Multiscaling in semiconductor nanowire growth

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The scaling behavior of diameter modulations in semiconductor nanowire growth was investigated. SiC nanowires with modulated diameter were grown via a self-organized process and examined by transmission electron microscopy. By calculating qth-order height-height correlation functions, we found multiaffinity.

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Since the concept of multifractality was extended to self-affine fractals for more suitable description of complex shapes and profiles [1,2], many theoretical works on multi-affinity have been reported so far [3–7]. Nevertheless, to our best knowledge, experimental observation of apparent multi-affinity in physical system is very limited: Myllys *et al.* [8] analyzed kinetic roughening of slow combustion fronts in paper and found spatial and temporal multiscaling at short scales.

Semiconductor nanowires grown via self-organized process sometimes show diameter modulations. Some of them are simply periodic [9], but some exhibit complex and non-periodic fluctuations [10,11]. In this Brief Report, we inspect two types of diameter modulations of SiC nanowires grown via a self-organized process in terms of scaling. We show that one type of the diameter modulations can be interpreted as multiaffine.

The growth procedure of the SiC nanowires is as follows: a (111) Si substrate (P-doped, 1000–3000 Ω cm) on which 7.5-nm-thick Au (99.95%) was deposited and a SiC powder (98.7%) were sealed in an evacuated silica container (10^{-5} torr) and heated to 1200 °C, and then cooled to room temperature. Source gas was formed at 1200 °C and the supersaturation during the cooling was the driving force of the crystal growth. SiC nanowires grown on the Si substrate were dispersed in ethanol by ultrasonic treatment, and then dropped on a microgrid for transmission electron microscopy (TEM) observations (JEOL, JEM-2000EX). Before TEM observations of the nanowires, any chemical or physical treatment has not been performed to remove homogeneous surface oxide of a few nanometers thick. Thus the diameters include the thickness of surface oxide. We speculate that a Au-containing droplet worked as a nucleation and growth site of a nanowire. Thus the growth mechanism of the nanowires is considered to be the vapor-liquid-solid (VLS) mechanism, where source materials are in gas phase, which are captured by a molten droplet, then supersaturated atoms in the droplet crystallize at the liquid-solid interface forming a wirelike crystal. In this process, the diameter of a wire is defined by the diameter of the liquid-solid interface.

In Fig. 1(a), we show a TEM image of a SiC nanowire which shows asymmetric abrupt changes in diameter and clear facets. The plane indices of the facets have not been revealed and are under investigation at present. We call this type of modulation type A. No catalytic droplet was found at the ends of the wire presumably because the wire was broken by the ultrasonic treatment. Thus the growth direction could not be decided. Figure 1(b) is its digitized profile with 0.94 nm resolution. The number of the data was 771. The mean diameter was 43.2 nm with the standard deviation of 4.6 nm. Detail of the multiscaling analysis is as follows. The digitized diameter modulation is defined as the set of diameters d_i at equally spaced points x_i where i is the index of discrete sampling points. In order to reveal scaling behavior of the diameter modulation, the qth-order height-height correlation function

$$C_q(i) = \langle |d_{j+i} - d_j|^q \rangle \tag{1}$$

was calculated, where angular brackets denote averaging over j. If d_i is scaling, we expect a power-law behavior at the limit of small i as follows:

$$C_q(i) \sim i^{q\alpha_q}, \tag{2}$$

where α_q is the generalized qth-order roughness (or Hurst) exponent. If the profile is monoaffine, α_q is independent of q. On the other hand, if the profile is monoaffine, α_q varies with q [1,2]. In Fig. 2, we show the behavior of the qth root of the qth-order height-height correlation function with $q = 1, 2, \ldots, 20$. Apparently the slopes of the curves vary with q for small i, while for large i, nonmultiscaling appears. The curves are sufficiently straight at around $0.6 < \ln i < 2$, thus the slopes were determined at this region to obtain α_q. The inset in Fig. 2 shows q dependency of α_q. The qth-order roughness exponent decreases monotonically with increasing q and approaches around 0.37. Accordingly, the diameter modulation can be interpreted as multiaffine. We also analyzed several similar nanowires which show abrupt changes and facets, and confirmed that they showed multiaffinity.

