Heteroepitaxial Growth of Diamond on 3C-SiC/Si Substrates by Antenna-Edge Microwave Plasma CVD

Takeru Suto¹, Junya Yaita^{1,2}, Takayuki Iwasaki^{1,2,3}, Meralys Natal⁴, Stephen E. Saddow⁴, Mutsuko Hatano^{1,2,3}

¹ Department of Physical Electronics, Tokyo Institute of Technology. Ookayama, Meguro, Tokyo 152-8552, Japan Phone : +81-03-3726-3999 Email : suto.t.ac@m.titech.ac.jp ² JST-CREST. Chiyoda, Tokyo 102-0076, Japan ³JST-ALCA. Chiyoda, Tokyo 102-0076, Japan ⁴Department of Electrical Engineering, University of South Florida. Tampa, Florida 33620

Abstract

Large-area diamond films are in demand as semiconductor materials for future devices and sensors. Diamond seed crystals were nucleated on both (100) and (111) 3C-SiC/Si substrates for the growth of heteroepitaxial diamond films by a unique antenna-edge microwave plasma chemical vapor deposition system. In this work, we found that lower voltage improved the diamond nuclei orientation resulting in highly epitaxial diamond nucleation on Si substrates via (100) and (111)3C-SiC buffer layers.

1. Introduction

Diamond is an attractive semiconductor material for future devices such as loss-less power devices, high sensitivity sensors [1], or quantum bits [2] due to its superior physical properties of high thermal conductivity, high breakdown field, and long spin coherence time of the nitrogen-vacancy (NV) centers.

Large-area diamond substrates are required to realize these applications. Heteroepitaxial growth of diamond on Si substrates via a 3C-SiC buffer layer offers the possibility to obtain large-size diamond films since the 3C-SiC film is epitaxially grown on Si and the lattice mismatch with diamond is smaller than that of Si. Highly-oriented diamond(100) films have been reported on Si(100) substrates via 3C-SiC(100) buffer layers [3, 4]. Although NV centers in (111) diamond substrates is of demonstrated value in high sensitivity magnetometry [5], heteroepitaxial growth of diamond on 3C-SiC/Si(111) has not been reported to date.

In this study, the diamond nuclei on 3C-SiC/Si of both (100) and (111) orientation were formed by a bias enhanced nucleation (BEN) process [6] in a unique antenna-edge microwave assisted plasma chemical vapor deposition (AE-MPCVD) system (Fig. 1), which was demonstrated to be useful for diamond nucleation [7, 8].

2. Experimental

Single crystal 3C-SiC thin films deposited on Si(100) and Si(111) were used for the diamond growth. Diamond nuclei were formed by the BEN process in AE-MPCVD. AE-MPCVD induces the discharge of gases at the edge of antenna placed over the substrates, and generates a plasma

of high power density, which enhances the nucleation probability. A gas mixture of H₂ and CH₄ were used. The distance from the substrate to the antenna tip was altered to control the temperature. Since the diamond nuclei were very small after the BEN process, we performed the sequential growth on the diamond nuclei to observe diamond morphologies and nucleation density. The nucleation and growth conditions were summarized in Table I. The diamond morphology and density were observed by scanning electron microscopy (SEM) and reflection high energy electron diffraction (RHEED).



Fig. 1 Schematic overview of AE-MPCVD.

	Table	I. AE-	MPCV	D cond	itions
--	-------	--------	------	--------	--------

	Nucleation		[111]	[100]			
	(111)	(100)	growth	growth			
Pressure [kPa]	10	20	5	5			
Power [W]	500	200	600	600			
Bias Voltage [V]	-15	-50	0	0			
CH4 ratio [%]	1	1	0.25	1			
Temperature [°C]	800	750	675	675			
Time [min]	5	10	-	-			

3. Results and Discussion

On the 3C-SiC(100) substrate, as shown in Fig.2a, a number of the nuclei having rectangle-shaped pyramids, which have an epitaxial relationship with the substrate, were observed. By two-step growth toward the [111] direction after the [100] direction growth [9], the nuclei merged and formed a continuous film (Fig.2b). When we applied the larger or smaller voltage, the nuclei lost their orientation.



