



Si seed layer thickness effect on the structure of ultrathin tetrahedral amorphous carbon films

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ABSTRACT

Tetrahedral amorphous carbon (ta-C) film is a promising materials serving as a protective coating layer due to its high sp³ content and its thickness up to 1–2 nm. In order to improve the adhesion of ta-C on metal, a Si film is usually pre-coated as seed layer. However, the Si film will increase the total thickness of protective layer. How to reduce the thickness of Si film is a requirement of industry applications. Here, we have studied the Si film thickness effect on the structure of ultrathin ta-C film. In our experiment, the thickness of Si film varies from 5 to 30 Å while the thickness of ta-C film is kept at about 2 nm. The Raman result shows that the sp³ fraction of ta-C film with pre-coated 5 Å Si film is a little lower than that of ta-C film without Si pre-coated. When the Si film becomes thicker from 5 to 30 Å, the sp³ fraction of Si/ta-C composite films is almost no changed. The XPS analysis shows that a part of Si bonding changes to SiC bonding which can enhance the adhesion of ta-C on metal. It also confirms that the SiC bonding fraction of Si film has no change when the thickness of Si film increases from 5 to 30 Å, which is consistent with the Raman analysis. Thus, an indication is that the fundamental limitation thickness of Si seed layer can be reduced to 5 Å.

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1. Introduction

At present, the demand for developing ultrathin and high-performance dielectric coating has rapidly grown, especially in the fields of plasmonics and magnetic disk drive technology [1–10]. Plasmonics is a flourishing field of science and technology that exploits the unique optical properties of metal nanostructures to manipulate light at a nanoscale [8,9]. In plasmonics, ultrathin dielectric layer coated metallic nanostructures is of much interest due to the following reasons. Firstly, compared with bare metallic nanostructures, an ultrathin dielectric layer coating can keep the metallic nanoparticles from agglomerating, separating them from a direct contact with photonic processes [1,4]. Secondly, dielectric layers can be functionalized to fulfill special requirements, for example, chemical stability [6,7]. However, a common drawback in the system of dielectric layer coated metallic substrates is that the strongest plasmonic electric field usually becomes trapped within the dielectric layer, and the electric field intensity outside the dielectric layer becomes attenuated as compared with that of bare metal nanostructures, especially under a thicker dielectric layer coating [1,2,7]. In this regard, how to release the strongest plasmonic electric field trapped inside

the dielectric layer to its outside surface is of tremendous scientific and technical importance. Compared with electron tunneling phenomena, if the dielectric coating is ultrathin (up to angstrom level), it cannot confine this field wholly inside the dielectric layer and the photons can spill over from the inside of the dielectric layer [2,3].

Among the various dielectric materials, diamond-like carbon (DLC) film becomes a promising material as ultrathin dielectric coating film [2,3,10,11]. As a type of DLC films, tetrahedral amorphous carbon (ta-C) film with sp³ bonding up to 90% in volume fraction and a thickness down to 1–2 nm can preserve its extraordinary mechanical properties, thus usually serving as an excellent protective coating layer [10–16]. The ta-C film, as an ultrathin dielectric coating, has many other desirable properties, such as pinhole-free, chemical inertness, thermal stability, biocompatibility, and high electrical resistance [17–19]. Additionally, ta-C film is atomically smooth and generally takes on the roughness of the substrate on which it is deposited, which is highly desirable for many optical applications [20]. Thus, ultrathin ta-C film is a promising coating in plasmonics [2,3].

However, direct deposition of ta-C films on metal has a drawback that the adhesion between the ta-C and metal is very weak. Thus, a thin Si film is often pre-coated as seed layer to improve the adhesion between the ta-C and metal substrate [21–24]. But the Si film will increase the total thickness of protective layer. How to reduce the thickness of Si film is a requirement of industry applications [2].

In addition, in magnetic disk drive technology, the reduction for the Si/ta-C thickness is more and more necessary due to higher data

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recording density. However, the ta-C film with thickness up to 1–2 nm is very difficult to be further reduced at present. Thus, the Si seed layer limitation thickness should be studied well.

Here, we have studied the Si thickness effect on the structure evolution of ultrathin ta-C film by Raman spectroscopy. In the experiment, the thickness of Si films varies from 5 to 30 Å while the thickness of ta-C films is kept at about 2 nm. The interface analysis by XPS between the Si seed layer and ta-C film was also performed. It will allow us to investigate the fundamental limitation thickness of Si seed layer. As far as we know, such ultrathin Si seed layer on the ta-C structure has not reported up to now.

2. Experimental

Firstly, the AlTiC (Al_2O_3 :64%; TiC: 36%) substrates were cleaned by RF-plasma etching using Ar ion plasma before coating. And then, a thin Si seed layer was coated on the substrate by DC magnetron sputtering using Ar ion plasma and thickness varying from 5 to 30 Å. Lastly, the ta-C films were deposited in a multi-film deposition system combining a FCVA (filtered cathodic vacuum arc) gun [10–12]. During the coating process, the substrate holder was rotated for better uniformity and ion beam was normal to the substrates. The deposition rate was ~ 0.5 Å per second and monitored by ellipsometer, and the thickness of ta-C film was kept at 20 Å.

Raman measurements were performed with a Renishaw inVia Raman spectrometer at 514 nm in backscattering geometry. The 514 nm laser output of 20 mW and a 50 X objective lens was used, which resulted in an incident power at the sample of approximately 3 mW. The line focus in Raman measurement was performed to avoid sample degradation. All the spectra were corrected by subtracting the system response signal obtained by measuring a background spectrum. The spectral resolution obtained in Raman spectroscopy was approximately 1 cm^{-1} . The Si and C bonding analysis by X-ray Photoelectron Spectroscopy (XPS) were performed on a Shimadzu ESCA 3600 system with an Mg Ka (1253.6 eV) source and the bonding energy (84.0 eV) of Au 4f_{2/7} as calibration. The Si/ta-C composite film was also investigated using a transmission electron microscope (TEM, JEOL, JEM 2100 F) cross-sectional analysis. The specimen was thinned by dual beam focused ion beam (FIB).