It is reported that vertical discontinuities in a self-affine profile can lead to multiaffinity [7]. As mentioned, the type-A nanowires have abrupt changes in diameter. Accordingly, the abrupt changes are considered to be a source of the multiaffinity in the diameter modulation. To confirm this, we also examined another type of diameter modulation (type B). In Fig. 3(a), we show a TEM image of a SiC nanowire which does not show abrupt changes in diameter nor clear facets. Figure 3(b) is its digitized profile with the same resolution of 0.94 nm and the number of the data is 1604. The mean diameter was 59.1 nm with the standard deviation of 7.8 nm. As shown in Fig. 4, the height-height correlation functions show scaling behavior; however, α_q does not show large change but nearly constant at around 0.8–0.9. Thus the di-

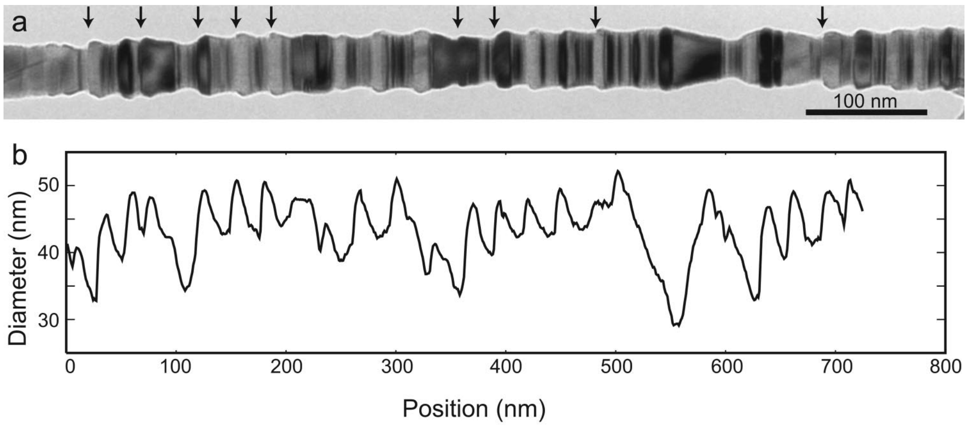

FIG. 1. (a) TEM image of a SiC nanowire showing abrupt changes in diameter (marked by arrows) and clear facets. (b) Digitized diameter profile.

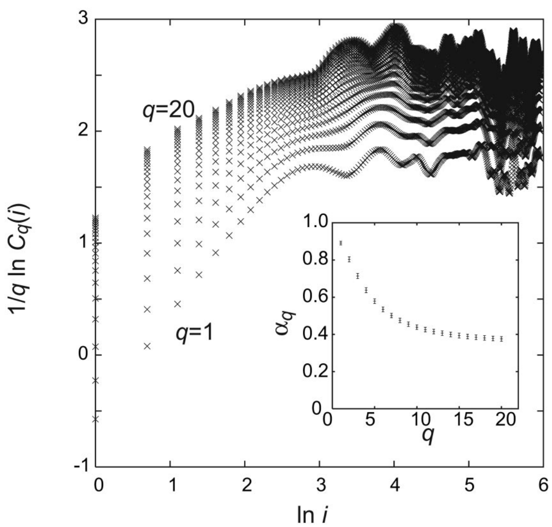

FIG. 2. The qth-order height-height correlation functions of the profile of Fig. 1(b) with $q=1,2,\ldots,20$. Inset: The q dependency of α_q.

ameter modulation of this wire is considered to be not multiaffine but monoaffine. This result on the type-B nanowire also confirms that the multiaffinity of the diameter modulation of type-A nanowires is not an artifact due to digitizing error when converting the TEM image to discrete data, since the same procedure was applied.

In our previous paper [12], we reported that the increments of the diameter modulation of a type-A SiC nanowire had an asymmetric power-law distribution. In other words, the distribution of increments of the type-A nanowire had a fat tail. In addition, in the previous paper, the origin of the asymmetric modulation was tentatively attributed to the stick-slip motion of the catalytic droplet at the end of the nanowire; however, we have no solid evidence for this. We also checked the distributions of increments of the diameter modulations of the nanowires shown in Fig. 1 (type A) and Fig. 3 (type B): the type-A nanowire exhibited an asymmetric power-law distribution for large increments (<-2 nm per a 0.94-nm step) while the type-B nanowire exhibited a Gaussian distribution (see Fig. 5). We think that the power-law nature of the increments of the type-A nanowire is due to the abrupt changes in diameter, since abrupt changes can yield large increments. We speculate that the difference in increments distribution also contributed the difference in fractality since other multiaffine systems also exhibited power-law distributions [6,8].