Fig. 2 SEM images of the diamond synthesized on 3C-SiC(100) after (a) 1 hours growth in the [100] direction and (b) 60 hour growth in [111] direction.

On the 3C-SiC(111) substrate, there seems to be no epitaxially-aligned diamond nuclei at the optimized voltage (-50 V) for the (100) substrate, as shown in Fig. 3a,b. The ringlike diffraction pattern indicates that diamond nuclei were not epitaxially formed on the 3C-SiC surface. By decreasing the bias voltage to -25 V (Fig. 3c,d) and -15 V (Fig. 3e,f), the epitaxial ratio of the diamond nuclei was improved. The epitaxial nuclei with a hexagonal-structure from top-view are observed in the SEM images. The nucleation density was estimated to be about 1.1×10^8 cm⁻² under a bias voltage of -15 V. It is worth noting that lower and higher bias voltages resulted in no nucleation and less epitaxial nucleation, respectively. Although the density decreased significantly at the low bias voltage (-15 V), to the best of our knowledge, this is the first demonstration of epitaxial diamond nucleation on 3C-SiC(111), which would be attributed to the high plasma density in the AE-MPCVD system. Despite the same material, the optimal bias voltage in synthesizing diamond nuclei on 3C-SiC(111) is 3 times smaller than that of (100), which would come from the difference in the bond energy between diamond and 3C-SiC, gas migration on the 3C-SiC surface, and/or stability of the nucleation sites.

4. Conclusions

Diamond epitaxial nucleation was performed on 3C-SiC(100) and (111) surfaces by using AE-MPCVD. At the BEN process on the (111) surface, the lower voltage improved the nuclei orientation compared with the (100) substrate. By using lower voltage with AE-MPCVD, the present work show it is possible to synthesize highly epitaxial diamond nuclei on 3C-SiC(111)/Si(111). Single crystal diamond (111) thin films would be expected after longer growth time.

Acknowledgments

This work was supported in part by MEXT/JSPS KA-KENHI Grant No. 26820110 and the Asahi Glass Foundation.

References

[1] Taylor, J. M. Cappellaro, P. Childress, L. Jiang, L. Budker, D.;

Hemmer, P. R. Yacoby, A. Walsworth, R. Lukin, M. D., Nature Physics 2008, 4 (10), 810-816.

- [2] Hanson, R. Awschalom, D. D., Nature 2008, 453 (7198), 1043-1049.
- [3] T. Suesada, N. Nakamura, H. Nagasawa and H. Kawarada, Jpn. J. Appl. Phys. 34 (1995) 4898.
- [4] J. Yaita, T. Iwasaki, M. Natal, S. E. Saddow, M. Hatano, Jpn. J. Appl. Phys. 54 (2015) 04DH13.
- [5] Fukui, T. Doi, Y. Miyazaki, T. Miyamoto, Y. Kato, H. Matsumoto, T. Makino, T. Yamasaki, S. Morimoto, R. Tokuda, N. Hatano, M. Sakagawa, Y. Morishita, H. Tashima, T. Miwa, S. Suzuki, Y. Mizuochi, N., Applied Physics Express 2014, 7 (5).
- [6] S. Yugo, T. kanai, T. Kimura and T. Muto, Appl. Phys. Lett. 58 (1991) 1036.
- [7] N. Taniyama, M. Kudo, O. Matsumoto, and H. Kawarada, Jpn. J. Appl. Phys. 40, L698 (2001).
- [8] T. Fujisaki, M. Tachiki, N. Taniyama, M. Kudo, and H. Kawarada, Diamond Relat. Mater. 11, 478 (2002)
- [9] C. Wild, P. Koidl, W. Muller-Sebert, H. Walcher, R. Kohl, N. Herres, R. Loncher, R. Samlenski and R. Brenn. Diam. Relat. Mater. 2 (1993) 158.



Fig. 3 SEM images and RHEED patterns of the diamond nuclei synthesized on 3C-SiC(111) after the 2 hours growth. The nucleation were performed under bias voltages of (a, b) -50V, (c, d) -25V, (e, f) -15V. The main diffraction spots from diamond were indicated by yellow triangles.