3. Results and discussion

Raman spectroscopy was widely used to characterize the structure evolution of ta-C films. Here, we determined the 514 nm Raman spectra for the Si/ta-C composite films. Fig. 1 showed the Raman spectra between the 900 cm^{-1} and 2000 cm^{-1} for the 20 Å ta-C films with the pre-coated Si seed layer thickness varying from 0 to 30 Å, respectively.

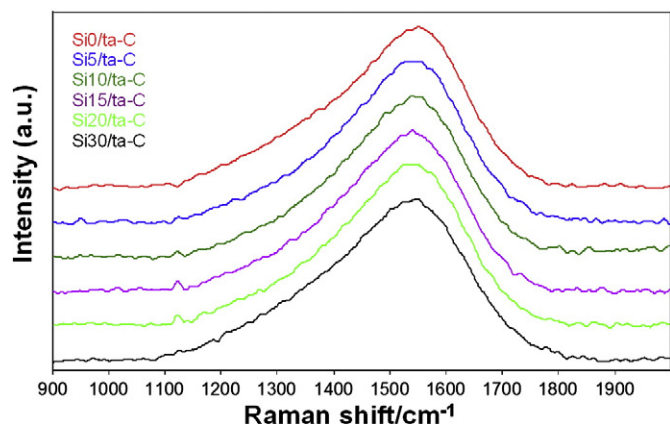


Fig. 1. 514 nm Raman spectra of Si/ta-C composite films. Si thickness is varied from 0 to 30 Å and the thickness of ta-C layer is kept to be 20 Å for all the samples.

The spectra have been displaced vertically for clarity, but otherwise are displayed using the same vertical scale. These visible Raman spectra of ta-C films exhibited one main feature, a broad almost symmetric Raman intensity distribution in the range $1100\text{--}1800\text{ cm}^{-1}$ centered around 1550 cm^{-1} , which included the bonding information of sp^2 and sp^3 sites [13,25–28]. This confirmed that the fabricated ultrathin ta-C films were amorphous [25–28]. With careful inspection of these Raman spectra, the peak position at around 1550 cm^{-1} shifted to a little lower wavenumber when the thickness of Si film increased from 0 to 5 Å. However, by further increasing of Si film thickness from 5 to 30 Å, the peak position at 1550 cm^{-1} seemed to be almost not changed.

For the further detailed information, these visible Raman spectra were fitted to two peaks using a mixture of Gaussian and Lorentzian profiles. For consistency with published literature, we defined these two peaks as the graphite (“G”) and disorder (“D”) peaks [25]. As it was well known, at the 514 nm excitation, the sp^2 sites had such a high cross-section that they dominate the spectra, the sp^3 sites were invisible and the spectrum responded only to the configuration or order of the sp^2 sites. So the G and D peaks were due to sp^2 sites [25–28]. The G peak was due to the bond stretching of all pairs of sp^2 atoms in both rings and chains. The D peak was due to the breathing modes of sp^2 atoms in rings, induced by disorder-activated aromatic modes of A1 symmetry [28]. Fig. 2 showed the parameters about G and D peak extracted from the Raman spectra as a function of the thickness of Si seed layer varying from 0 to 30 Å for the composite Si/ta-C films. The intensity ratio of I_d/I_g was generally a measure for zone edges or border phonons of the carbon clusters, which depended on cluster sizes and distributions. The increase in the D peak content was an indication of the state of development of the sp^2 phase and suggested that the sp^2 sites were beginning to organize into small graphitic clusters [28]. Thus, the increase of I_d/I_g ratio indicated a decrease of the sp^3 fraction. The G peak position was also related to the sp^3 fraction. When the sp^3 fraction increased, the sp^2 fraction and sp^2 cluster size would decrease, and then the bond order increased with a resultant increase in the Raman frequency, which meant that the upper shift of G peak position corresponded to higher sp^3 content [25–28]. Thus, the trend of I_d/I_g vibration was

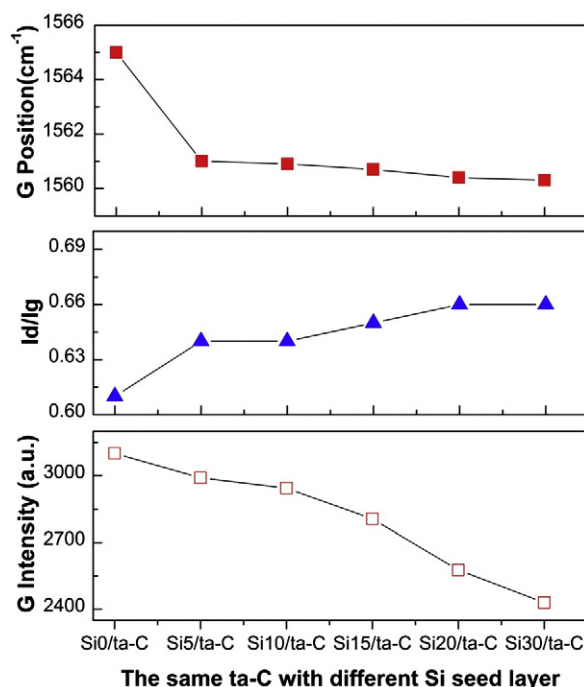


Fig. 2. Variation of G position, I_d/I_g and G intensity with the Si seed layer thickness varying from 0 to 30 Å.

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