Fractal characteristics of surface morphology for hydrogenated silicon films*

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Hydrogenated nanocrystalline silicon films (nc-Si:H) are a developing novel man-made functional semiconductor material in recent years. Their strange microstructure and physical properties of nc-Si:H films are attracting our attention. We have used the conventional structure analysis procedures, such as X-ray diffraction pattern, Raman scattering spectrum, high resolution transmission electron microscopy (HREM), and the scanning tunnelling microscope (STM), to analyze their microstructure[1, 2]. Since Mandelbrot proposed the fractal theory in 1982[3, 4], it has become a quantitatively characterizing means to distinguish the microstructure morphology of materials. Recently, the fractal formation in a-Ge/metal and a-Si/metal bilayer films was studied by Wu and Bo Bian *et al.*[5] They found that the dendrite patterns of Si and Ge are indeed fractals and suggested a randomly successive nucleation mechanism to explain the fractal formation.

In this note, we study the morphology of surface microstructure of silicon films by using STM technology in the range of nanometer scale and the fractal dimension D of surface morphology of samples is calculated with the fractal theory[6]. The silicon film samples (μc-Si:H, nc-Si:H, pc-Si:H) are deposited in a conventional ultrahigh vacuum plasma-enhanced chemical vapour deposition (PECVD) system[1]. The results lead us to the conclusion that the surface morphology of silicon films exhibits fractal feature at nanometers. It is interesting to note that there is a maximum fractal dimension D when the silicon films varied into nanocrystalline phase (nc-Si:H).

1 Experimental procedure

The silicon films, used in our experiment, were deposited in an ultrahigh vacuum

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PECVD system. The technological parameters were selected under the conditions of deposition temperature T_s=250—300℃, r.f. power 60—80 W, and the total pressure of reactive gases in the deposition process of 100 — 150 Pa. There are two main technological factors which are crucial in forming the micrograins in the growing films. One is the percentage content of saline, C=SiH_4/SiH_4+H_2 and the other is negative bias-V_b applied to the parallel electrodes of the deposition system[1]. The content of micrograins in the growing films is defined by the volume fraction ratio of grains X_c, which may be evaluated from the Raman scattering spectrum. Therefore, according to X_c values we may classify the silicon films into three kinds: the μc-Si: H (X_c<47%), nc-Si: H (X_c=48%−58%) and pc-Si: H (X_c>60%) films. The mean grain size d of experimental samples was 3— 6 nm, which was estimated from the peak value of Raman scattering spectrum. The thickness of these samples was 1 μm and the substrates were glass and quartz.

The morphology of surface microstructure was observed with STM images. The STM equipment produced by the Institute of Chemistry, Chinese Academy of Sciences, was operated in the air at room temperature. Tips were prepared by mechanically shearing platinum-iridium (Pt:Ir=88:12) alloy wire (0.01 mm in diameter). Images were performed in the constant-current mode with positive bias voltages at 1.50 V and a tunnelling set-point current of 0.2 nA. The fluctuation of the morphology of sample surface can be reflected by the variation of gray scale in view of observation. Before observation, samples were etched by dilute HF in order to eliminate the thinner oxide layer on surface.

The morphological outline of sample surface could be obtained by using image processing of surface microstructure with image software and by calculating the fractal dimension D from these morphological outlines. Moreover, there are many methods to calculate the fractal dimension D from these morphological outlines[7]; we select the Fourier analysis method to calculate the D values for the silicon film surface morphology. The actual procedures are as follows:

(i) Using the image software to deal with the observed morphology picture of sample surface, which has been done by STM technology, may obtain the morphological outline on different positions of sample surface.

(ii) Then, we do the Fourier transformation for these observed outlines, and get the correlative powerful spectrum.

(iii) In order to decline the image processing errors, we make the powerful spectrum average.

(iv) Integrate the average powerful spectrum from high-frequency terminal to lower-frequency terminal.

(v) Draw the powerful spectrum curve on double logarithm coordinate after

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integration, and calculate the slope m from the linear part of the curve.

So the fractal dimension D of the surface morphology of silicon films can be obtained from the following formula:

$$D=2.0+m/2.$$

The fractal demension D obtained in this procedure can be regarded as a measure of random surface roughness. It can be used to quantatively describe the fractal characteristics of sample surface morphology outline curve. This computation program has been written in C language and completed at AST-486 personal computer.

