[I~13] [초청]

MICROWAVE PLASMA-ENHANCED CHEMICAL VAPOR DEPOSITION AND CHARACTERIZATION OF (001) HOMOEPITAXIAL DIAMOND FILMS

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The surface morphologies and structure of (001) homoepitaxial diamond films prepared by microwave plasma-enhanced chemical vapor deposition (MPECVD) were investigated. A particular emphasis was placed on the description of the epitaxial growth mechanisms by linking the surface morphologies and structure to the deposition parameters.

A systematic study of the surface morphologies and structure of (001) homoepitaxial diamond films with the deposition parameters indicates that growth of these films depends strongly on the surface misorientation angles of substrates toward the [110] or [1$\bar{1}$0] direction, methane concentrations, and growth temperatures.

Remarkably different surface morphologies were observed on etched substrates and homoepitaxial diamond films with different misorientation angles. On the etched surface, the density of etch pits decreased drastically by increasing the misorientation angles. For the as-grown films, growth hillocks and macrosteps occurred on the well-oriented and misoriented substrates, respectively. Step-flow growth resulted in higher growth rates than hillock growth. The dependence of the etching and growth morphologies as well as the growth rates on the misorientation angles indicates that surface steps play a significant role in diamond etching and growth. In this study, therefore, etching and growth mechanisms on the diamond (001) surface were considered in terms of the reactions of atomic hydrogen and hydrocarbon precursors with the atomic steps and terraces on the surface, respectively. It is proposed that hillock growth occurs through two-dimensional nucleation on terraces when the density of surface steps is low, while the step-flow growth proceeds along the <110> directions on the substrates with the high density of steps.

The homoepitaxial diamond growth with a low methane concentration of 1% produced macrosteps with the surface close to the single-domain structure, while at high methane concentrations of 2% and 6%, growth hillocks and random growth features occurred with the double-domain surface, respectively. The variation of surface morphologies and structure with the methane concentrations is attributed to lower mobility and shorter diffusion length of adsorbates on the surface at higher methane concentrations.

During the step-flow growth, step bunching was more apparent at a lower temperature of 875 °C than at 1200 °C. In terms of surface diffusion of adsorbates as well as step bunching, the high temperature growth seems to be more promising for the deposition of high-quality and smooth films. Step-flow growth with the single-domain surface is believed to produce higher-quality films with fewer lattice defects than other growth modes. It is likely that the step-flow growth is favored by increasing the misorientation angles, lowering the methane concentrations, and increasing the growth temperatures. This study suggests that the deposition condition for (001) homoepitaxial diamond films is optimized with misorientation angles of approximately 3-5°, methane concentrations of less than 1%, and substrate temperatures of around 1200 °C.

Reflection high-energy electron diffraction (RHEED) has been used to study the surface structure of the diamond (001) substrates annealed in H plasma and of as-grown films. The 2x1 surface reconstruction occurred on all diamond (001) substrates annealed at 650 to 1300 °C in H plasma. It was found that reconstruction takes place at a much lower temperature in H plasma than in ultra-high vacuum. This indicates that CVD diamond growth proceeds on the reconstructed surface over the range of temperatures investigated.

The domain and step structure of the diamond (001) surface was investigated by comparing the intensities of two series of the half-order RHEED spots. The surface annealed in H plasma showed the

transition from the double-domain to the nearly single-domain structure by increasing the misorientation angles. This transition is temperature-dependent. The as-grown films exhibited the double-domain structure on the well-oriented surface and the surface close to the single-domain structure on the misoriented surface. When the surface was close to the single-domain structure, type-A terraces dominated the H-plasma annealed surface while type-B terraces were the major domain of the as-grown film surface. This implies that D_A and D_B steps are present on the H-plasma annealed surface and on the as-grown surface, respectively. D_A steps have never been observed even on the Si (001) surface. It is inferred that the step formation energies increase in the order of D_A, S_A+S_B, and D_B on the surface annealed in H plasma and in the order of D_B, S_A+S_B, and D_A on the as-grown films.

The diamond (001) films which were homoepitaxially grown with boron doping and subsequently followed by H-plasma annealing, were also investigated using scanning tunneling microscopy (STM). In this study, an atomic resolution was achieved so that individual dimers and even individual atoms in the dimers could be seen. High-resolution atomic images showed that on the H-terminated diamond (001) surface, the 2x1:H monohydride structure was predominant, but the 1x1:2H structure and local 3x1:1.33H configuration existed only in a very local area and in a very low concentration. For the H-plasma annealed diamond (001) surface, the double-domain structure was observed at a low misorientation angle while the single-domain structure occurred with type-A terraces and D_A steps at a high misorientation angle. The STM observations of the double-domain and single-domain structures with the misorientation angles were in good agreement with the RHEED results. D_A steps which were predicted to be present on the single-domain surface annealed in H plasma by a RHEED study, were confirmed by STM. Atomic images also revealed several types of surface defects including antiphase boundaries, islands, and dimer vacancies on the diamond (001) surface.