Epitaxial Nucleation and Growth of Diamond Films on Silicon

X. T. ZHOU, and C. H. LEE (Ph. D. Student)

Center of Super-Diamond and Advanced Films (COSDAF) and
Department of Physics and Materials Science

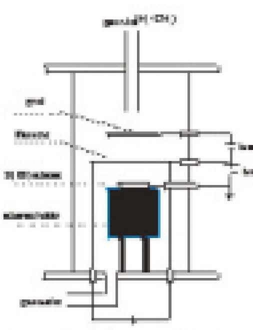
1. Introduction

The quest for artificial methods of diamond production is motivated not only by its gemstone quality but also by its unique set of properties, which make it an excellent candidate for numerous important applications. Diamond was successfully produced in the 1950s by the high-pressure, high-temperature (HPHT) method. An alternative method, chemical vapor deposition (CVD) of diamond at low pressure (typically with the use of a methane-rich CH4/H2 mixture on substrates held at ~700° to 800°C), has also been applied successfully over the last 15 to 20 years. The homoepitaxial growth of diamond on a diamond substrate by CVD methods is relatively well understood. Experimental methods for diamond nucleation on nondiamond substrates (the first necessary step of a heteroepitaxial growth process) have also been developed. The most effective method is bias-enhanced nucleation (BEN), in which the target is biased, with a relatively methane-rich CH4/H2 mixture (several percent methane) as a first step, followed by a conventional CVD step (typically 1% methane or less). However, the nucleation mechanism of diamond on nondiamond substrates remains poorly understood, largely because of the tremendous difficulty of locating and identifying the nucleation sites. This is a major obstacle to further advances in diamond science and technology.

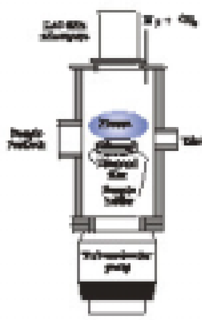
Here we present direct high-resolution transmission electron microscopy (HRTEM) evidence that a step on a single crystalline Si surface serves as a nucleation site for heteroepitaxial diamond growth, and we propose a scheme for the growth of epitaxial diamond films on Si wafers.

2. Experimental

In our Experiment, two-bias assisted hot filament chemical vapor deposition (HFCVD) technique was used for nucleation of diamond on Si(100) and then microwave plasma chemical vapor deposition (MPCVD) technique for growth. After nucleation and growth, the diamond films were characterized by Raman spectroscopy, scanning electron microscopy (SEM) and transmission electron microscopy.



Schematic diagram of the two-bias assisted HFCVD system

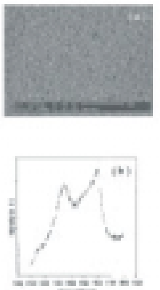


Schematic diagram of MPCVD system

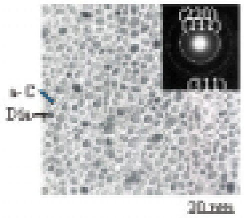
The parameters of nucleation and growth of diamond films

	Total flow rate (SCCM)	CH4 conc. [vol%]	Filament temp. [°C]	Substrate temp. [°C]	Pressure [Torr]	Microwave power [W]	Grid voltage [V]	Grid current, Ig [A]	Substrate voltage [V]	Substrate current, Is [mA/cm²]	Duration [h]
Nucleation	200	3.5	2100	800	30	–	28	0.4	-130	0.3	0.5
Growth	200	1	--	800	30	1300	–	–	--	–	16

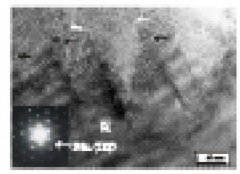
3. Results



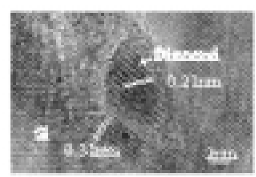
SEM image (a) and a Raman spectrum (b) of the film after nucleation.



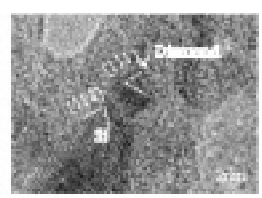
Plan view TEM image at lower magnification of diamond film after nucleation. The nanoparticles with high density can be seen. The inset shows the corresponding transmission electron diffraction pattern of diamond.



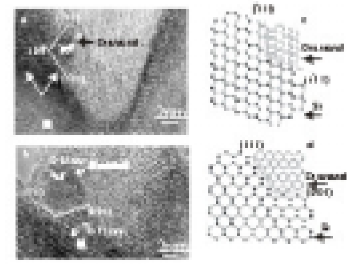
Low-magnification cross sectional view of the interface between the Si substrate and the C film indicates small (2 to 6 nm in diameter), randomly dispersed diamond crystallites. Some have grown on the Si (black arrows) and some are embedded in the a-C matrix (white arrows). Neither a SiC layer nor SiC crystallites are present.



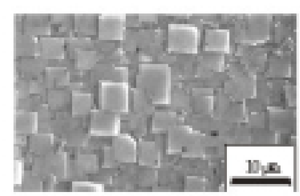
HRTEM image of a diamond crystallite (diameter: 6 nm) grown directly on Si with a random alignment.



HRTEM image of a diamond crystallite (diameter: 3 nm) grown directly on Si with a partially epitaxial alignment.



(a) HRTEM image of a diamond crystallite (diameter: 2nm) grown directly on a Si step with an epitaxial alignment. The interfaces between the Si and the diamond are (111) and (111), intersecting at an angle of 109.5°. (b) HRTEM image of a diamond crystallite (diameter: 6nm) grown directly on a Si(001) surface with an epitaxial alignment. No misorientation between the diamond nucleus and the Si substrate was detected. (c and d) Ball-and-stick diagrams illustrate the interfaces between the diamond crystallites and the Si substrate in (a) and (b).



SEM image of epitaxial diamond film on Si(001) after growth

4. Conclusions

A diamond nucleation site responsible for epitaxial growth of diamond on silicon by chemical vapor deposition is identified in high-resolution transmission electron microscopic images. Other sites in the same sample leading to polycrystalline growth, but deleterious to epitaxial CVD growth, are also described. A mechanism for the heteroepitaxial growth of diamond is suggested, in which etching of the nondiamond carbon binder exposes and removes misoriented nanocrystalline diamond nuclei, leaving intact only those directly nucleated on the Si substrate. This work enhances our understanding of diamond nucleation and heteroepitaxial growth and its potential applications.

5. Publications

1. S. T. Lee, H. Y. Peng, X. T. Zhou, N. Wang, C. S. Lee, I. Bello, and Y. Lifshitz, *Science*, 287 (2000) 104
2. X. T. Zhou, Q. Li, F. Y. Meng, I. Bello, C. S. Lee, S. T. Lee, and Y. Lifshitz, *Appl. Phys. Lett.*, (in press)
3. C. H. Lee, Z. D. Lin, N. G. Shang, L. S. Liao, I. Bello, N. Wang, and S. T. Lee, *Phys. Rev. B61 Issue* 8 (2000) 5579
4. X. T. Zhou, H. L. Lai, H. Y. Peng, C. Sun, W. J. Zhang, N. Wang, I. Bello, C. S. Lee, and S. T. Lee, *Diam. & Relat. Mater.*, 9 (2000) 146