Thermal and Electrical Properties of Ultrananocrystalline Diamond/Amorphous Carbon Composite Films

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（超ナノ微結晶ダイヤモンド／アモルファスカーボン混相膜の熱および電気特性）

区　　分　：甲

論 文 内 容 の 要 旨

This doctoral thesis describes the relationship of structure of grain boundaries and the thermal and electrical properties of ultrananocrystalline diamond (UNCD)/hydrogenated amorphous carbon (a-C:H) composite (UNCD/a-C:H) films toward their semiconducting application.

UNCD/a-C:H films comprise a large number of nanodiamond with diameter of less than 10 nm and a-C:H matrix. The existence of a huge number of grain boundaries is structural specific to UNCD/a-C:H films. UNCD/a-C:H films has the following specifics: (1) the growth of UNCD/a-C:H films on foreign substrates is easier than poly- and single-crystalline diamond; (2) the control of conduction type to both p- and n-type by boron and nitrogen doping, respectively. While the preparation of UNCD/a-C:H films have been carried out mainly by chemical vapor deposition (CVD), we have succeeded in the formation of UNCD/a-C:H films by coaxial arc plasma deposition (CAPD). Furthermore, a rectifying action in pn junctions comprising UNCD/a-C:H films and Si substrates were observed. While preparation of UNCD/a-C:H films and control of conduction type for application to semiconductor devices were succeeded, the relationship between structure of UNCD/a-C:H films and the origin of their physical properties such as thermal and electrical properties are not well understood.

The main goal of this study is to reveal the relationship between structural, chemical properties of grain boundaries and the thermal and electrical properties of UNCD/a-C:H films prepared by CAPD method for application to semiconducting devices. The doctoral thesis is consisted of 6 chapters.

Chapter 1 introduces the classification of carbon-based nanomaterials, and concretely compares UNCD/a-C:H with related materials. In addition, it introduces the study on the grain size dependence of electrical and thermal properties of UNCD/a-C:H prepared mainly by chemical vapor deposition (CVD). Furthermore, historical background of UNCD/a-C:H prepared by CAPD in our laboratory are described. Finally, we show the scope of this study.

Chapter 2 presents experimental techniques employed in our study, including film preparation method, structural evaluation, chemical bonding evaluation, and physical properties measurements, and explains the principle and detail conditions .

In chapter 3, hydrogenation effects on the thermal conductivity and interfacial conductance in grain boundaries were investigated using a time-domain thermoreflectance method. The interfacial conductance was estimated to be 1,010 and 4,892 MW/(m^2·K) for UNCD/a-C:H and UNCD/non-hydrogenated amorphous carbon (a-C) composite (UNCD/a-C) films, respectively. Reasons for the hydrogenated film having lower interfacial conductance than the non-hydrogenated film are 1) the reduced number of carriers that contribute to heat transport and 2) the hydrogen atoms, which are preferentially located at the grain boundaries and enhance phonon scattering. It was experimentally demonstrated that the thermal conductivity of UNCD/a-C(:H) films is reduced by hydrogenation; i.e., the thermal conductivity of UNCD/a-C(:H) films is controllable with hydrogenation.

In chapter 4, defect structures of boron-doped UNCD/a-C:H films prepared by CAPD were investigated from the viewpoint of chemical bonding structures and their carrier lifetime were measured. The near-edge X-ray absorption fine-structure spectrum and Fourier transform infrared spectrum implied that boron incorporated into the films are preferentially replaced by hydrogen atoms connected to sp^2 carbon. Electron spin resonance measurement revealed the number of defects decreased with increasing boron content in the films, which might be due to the suppression of the sp^2 carbon which act as a carrier source and efficient termination of dangling bonds by incorporated boron. From the carrier lifetime measurement, the lifetime of minority carriers (τ) of 0 (undoped), 0.5, and 2.0 at.% boron-doped UNCD/a-C:H films were estimated to be 0.43, 0.31, and 1.96 μs, respectively. A reason why τ enlarges with increasing boron content is that trap centers such as dangling bonds for minority carriers are effectively terminated by boron atoms.

In chapter 5, spin injection was studied for trilayers comprising UNCD/a-C:H interlayers, which have not been reported previously. The trilayered spin-valves can be fabricated by physical vapor deposition (PVD) with a mask method. Whereas spin-valve signals are observed at low temperatures in GMR and TMR, the trilayers consisting of UNCD/a-C:H films shows spin-valve effect at room temperature, which might be attributed to the hopping conduction of UNCD/a-C:H films. Even at an interlayer thickness of 50 nm, they clearly showed spin-valve signals. Its interlayer thickness value might be a sufficient length for application as gate materials for spin transistors. This is the first demonstration of spin-injection into and spin-transport in diamond related materials.

Chapter 6 summarizes obtained achievements from chapter 3 to 5 and shows a future perspective.