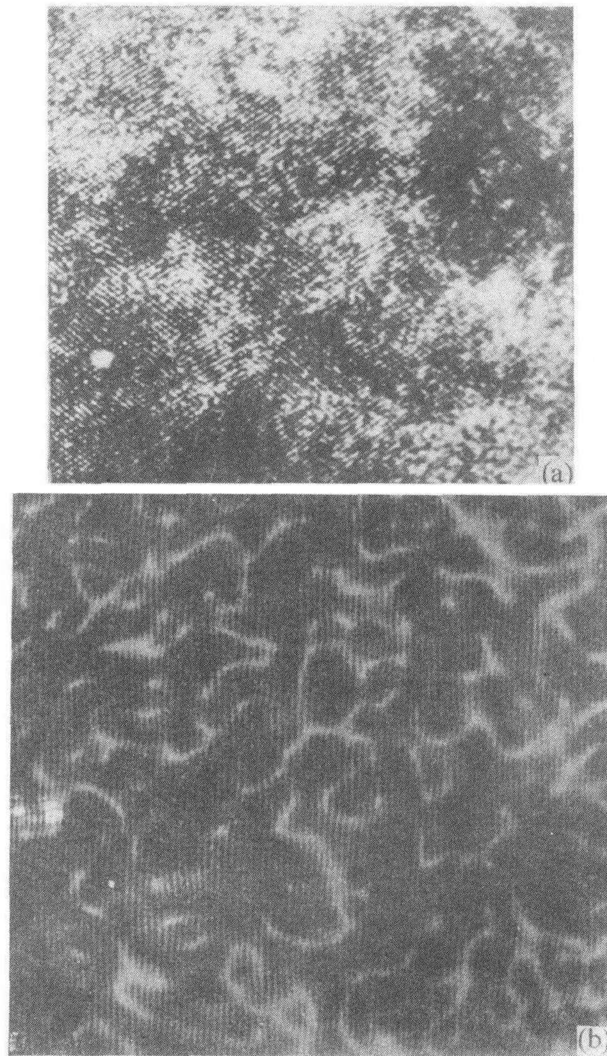

Fig. 1. HREM picture of nc-Si:H films. (a) HREM image of nc-Si:H film; (b) Fourier-filtered image of fig. 1(a), revealing a lot of interfaces' texture.

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2 Experimental results

Figure 1 is a typical HREM picture of nc-Si: H film, which reveals a novel microstructure of silicon materials. Fig. 1(a) is an original HREM picture and fig. 1(b) is the picture after image processing. We can see from fig. 1 that there are a great number of randomly arranged micrograins, with the size about 3—6 nm measured by Raman scattering spectrum. Between grains there is an interface regime, its thickness being $l<1$ nm.

Figûre 2 is the microstructure morphology of nc-Si:H film surface with different X_c values, which was observed by STM technology. The scanned area was 400 nm × 400 nm. For every sample, we measured 10 different position outlines on the surface, and then made out the mean integrated powerful spectra by using Fourier analysis method. Finally, we drew these data into a double logarithm coordinate as shown in fig. 3. We can see from fig. 3 that there exists a very good linear regime in the middle part of the whole curve and the slope of line is $m=-1.362$. According to formula in ref. [3], we get the fractal dimension $D=1.319$. We found the correlative coefficient of linear simulation to be 0.993. This demonstrates that there really exist the fractal characteristics for the

