Application of Nitrogen-doped Ultrananocrystalline Diamond/Hydrogenated Amorphous Carbon Composite Films to Electronic Devices

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論　文　内　容　の　要　旨

Thesis Summary

Nanostructured carbon films including diamond, diamond like carbon (DLC), single-layer graphene, and carbon nanotubes (CNT), have received a great scientific and technical interest for applications in many fields. Ultrananocrystalline diamond (UNCD) film is a unique formation of diamond, which was grown through a patented microwave plasma-enhanced chemical vapor deposition (MPCVD) technique. The ultra-smooth surface offered by nanometer scale grain sizes of UNCD films, together with other outstanding properties, such as, thermal stability, chemical inertness, high electron field emission, low friction coefficient, as well as the ability to incorporate n-type dopants, have attracted many scientists towards the application of UNCD films in optoelectronics devices, microelectromechanical systems (MEMS), field emission source, and biomedical applications. Over many trials, one of the most challenging issues in the diamond field is the growth of n-type diamond. Although, nitrogen is often used to create n-type conduction, nitrogen doped diamond is an electrical insulator at room temperature due to the deep level of 1.7 eV of the nitrogen impurity. On the other hand, nitrogen dopant in UNCD films (in pure form or as composites of nanocrystalline diamond embedded in an amorphous matrix UNCD/a-C:H) realize n-type conduction with enhanced electrical conductivities.

In this study, nitrogen-doped ultrananocrystalline diamond/hydrogenated amorphous carbon composite (UNCD/a-C:H) films were synthesized by coaxial arc plasma deposition. The results of temperature dependence of electrical conductivity imply that carriers are transported in hopping conduction. In addition, heterojunctions comprising nitrogen-doped UNCD/a-C:H films and p-Si substrates exhibited a typical rectifying action. It was experimentally demonstrated that nitrogen-doped UNCD/a-C:H is applicable as an n-type semiconductor. The origin of the n-type conduction was correlated to the chemical bonding structural evaluation results. Briefly, this study divided into seven chapters as follows:

In Chapter 1, we introduce the background of carbon-based nanomaterials include diamond films and their feature properties, with highlighting the ultrananocrystalline diamond films, as a core of our study.

Chapter 2 presents the experimental techniques that have been employed along the study, with the explanation of physical meaning and mechanisms behind each techniques. It includes the film preparation methods and evaluation techniques.

In Chapter 3, undoped UNCD/a-C:H films prepared by CAPD method was discussed based on our previous work. The chapter shows hydrogenation effects on the growth of UNCD/a-C:H films, effects of boron doping on the electrical properties of UNCD/a-C:H films as well as the application of UNCD/a-C:H films in hard coating.

In Chapter 4, we focused on the effects of nitrogen incorporation on the physical properties of CAPD-deposited UNCD/a-C:H films. the origin of the n-type conduction accompanied by an enhancement in electrical conductivity of nitrogen-doped UNCD/a-C:H films is discussed and correlated to the chemical bonding

structural evaluation results obtained by sensitive spectroscopic measurements, such as, XRD, XPS, and NEXAFS, measured in Saga Light Source (Saga-LS) synchrotron center. The results suggested that hydrogen atoms that terminate diamond grain boundaries will be partially replaced by nitrogen atoms and, consequently, π C–N and C=N bonds that easily generate free electrons will be formed at grain boundaries. Moreover, from the temperature dependence of the electrical conductivity, consideration of the carrier conduction mechanism was studied, it was found that the carrier follow variable range hopping conduction mechanism. This is an electronic property is considered to be an important knowledge for device application. Beside their attractive electrical properties, nitrogen-doped UNCD/a-C:H films possess large absorption coefficients of more than 10^5 cm^{-1} at photon energies from 3 to 5 eV. The optical band gap which should be attributable to an a-C:H matrix in UNCD/a-C films was shrinkage by nitrogen doping, which refers to increased sp2 fractions in the films by nitrogen doping.

Chapter 5 figures out the fabrication of heterojunction diodes comprising nitrogen-doped UNCD/a-C:H films as n-type semiconductor and p-type Si substrates. The fabricated diodes evaluated based on current-voltage (I-V) and capacitance-voltage (C-V) characteristics. Dark I-V characteristics reveal a rectification ratio of more than four orders of magnitude, which confirm that the fabricated junctions are working as typical diodes. Furthermore, C-V measurements afford a valuable information about carrier density and energy band diagram. Moreover, .n- UNCD/a-C:H/p-Si diodes were employed as an ultraviolet photodetectors. Under 254 nm monochromatic light, photodiode show a capability of deep ultraviolet light detection, which attributed to the existence of UNCD grains.

In chapter 6, Current transport mechanisms through heterojunction diodes comprising n-type UNCD/a-C:H films and p-type Si substrates, were electrically characterized by *J–V–T* characteristics method, measured in the temperature range of 80–300 K. The results suggested that the multi-step trap-assisted tunneling is the dominant transport mechanism in this temperature range at low forward bias voltage below 0.45 V.

Chapter 7 summarizes the results and discussions from chapter 4 to chapter 6 and looks forward to future research perspectives. It is thought that the application of UNCD/a-C:H ultraviolet sensor and device application as a solar cell absorption layer will be embodied by efforts to further improve the light receiving characteristics in the near future, as all-carbon solar cell made solely from nanocarbon material, namely UNCD/a-C:H films.