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New insights into selected-area deposition of diamond films by means of selective seeding

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Abstract

Polycrystalline diamond films have been patterned on a polished Si substrate by means of selective seeding via hot-filament chemical vapour deposition. In addition to the process of selective seeding, the CH_4/H_2 concentration and the sizes of the patterns have effects on the selectivity. The mechanism of selective growth of diamond is also discussed in this paper.

1. Introduction

Excellent patterning of thin solid films is the basic requirement for realizing their microelectronic applications, especially for diamond films which have many unique chemical and physical properties. However, diamond is difficult to pattern by conventional etching methods because of its chemical inertness and physical hardness. It is also difficult to find a suitable masking material to withstand the erosion even if it can be etched. Therefore, a feasible technique for obtaining patterned diamond films is selected-area deposition (SAD). SAD can be generally achieved if the two regions on the substrate have nucleation density differing by several orders of magnitude.

Since Hirabayashi and Taniguchi reported the selective deposition of polycrystalline and single-crystal diamonds for the first time in 1988 [1], many methods for SAD of diamond have been proposed [2–4]. A new SAD method has been presented by us [5, 6]. Polycrystalline diamond films have been successfully patterned on a variety of substrates by means of selective seeding. In particular, a higher selectivity (about 10^8) was obtained by selective seeding with a double-layer mask. In addition to this process, the CH_4/H_2 concentration and the sizes of the patterns have also influenced the selectivity, as is illustrated in this paper. Further, the mechanism of the selective growth of diamond is also discussed.

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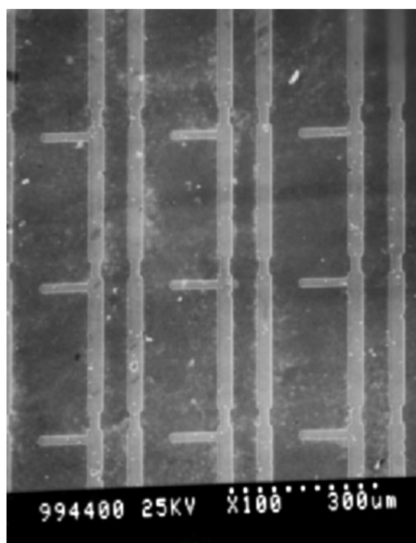

Figure 1. A scanning electron microscopy (SEM) image of selectively deposited diamond film grown at 0.05% CH_4/H_2 concentration.

2. Experimental details

A 300 nm thick TEOS SiO_2 film was grown on a cleaned Si wafer and a 200 nm layer of Mo was then deposited on the SiO_2 film by rf magnetron sputtering. The Si wafer with double layers was patterned by standard photolithography and etched completely in the opening of the photoresist. The Mo layer was etched in a 9:4:14:7 solution of HNO_3, H_3PO_4, CH_4COOH, and H_2O, and the SiO_2 layer was then etched in BOE solution. After the photoresist was removed, the wafer with the pattern of Mo/SiO_2 mask was cleaned in DI water, acetone, and methanol. The seeding processes are the same as in [6]. The wafer with the Mo/SiO_2 mask was turned upwards and shaken slightly in 45% HF acid for about 7 min. The Mo layer with the diamond powder sticking to it was precipitated at the completion of the erosion of the TEOS SiO_2 film. After cleaning and evaporation, the wafer was used to grow the diamond films. The process flow steps are shown in [6]. The conditions of deposition of the diamond films are as follows.

Base pressure:	10^{-3} Torr
Chamber pressure:	40 Torr
Filament temperature:	1900 °C
Substrate temperature:	750 °C
Hydrogen flow rate:	200 sccm
Methane flow rate:	1.0 sccm
CH_4/H_2 ratio:	0.05%
Deposition time:	1 h
Distance between filament and substrate:	1 cm.

3. Results and discussion

The diamond film selectively deposited on the polished Si surface is shown in figure 1. The selectivity of diamond forming between the seeded Si surface and the polished Si surface

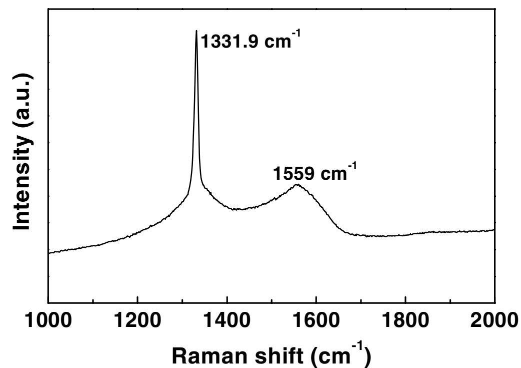

Figure 2. A micro-Raman spectrum of the patterned diamond film deposited at 0.05% CH_4/H_2 concentration after 1 h of deposition.

