Oblique Angle Deposition of Germanium Film on Silicon Substrate

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Abstract—The effect of flux angle, substrate temperature and deposition rate on obliquely deposited germanium (Ge) films has been investigated. By carrying out deposition with the vapor flux inclined at 87° to the substrate normal at substrate temperatures of 250° C or 300° C, it may be possible to obtain isolated Ge nanowires. The Ge nanowires are crystalline as shown by Raman Spectroscopy.

Index terms—Oblique angle deposition, Germanium nanowires

I. INTRODUCTION

Oblique angle deposition basically combines a typical deposition system with a tilted and rotating substrate. [1] The oblique angle flux incidence enhances atomic shadowing and creates an inclined columnar microstructure under conditions of limited adatom diffusion. The column inclination angle (β) lies between the film normal and the vapor incidence angle (α) and is dependent upon many factors such as substrate temperature, deposition rate, angular distribution of the deposition flux and background pressure.

Dirk and Leamy [2] had provided an explanation for the inclination of the columns from the vapor direction towards the substrate normal. Fig. 1 shows that oblique incidence causes atoms to be shadowed by neighboring atoms. As a result, the average pair orientation will shift from the vapor incidence direction towards the film normal. The mean pair orientation will depend on the average distance between the atoms and their relative vertical position.

The conservation of parallel momentum has also been used to explain this situation where upon impact the adatoms keep a part of their momentum parallel to the substrate surface. [3] As shown in Fig. 2, when adatoms arrive at the top of a column, it is possible that they will continue to move for a short distance in the direction defined by the vapor beam, causing the column to tilt from the vapor incidence direction towards the substrate normal. This effect is only visible if the random surface diffusion after relaxation is negligible. It has been accepted that there is a relationship between the flux angle (α) and the column angle (β) for a given set of deposition conditions. However, this relationship is complex and poorly understood. The empirical "tangent rule", namely, $\tan(\beta) = (1/2) \tan(\alpha)$, is a simple relationship based on near normal incidence deposition and gives poor results for $\alpha > 50^{\circ}$. [2] Tait *et al.* [4] suggested $\beta = \alpha - \sin^{-1}[1-\cos(\alpha)/2]$ and this gives better results for larger α .

Thus, qualitatively, it can be said that as flux angle increases, β would increase correspondingly, although the exact amount of increase is hard to predict. Under conditions of low adatom mobility, the vapor flux arriving at an increasingly oblique angle would enhance atomic shadowing. This will lead to an increasingly porous columnar microstructure of isolated grains inclined towards the vapor source. [5] This occurs because atomic shadowing produces areas that vapor flux cannot reach directly and the adatom mobility is too low for surface diffusion to fill the voids.



Fig. 1. Shadowing caused by neighboring atoms will shift the mean pair orientation towards the substrate normal [2].



Fig. 2. Conservation of parallel momentum causes the columns to tilt from the vapor incidence direction towards the substrate normal [3].

II. EXPERIMENT

An electron beam evaporator was used to deposit the germanium (Ge) film onto an n-type silicon substrate of resistivity 8-12 Ω -cm that was cleaned in RCA I and II prior to deposition. The substrate was mounted firmly onto a platform which was rotated towards the normal of the source to fix the deposition angle. Deposition was typically carried out at a base pressure of 1x10⁻⁶ Torr and an evaporation pressure of 1×10^{-5} Torr. The evaporation current ranged from 20 to 250 mA and the voltage varied from 3.5 to 4 kV. The substrate is heated by halogen lamps and the substrate temperature can be varied between room temperature and 300°C. Ge pieces of 99.999% purity was loaded into a copper crucible and positioned directly under the centre of the platform. The source to substrate distance was approximately 25 cm. A crystal oscillator was used to monitor the thickness of the Ge film.

Scanning electron microscopy (SEM) was used to examine the film morphology after deposition and Raman spectroscopy was used to examine the crystal quality of the film.

III. RESUTLTS & DISCUSSIONS

Initially, the deposition of the Ge film was carried out at room temperature at three different flux angles of 70°, 80° and 87° at similar deposition rates. This was to investigate the influence of α on the morphology of the film. The SEM images of the Ge film deposited at these α values are shown in Fig. 3. As can be seen, the porosity of the film increases when α increases. A larger flux angle will lead to more anisotropic atomic shadowing, causing more voids to be incorporated into the film. Vick *et al.* [6] noted that a flux angle of greater than 80° is necessary for the shadowing effects of oblique angle deposition to be significant.



