



Effect of substrate morphology on the roughness evolution of ultra thin DLC films

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ABSTRACT

The effect of substrate roughness on growth of ultra thin diamond-like carbon (DLC) films has been studied. The ultra thin DLC films have been deposited on silicon substrates with initial surface roughness of 0.15, 0.46 and 1.08 nm using a filtered cathodic vacuum arc (FCVA) system. The films were characterized by Raman spectroscopy, transmission electron microscope (TEM) and atomic force microscopy (AFM) to investigate the evolution of the surface roughness as a function of the film thickness. The experimental results show that the evolution of the surface morphology in an atomic scale depends on the initial surface morphology of the silicon substrate. For smooth silicon substrate (initial surface roughness of 0.15 nm), the surface roughness decreased with DLC thickness. However, for silicon substrate with initial surface roughness of 0.46 and 1.08 nm, the film surface roughness decreased first and then increased to a maximum and subsequently decreased again. The preferred growth of the valley and the island growth of DLC were employed to interpret the influence of substrate morphology on the evolution of DLC film roughness.

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1. Introduction

The ultra thin diamond-like carbon (DLC) films are widely used as protective films for many industrial applications, such as data storage and gas barrier, due to their superior structural and functional properties [1,2]. An important application of ultra thin DLC film is used as protective overcoat on the head and disk in magnetic storage device [3–5]. In order to increase the storage density over 1 Tbit/in.², it is desirable to reduce the head/disk magnetic spacing to less than 10 nm, so as to the DLC film with 1–2 nm thickness and extremely small roughness is required [5].

The evolution of the surface roughness of growing films provides much information about their growth mechanism. There are many growth models, such as layer-by-layer growth mode, island growth mode, Edwards–Wilkinson (EW) growth mode [6] and so on. It is found that the roughness evolution behaves in different ways depending on the dominant process. Casiraghi et al. [7] first presented the measurement of the roughness evolution of ultra thin DLC deposited on silicon (1 0 0) substrate with roughness of ~0.2 nm. They found that the surface had an extremely small roughness and growth exponent. Quantum and classical molecular simulations have shown that an efficient damping of

surface fluctuations is achieved through impact-induced downhill currents, eroding hills on the film surface, in agreement with the EW model. Liu et al. [8] found that the silicon substrate surface showed the highest R_q value (0.195 nm for tapping mode AFM image) compared to those of ta-C coatings and the R_q value of ta-C coatings decreased when their thickness increased from 1.4 to 11 nm. This is resulting from that the kinetic energy of the ions could efficiently eliminates voids, asperities and rough particles by their surface mobility during the DLC coating growth [9]. However, the effect of the silicon substrate surface morphology on the ultra thin DLC film growth, especially in the case that the substrate is not atomically smooth, have not been investigated experimentally. In the computer simulation aspect, the Monte Carlo [7] and molecular-dynamics (MD) simulations are usually used to study the film growth mechanism. Especially, the MD simulations are performed to study the thin film growth by ion beam deposition method [10–12], where the energetic carbon atoms are deposited onto the diamond substrate with normal incidence. The effects of surface roughness on DLC film growth on diamond substrate was first theoretically investigated by Ma [13] using MD simulation. They found that the film showed a preferred growth at the valley resulting in smoothening of the film in the initial stage and showed a homogenous growth mode in the later stage. However, the effect of substrate material on the DLC growth has not been taken into account because of that the substrate in the simulation was diamond instead of silicon. In this paper, the evolution of the

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surface roughness as a function of the film thickness of ultra thin DLC film deposited on silicon substrate with different surface roughness was investigated to reveal the effect of substrate roughness on film growth.

2. Experimental details

Among the present methods for the preparation of DLC films, the filtered cathodic vacuum arc (FCVA) deposition method has been known to produce a highly ionized plasma and consequent produce highly tetrahedrally bonded amorphous carbon (ta-C) films and can deposit onto insulating substrates as the deposition beam is a neutral plasma beam [14–16]. Considering the advantageous, the ultra thin DLC films studied in this paper were deposited on silicon wafers by a FCVA system. The silicon wafers were cleaned ultrasonically in the acetone and alcohol for 15 min, respectively and then immersed in hydrofluoric acid solution with concentration of 0.5% for 15 s to remove the oxide layer on the wafer surface. The surface roughness of silicon wafer after HF solution treated was 0.15 nm. In order to obtain different surface morphology, the substrates were sputtered by Ar⁺ ions with energy of 500 eV, beam current of 50 mA, and incident angle of 45° to roughen the silicon wafer surface. Different surface roughness can be achieved depending on sputtering time. The chamber was evacuated to a base pressure less than 5×10^{-4} Pa, and the working pressure increased to about 2×10^{-3} Pa. The distance between the substrate and the filter exit was 600 mm. The films were prepared under a pulse bias with duty ratio of 20% and amplitude of -100 V. The deposition duration was varied in the range of 0–15 min in order to obtain ultra thin DLC films with thickness of 0–20 nm.