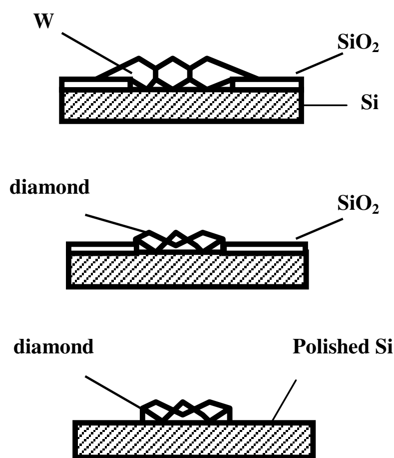

Figure 3. Schematic diagrams of the selective growth of (a) W and (b) diamond film using a SiO_2 film as a mask, and (c) diamond film using a polished Si surface as a 'mask'.

is very high, and the edge delineation follows the defined patterns closely. Therefore, the selective growth of diamond is good.

A Reinshaw Raman microscope (System 1000) was used for analysing the patterned diamond films. The measurement was performed using the 514.5 nm line of an Ar-ion laser and the laser spot size was about 1 μm. Figure 2 shows the micro-Raman spectrum of the patterned diamond films. The peak at 1331.9 cm^{-1} is the characteristic of diamond and the broad peak at about 1559 cm^{-1} is usually observed for graphite and amorphous carbon. Although the CH_4/H_2 concentration is far lower than that under the conventional deposition conditions, the Raman spectra reveal that the quality of the patterned diamond films is good.

In addition, if the polished silicon surface can be abstractly regarded as the mask in the SAD method, no obvious overgrowth on the mask is seen in the selective growth of diamond. The phenomenon has been also found in other SAD methods where a Si_3N_4 or SiO_2 film is used as a mask. This is unlike the case in selective deposition of tungsten, silicon, and GaAs [7–9], where the proximity effect on the mask is obviously observed. Figure 3 shows

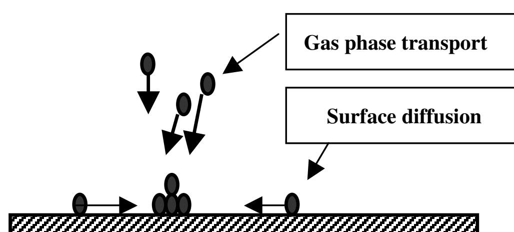

Figure 4. The two main ways of achieving nucleation in the diffusion theory: gas phase transport and surface diffusion.

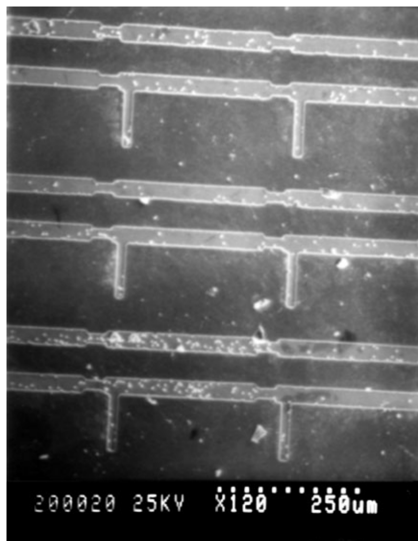

Figure 5. A scanning electron microscopy image of selectively deposited diamond film grown at 0.8% CH_4/H_2 concentration after 1 h of deposition.

schematic diagrams of the selective growth of (a) W and (b) diamond film using a SiO_2 film as a mask, and (c) diamond film using a polished Si surface as a 'mask'.

The two main ways of achieving nucleation in the diffusion theory, gas phase transport and surface diffusion, are shown in figure 4. The proximity effect of the selective growth of W, Si, and GaAs is explained by the fact that the transport of reaction species in the process of growth is by the gas phase, rather than by surface diffusion. The phenomenon of no obvious overgrowth in the selective growth of diamond demonstrates that the transport of the reaction species should be by surface diffusion rather than by the gas phase, and the reaction species have large diffusion lengths.

Figure 5 shows scanning electron microscopy images of selectively deposited diamond films grown at 0.8% CH_4/H_2 concentration after 1 h of deposition by the novel SAD method. Compared with figure 1, more sparse diamond particles appear in the polished area. This is due to the random nucleation of diamond formed on the polished Si surface when the growth condition is 0.8% CH_4/H_2 (within the ranges of conventional diamond deposition conditions). This means that the CH_4/H_2 concentration has affected the selectivity. In fact, CH_4/H_2 concentrations lower than those in traditional diamond deposition conditions are of benefit to the selectivity [10].

In figure 5, most of the random nucleations are formed in 30 μm width lines, whereas few of them are formed in 15 μm width lines. The results reveal that the diffusion length of the reaction species in the process of diamond formation on the polished Si substrate is between 7.5 and 15 μm. More accurate results can be achieved by changing the width of the lines. Further, the diffusion length of the reaction species in the process of diamond formation on the different substrates under the different growth conditions can be also estimated by the SAD method. This work is still in process.

4. Conclusions

By selective seeding with a Mo/SiO_2 mask, polycrystalline diamond film was successfully patterned on a polished Si substrate. Experiments show that CH_4/H_2 concentration lower than those in traditional diamond deposition conditions is beneficial to selectivity. The phenomenon of no obvious overgrowth in the selective growth of diamond means that the transport of the reaction species should be by surface diffusion rather than by the gas phase, and the reaction species have a large diffusion length. The diffusion length can also be estimated by this new SAD method. Work on this is still in process.

Acknowledgment

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