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Structural analysis of Nitrogenated Ultrananocrystalline Diamond/Amorphous Carbon Composite Films Fabricated by Coaxial Arc Plasma Deposition

Hiroki Gima, Abdelrahman Zkria, and Tsuyoshi Yoshitake

Department of Applied Science for Electronics and Materials,

Interdisciplinary Graduate School of Engineering, Kyushu University

Kasuga-koen, Kasuga, Fukuoka 816-8580, Japan

Abstract

Chemical bonding structure of nitrogenated ultrananocrystalline diamond/amorphous carbon composite (UNCD/a-C) films fabricated by coaxial arc plasma deposition (CAPD) was mainly investigated by X-ray photoemission (XPS) and Near-Edge X-ray absorption Fine Structure (NEXAFS) spectroscopies. Nitrogen content in the film was estimated 3 at. % by XPS spectra,. From NEXAFS spectra, intensity of π*C=N bonding increased by nitrogenation. Additionally intensity of σ*C-H slightly decreased with adding nitrogen into the film. These results suggest that hydrogens located interfaces of diamond grain and grain boundaries were replaced to nitrogen and it contribute to n-type conduction and enhancement of electrical conductivity.

1. Introduction

UNCD/a-C is a new candidate carbon semiconductor, and the application to electrical devices has received much attention [1]. It has been known that nitrogen-doped for single crystal diamond is ineffective for realizing n-type conduction at room temperature because nitrogen form a deep donor level of 1.7 eV below the bot-tom of the conduction band in diamond. For a-C, n-type conduction is realized by nitrogen incorporating, however it is difficult for the carrier density to be controlled widely. On the other hand, it has been reported that nitrogen incorporating makes possible n-type conduction accompanied by an enhancement in the carrier density for UNCD/a-C films prepared by chemical vapor deposition (CVD)[2] and pulsed laser deposition (PLD)[3-5]. On the other hand, recently, CAPD is paid attention from industrial viewpoints such as high deposition rate (400 nm/min) and a simple apparatus constitution and so on. So far, It has been experimentally demonstrated n-type UNCD/a-C film prepared by CAPD under hydrogen and nitrogen mixture gas atmosphere[6, 7]. However, its origin has not been investigated enough yet. Thus, in this work, chemical bonding structures of nitrogenated UNCD/a-C film fabricated by CAPD were studied by spectroscopic analysis.

2. Experimental methods

Undoped and nitrogenated UNCD/a-C films with a thickness of approximately 300 nm were deposited by CAPD at a substrate temperature of 550 C and an ambient hydrogen and nitrogen mixture gas pressure of 53 Pa. Inflow ratio of hydrogen and nitrogen ($I_{N/H}$) was set 0 and 0.25. The quartz and p-type Si (100) were used as substrates for the evaluation of electrical conductivity and chemical bonding configurations, respectively. XPS spectroscopy was carried out using the Mg Kα line (source energy of 1253. 6 eV). NEXAFS was measured at beamline 12 (BL12) of the SAGA Light Source (SAGA-LS).

3. Result and discussion

X-ray photoemission spectra, which were measured with MgKα line, of the UNCD/a-C films prepared at different $I_{N/H}$ are shown in **Fig. 1**. N1s peaks strengthened with the $I_{N/H}$, which evidently indicates that the nitrogen content in the film increased with the $I_{N/H}$. The nitrogen contents of the films deposited at $I_{N/H}$ of 0 and 0.25 were estimated to be 0 and 3 at. %, respectively. O1s is caused by adsorbates of film surface. The adsorbates is only the surface and also can be removed easily by Ar sputtering so that it should not be critical influence as semiconducting bulk property. And the nitrogenated film acts as semiconductor and n-type film as reported previously[6, 7].

The C K-edge NEXAFS spectra of undoped and nitrogenated films are shown in **Fig. 2**. All spectra nomarized at 330 eV along with previous report[5]. The nitrogenated film exhibited three additional component spectra compared with the undoped film. The spectra with peak positions at 286.8 and 297.8 eV are probably due to π*C=N and σ*C=N. It suggest that π*C=N is located in diamond grain interfaces or grain boundaries and work as donor.

4. Conclusion

It was revealed that intensity of π*C=N observed by NEXAFS spectra increased with decreasing σ*C-H. It suggests that hydrogens located interfaces of diamond grain or grain boundaries were replaced to nitrogen and it contribute to n-type conduction and enhancement of electrical conductivity. Thus, it intimated that controlling state of π*C=N bonding was key of the electrical properties on nitrogenated UNCD/a-C prepared by CAPD.

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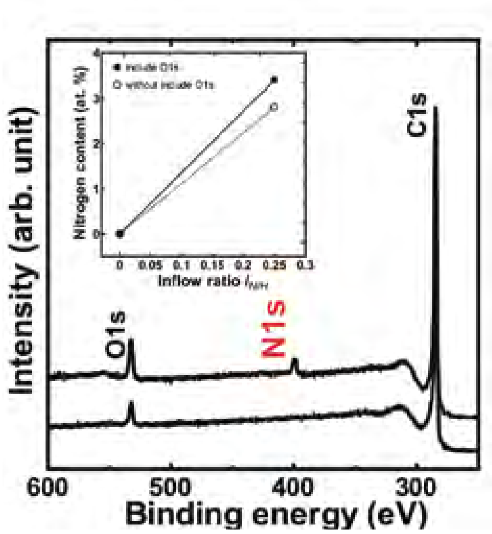

Figure 1. XPS wide-scan spectra of undoped and nitrogenated UNCD/a-C.

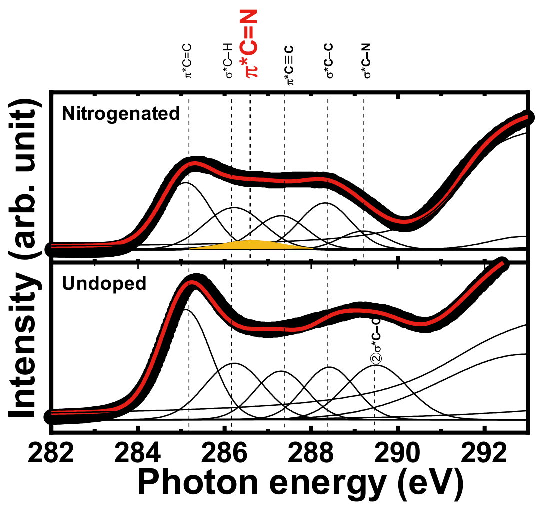

Figure 2. C K-edge NEXAFS spectra of undoped and nitrogenated UNCD/a-C.

Email: hiroki_gima@kyudai.