(a)





Fig. 3. SEM top-view images of Ge film evaporated at room temperature and under similar deposition rates at flux angles of (a) 70° , (b) 80° and (c) 87° . The shadowing effect becomes enhanced as the flux angle increases, causing columnar morphology to occur at larger flux angles.



Fig. 4. Raman spectra of samples deposited at room temperature with flux angles of $70^\circ,\,80^\circ$ and $87^\circ.$

The Raman spectra in Fig. 4 reveal that all the samples deposited at flux angles of 70° , 80° and 87° are amorphous with broad Ge-Ge peaks at 272 to 276 cm⁻¹.

In order to improve the crystal quality of the Ge film and to maximize the shadowing effect, depositions were carried out at higher substrate temperatures with the deposition rate and α fixed. A higher substrate temperature changes the film morphology towards columnar grains with a smooth matt surface [7]. With a higher substrate temperature, the surface diffusivity of the adatoms will increase, thus improving the crystal quality of the Ge film. The Raman spectra of samples deposited at room temperature, 120°C, 200°C and 250°C (SEM micrographs shown in Fig. 5) are shown in Fig. 6. With an increase in substrate temperature, the FWHM of the Ge-Ge peaks at ~300 cm⁻¹ gets narrower indicating better crystal quality. The SEM image of the sample deposited at 250°C is shown in Fig. 5(a). In agreement with results in the literature [8,9] the column cross section becomes elliptical in shape. This elongated column chain [10], or agglomeration of the columnar film in the direction perpendicular to the vapor flux direction, is commonly known as "bundling". The bundle shape is the result of anisotropic shadowing with more shadowing in the vapor incidence plane than the transverse direction. Thus in order to produce isolated crystalline Ge nanowires, the deposition parameters must be optimized further.

Consequently, the Ge film was deposited at a lower rate of between 0.2-1.5 Å/s, in contrast to the usual rate of 0.2-6Å/s. The SEM image of the resulting morphology is shown in Fig. 5(b). As can be observed, bundling is reduced and isolated nanowires are present. Note that even



WJ5 30.0kV X100K 300mm



Fig. 5. SEM image of Ge film evaporated with a flux angle of 87° and substrate temperature of 250°C and (a) deposition rate of 0.2-6Å/s, (b) deposition rate of 0.2-1.5 Å/s and (c) substrate temperature of 300°C and deposition rate of 0.2-1.5 Å/s. The view is taken with the SEM probe beam parallel to the columns. The image in (a) shows significant bundling whereas in (b) columns are more separated with the presence of isolated nanowires. Inset in (c) is the near cross section SEM image of the Ge film deposited under the same conditions.



Fig. 6. Raman spectra of samples deposited at a flux angle of 87° at room temperature (RT), 120° C, 200° C and 250° C.

though low adatom mobility is a prerequisite for column formation, the mobility of the incident vapor atoms over the substrate must be distinguished from the mobility over the growing film material [2]. Although a lower deposition rate will lead to an increase in diffusivity, as long as this increase in diffusivity does not exceed that of bulk diffusion, adatoms are able to fill the voids between columns.

During nucleation, a larger diffusion distance would lead to a larger capture radius of a nucleus. Consequently, nuclei will be spaced further apart. As growth continues, oblique angle deposition leads to anisotropic shadowing such that columns grow towards the flux direction. However, there is less shadowing in the transverse direction which results in bundling. By ensuring that the spacing between all nuclei is larger in all directions by having a lower deposition rate, bundling can be reduced even though shadowing could not be enhanced in the transverse direction.

Fig. 7 shows the TEM image of the film deposited under the same conditions as that of Fig. 5(b). It reveals that the Ge nanowires are polycrystalline.

By maintaining a lower deposition rate, the substrate temperature was increased to 300° C. The SEM images of the sample deposited under such conditions are shown in Fig. 5(c). Bundling was further reduced and the occurrence of isolated nanowires is increased (see inset of Fig. 5(c)). Increasing the substrate temperature also has the effect of increasing the diffusivity. As long as this increase in temperature is such that bulk diffusion does not become dominant, the morphology will still be columnar. Increasing the substrate temperature has the added



Fig. 7. TEM image of Ge film deposited under the same conditions as Fig. 5(b).

advantage that the film will be more crystalline. The higher substrate temperature will also result in a larger critical nucleus size, thus enhancing atomic self shadowing.

IV CONCLUSION

It has been shown that it is possible to produce crystalline Ge nanowires by the use of oblique angle deposition. By proper adjustment of the deposition parameters, it is possible to reduce the effect of bundling to produce isolated Ge nanowires.

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