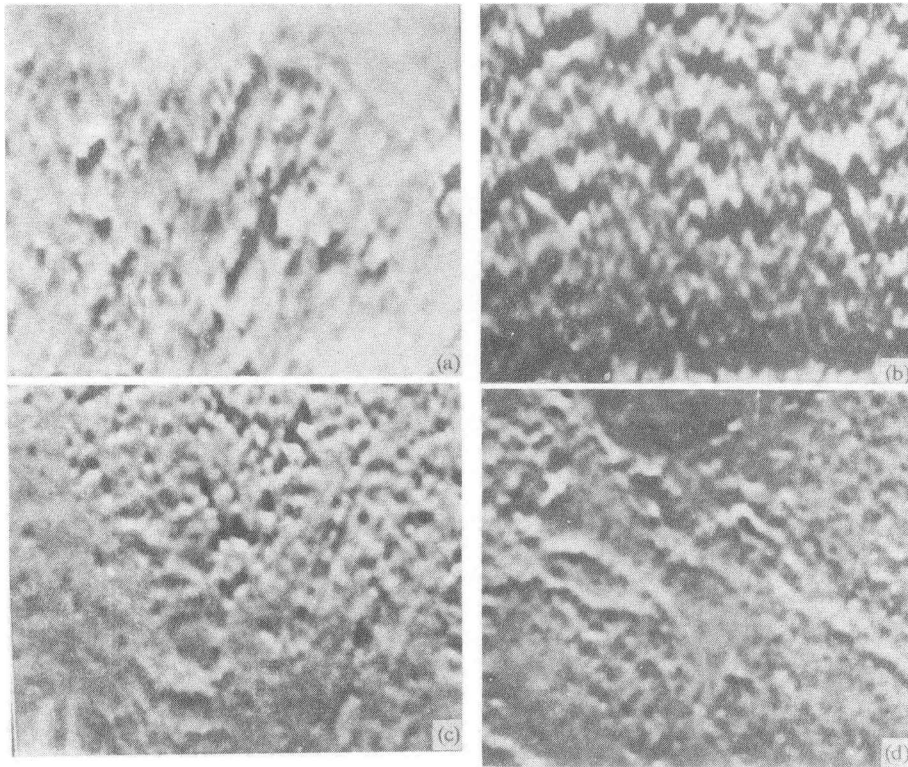

Fig. 2. Surface morphology of nc-Si:H films employed STM technology. (a) $X_c=0.51$, $D=1.43$; (b) $X_c=0.57$, $D=1.43$; (c) $X_c=0.66$, $D=1.33$; (d) $X_c=0.70$, $D=1.31$.

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micromorphology of film surface.

According to the mathematical theory of fractal, the self-similarity has an infinite texture for a set of patterns embedded in each other, for the physical texture. On the other hand, the physical substances will have the fractal characteristics only at a certain dimension region. This is evidence for the fact that there is a very good linear regime at the middle part of the curve in fig. 3; the corresponding dimension region is about 2— 6 nm.

The experimental data of fractal dimension D varying with the crystalline volume fraction X_c for the morphology of hydrogenated silicon film surface are scheduled in table 1 and figure 4.

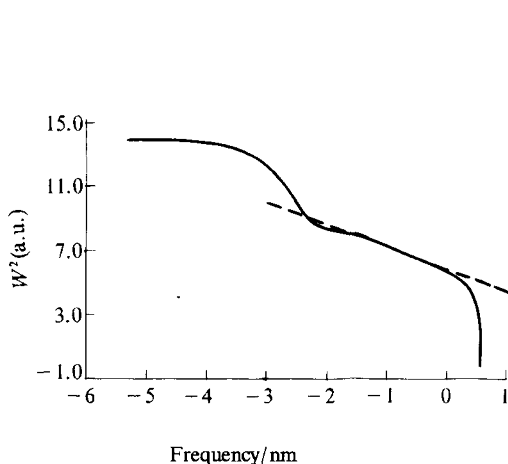

Fig. 3. A diagram of integrated powerful spectra of nc-Si:H film.

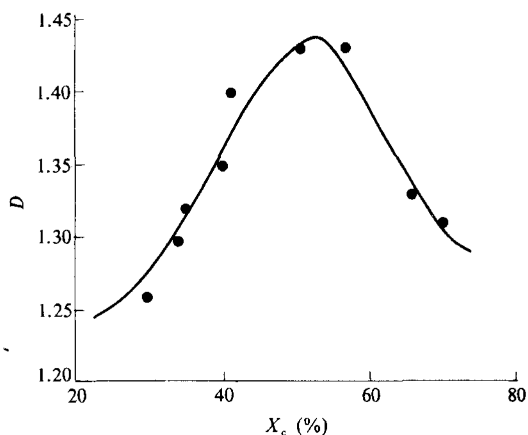

Fig. 4. Fraction dimension D varies with X_c for a series of hydrogenated silicon films.

Table 1 D varies with X_c for a series of hydrogenated silicon films

X_c (%)	29.4	33.4	35.0	40.0	41.3	51.0	57.0	66.0	70.0
D	1.26	1.30	1.32	1.35	1.40	1.43	1.43	1.33	1.31

3 Discussion

As mentioned above, the fractal dimension D used to quantitatively describe the morphology fractal characteristics of film surface has certain random process of itself for the degree of magnanimity of random surface roughness. In order to study the relationship between the morphology fractal dimension D and the related microstructure parameters, such as X_c, of hydrogenated silicon films, we employed the STM technology to observe the morphologies of different positions of film surface. We may get several fractal dimensions for every microscopy morphologies' pictures of film surface. Because the actual fractal dimensions of film surface could not be less than any of the measured D values, we may take the largest of measured D values as the actual fractal dimension of measured samples.