Raman spectroscopy was conducted to investigate the structure of the as-prepared film with a Renishaw R-1000 system using wavelength of 514.5 nm. The deposition rate was calculated from the film thickness measured by a JEM-2010F transmission electron microscope (TEM) and the deposition duration. A digital instruments NanoScope IIIA atomic force microscopy (AFM) was employed to observe the surface morphology of the films with tapping mode. For each sample, four images were taken and an average roughness was obtained.

3. Results

To characterize the structure and the thickness of the DLC film deposited on the silicon substrate, the cross-sectional sample of DLC film was prepared for TEM observation. Fig. 1 shows the cross-sectional TEM image of the DLC film with deposition duration of 15 min. As shown, the typical amorphous structure of DLC film was observed and the film thickness of DLC was measured to be 20 nm. As a consequent, the deposition rate was calculated to be 1.33 nm/min. Thus, a series of DLC films with different nominal thickness can be obtained via controlling the deposition duration.

The Raman spectrum in Fig. 2 shows three features at the positions of 1360, 1450, and 1560 cm^{-1} , which are labeled as D, Si, and G peak, respectively. The G peak is due to the bond stretching of all pairs of sp^2 atoms in both rings and chains, and the D peak is due to the breathing modes of rings. Due to the ultra thin thickness of DLC film, the Si peak comes from the Si substrate also can be found. In order to obtain the position and intensity of the D and G peaks, we fitted the spectra using Gaussian functions. The positions of D and G peaks were calculated to be 1389 and 1544 cm^{-1} , and the sp^3 content was estimated to be about 60% according to Ref. [17].

The surface morphology of the three substrates with different initial roughness of 0.15, 0.46 and 1.08 nm measured by AFM is

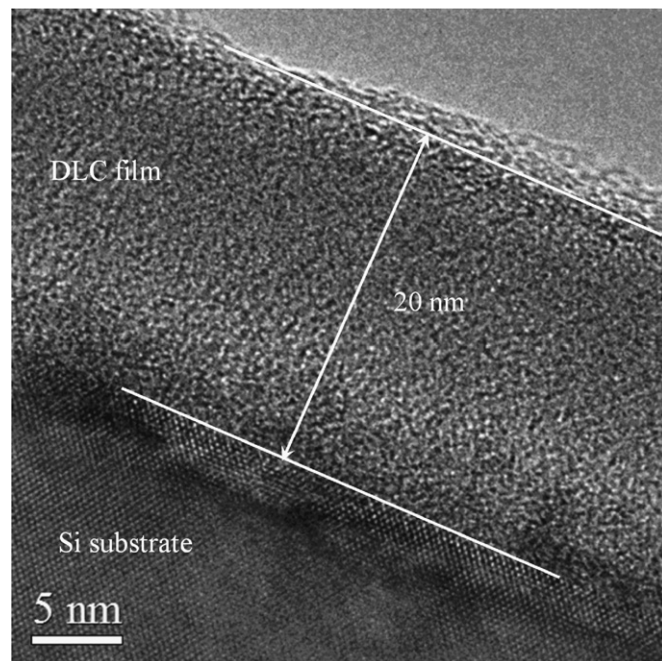


Fig. 1. Cross-sectional TEM image of DLC film with deposition duration of 15 min.

shown in Fig. 3. According to the section analysis, the peak-to-valley distance, the valley radius and the valley obliquity of the three substrates are listed in Table 1. To study the effect of the substrate surface roughness on the ultra thin DLC film growth, especially the substrate is not atomically smooth; DLC films deposited on Si substrates with the three different initial roughnesses were experimentally investigated.

Firstly, DLC film deposited on silicon substrate with a low initial roughness was tested. Fig. 4(A) shows the surface roughness evolution as a function of the DLC film thickness for Si substrate with an initial roughness of 0.15 nm. The surface roughness decreases from 0.14 to 0.10 nm, while the film thickness increases from 1 to 20 nm. The typical surface morphology of silicon substrate and DLC films with thickness of 2, 6 and 20 nm were shown in Fig. 4(B). The scan size is 1 μm and the vertical scale is 2 nm. It can be seen that the hills are merging to each other at the bottom of the valleys as the deposition progresses. This result

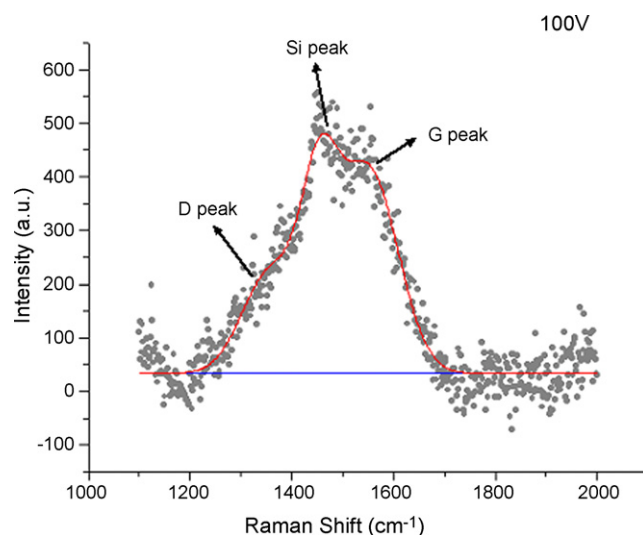


Fig. 2. Raman spectra of DLC film.

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