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Step growth in single crystal diamond grown by microwave plasma chemical vapor deposition

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Abstract

Single crystal diamond films of varying quality are deposited using microwave plasma chemical vapor deposition (MPCVD) apparatus. Unpolished natural diamond seeds are used as substrates in the temperature (T_s) range 850–1200 °C. The gas mixture of methane (CH₄), hydrogen (H₂) and oxygen (O₂) is used for the deposition of diamond. The deposition pressure is varied in the range 90 to 150 Torr. The films are characterized using scanning electron microscopy (SEM), Atomic force microscopy (AFM) and Raman spectroscopy techniques. The growth morphology of the films is found to be a sensitive function of the deposition parameters. The crystalline nature of the films change from polycrystalline to single crystal as we increase T_s and for a certain set of parameters the filamentary growth of the diamond crystals can be seen. The films are polycrystalline in the range of substrate temperature 850–900 °C and oriented grains of diamond crystals are evident as the T_s increases. The single crystal diamond growth is observed to proceed via the step growth mechanism with the evidence of bunching of the steps. Our study explores evolution of the growth of single crystal diamond in a wide range of parameters. (© 2005 Elsevier B.V. All rights reserved.

Keywords: Single crystal diamond films; MPCVD; Step growth\

1. Introduction

Diamond thin films deposited on natural single crystal diamond substrates have attracted great deal of attention recently [1,2], since diamond is one of the most attractive materials for both high power and frequency electronic devices suitable for high temperature and chemically harsh environment [3–7]. Large area crystal diamond films have great value commercially and scientifically due to their colour, quality and size. Diamond synthesized under high pressure and temperature environment (popularly known as HPHT diamond) always contain catalytic impurities such as nickel, iron, or cobalt [6–10]. A key limitation in the development of the homoepitaxial diamond films is the low growth rate. Recently, however the

growth rate of the diamond has increased substantially in a narrow parameter window using microwave plasma chemical vapor deposition technique (MPCVD) [1,11]. The high growth rate of chemical vapor deposited diamond would ensure that single crystal diamond films could easily be available for a variety of applications such as electronics devices, SAW devices and heat sinks. Single crystal diamond films have obvious advantages over its polycrystalline counterparts in that the grain boundaries are completely absent and thus the electrical and optical properties can be accurately and precisely determined.

As the single crystal diamond films grown in high growth regime are recent materials, it is essential to explore the parameter range thoroughly and grow the films encompassing the whole range. Moreover, it would be also a unique opportunity to study the morphology of the single crystal diamond films, their growth habitat, faceting and growth mechanism. In this paper we report on the morphology of single crystal diamond films grown at various pressure, temperature and flow rates of gases. We find that the single

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Fig. 1. The SEM images of the diamond films; (a) the image of diamond film grown at higher CH₄ percentage showing filamentary structure, (b) steps on the surface of a single crystal film, (c) and (d) show respectively the morphology of the film.

crystal diamonds grow via step growth route with a spiral growth model. Scanning electron microscope (SEM), Raman spectroscopy and Atomic force microscopy (AFM) results are reported.

2. Experimental

Single crystal diamond films were deposited using a MPCVD reactor of 2 kW microwave power coupled into an optimally designed plasma chamber. The plasma chamber is

a stainless steel double walled water-cooled reactor. A molybdenum substrate holder is modified to get uniform dense plasma of 2 in. diameter. The substrate temperature was controlled by controlling microwave power and the distance between plasma ball and substrate. No external substrate heater was used. I_s was measured using an IRCON make Optical Pyrometer. Unpolished natural IIa diamond seeds $(3 \times 3 \times 1 \text{ mm}^3)$ were used as substrates. A gas mixture of methane (CH₄), hydrogen (H₂) and oxygen (O₂) was used for the deposition. The rates of methane and hydrogen flow



Fig. 2. Bright field (TEM) image of the carbon filament grown on the single crystalline diamond substrate with higher CH₄ percentage.

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