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Then, a series of measured D values varying with crystalline volume fraction ratio X_c of hydrogenated silicon films are given in table 1 and figure 4.

Mitchell has suggested[6] that in the case of calculating the fractal dimension D by employing the Fourier transformation method, the precision of Fourier transformation is the main factor to decide the errors. Owing to the precision of Fourier transformation is closely related to the numbers of measured data which were used to describe the outline for film surface, if the data are taken much less, then the errors of measured fractal dimension D are much greater. When we take the measured data as 100—200, the errors of D values may be higher than 35%. However, in our calculation, we take the data value of describing the outline for film surface as 1600, so the error should be less than 3%. It can be seen that the results scheduled in table 1 can fully explain that the fractal dimension D revealed a maximum value as the crystalline volume fraction ratio X_c entered into nanophase ($X_c=0.51—0.57$) for hydrogenated silicon films. This is an important discovery for this note.

We can see from fig. 4 that the D values monotonously decreased with X_c decreasing as the crystalline volume fraction of hydrogenated silicon films $X_c<0.50$. As extending $X_c \to 0$, we get $D=1.20$ for a-Si:H film. On the other hand, as $X_c>0.57$, the D values suddenly dropped. This reveals a maximum peak of D at $X_c=0.54$ in the curve of fig. 4. This is another special characteristic of nc-Si:H film observed from the fractal texture.

This review suggests that the fractal dimension D of film-surface morphology is a degree of magnanimity of random surface roughness. Moreover, the D value of a-Si:H film ($D=1.20$) is less than that of nc-Si:H ($D=1.43$). This fact presents a question: Although the amorphous silicon film (a-SiH) has a complete disorder morphology on its whole structure, it can exhibit a more even surface morphology than that of nanocrystalline silicon film (nc-Si:H) in a certain small region, such as the nm-order sized. Fig. 2(b) shows that a film possesses maximum values of $X_c=0.57$ and $D=1.43$, but its surface morphology reveals most roughness in all of the pictures. In our previous works, we have suggested that the nc-Si:H film contained a large amount of interface texture ($X_i \approx 50\%$). Therefore, there existed an outstanding complexity of film-surface morphology. As X_c values continuously increased, the content of interfaces decreased. Undoubtedly, this is the main reason for D values dropping down over from $X_c>0.57$. Compared with our previous works, this note developed the idea that the hydrogen content C_H is maximum[8] and the coefficient of piezo-resistance coefficient k is the largest as $X_c \approx 50\%$[9]. All of these reflect the special characteristic of nanocrystalline silicon films.

4 Conclusion

In this note, we suggest that there is an extremely powerful means to study the

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fractal characteristic of film-surface morphology by using the fractal theory and STM technology. The surface morphology of hydrogenated silicon films has a gross fractral characteristic and the fractal dimension D reveals a maximum value in its nanophase state. All of this fully reflects the special structural characteristic of nc-Si:H films. We consider that there is a functional relationship between fractal dimension D and structure parameter X_c, if we make a further investigation on this title, such as $X_c=f(D)$ or $D=f'(X_c)$.

References

1 He Yuliang, Liu Xiangna, Wang Zhichao *et al.*, Study of nano-crystalline silicon films, *Science in China*, Ser. A, 1993, 36(2): 248.

2 He Yuliang, Yin Chenzheng, Cheng Guangxu *et al.*, The structure and properties of nanosize crystalline silicon films, *J. Appl. Phys.*, 1994, 72(2):797.

3 Mandelbrot, B. *Fractal Geometry of Nature*, San Francisco: CA, 1982.

4 Mandelbrot, B., Passoja, D. E., Paullay, Alvin. J., Fractal character of fracture surface of metals, *Nature*, 1989, 308:721.

5 Bo Bian, Jian Yie, Li Boquan *et al.*, Fractal formation in a-Si:H/Ag/a-Si:H films after annealing, *J. Apl. Phys.*, 1993, 73(11):7402.

6 Feder, J., *Fractals*, New York: Plenum Press, 1988.

7 Morgan, W. M., Down, D. B., Quantitative topographic analysis of fractal surfaces by STM, *J. Mater Res.*, 1990, 5(10):2344.

8 Yu Minbin, He Yuliang, Liu Hongtao *et al.*, Annealing characteristics of nc-Si:H films, *Acta Phys. Sinica* (in Chinese), 1990, 39(11):1796.

9 He Yuliang, Wu Xuehui, Lin Hongyi *et al.*, Structure characteristics and piezoresistive effect of nc-Si:H films, *Chinese Science Bulletin*, 1995, 40(20):1684.

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