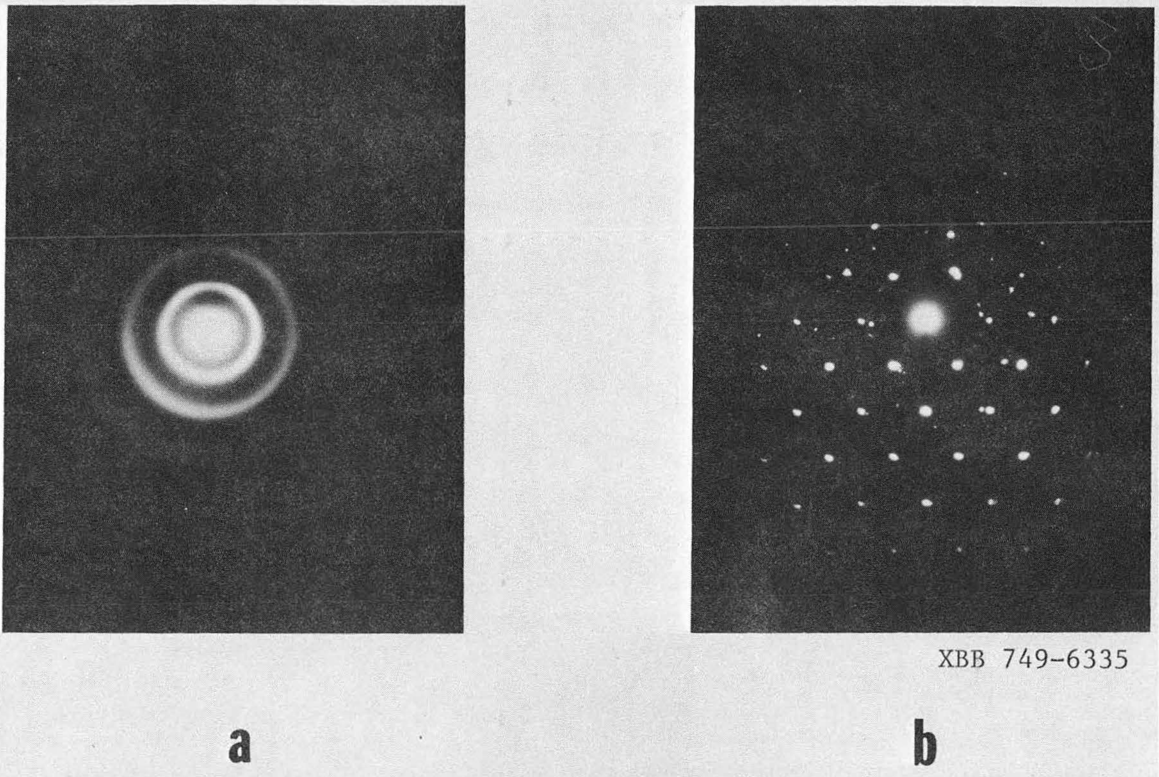

Fig. 1

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