So far, the origin of the diameter modulations has not been revealed. At this moment, we speculate that wetting property of catalytic molten droplets is related to the complex modulations similarly to reported periodic modulations

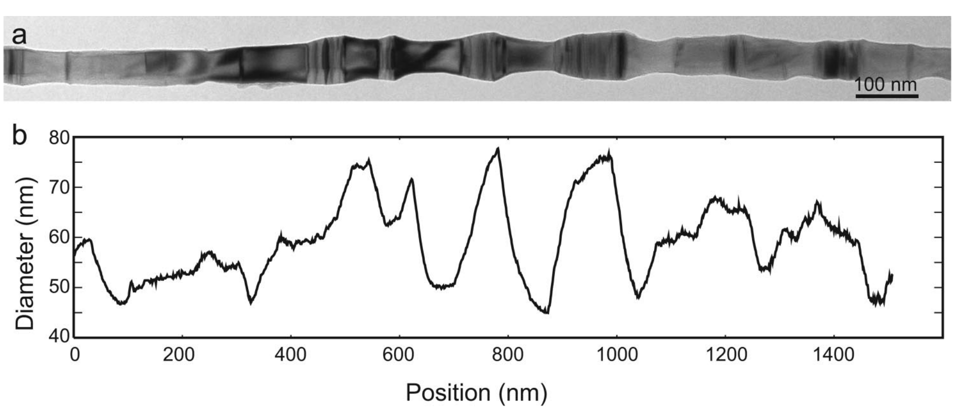

FIG. 3. (a) TEM image of a SiC nanowire of type B which shows no abrupt changes in diameter. (b) Digitized diameter profile.

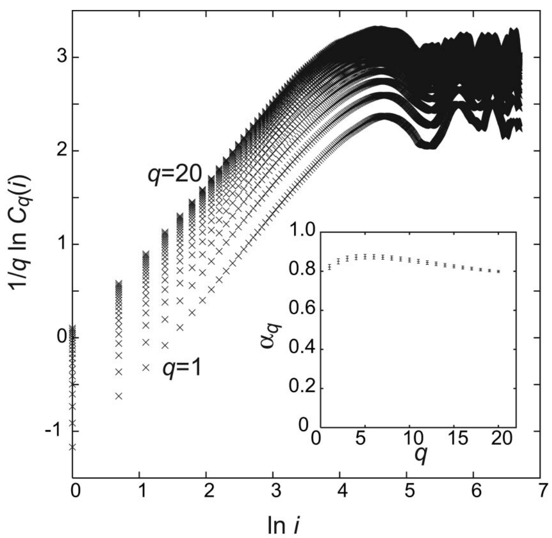

FIG. 4. The qth-order height-height correlation functions of the profile of Fig. 3(b) with $q=1,2,\ldots,20$. Inset: The q dependency of α_q.

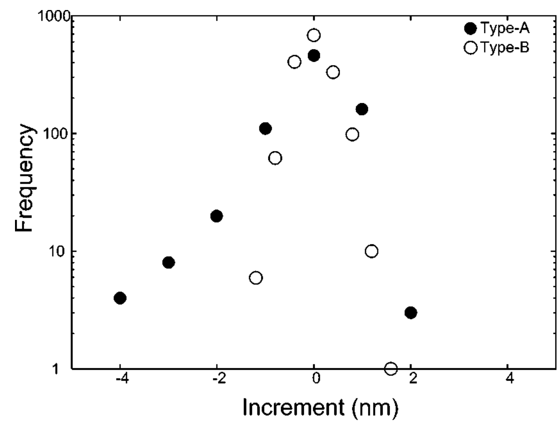

FIG. 5. Probability distributions of the diameter increments per an 0.94-nm step for the type-A and type-B nanowires.

[9,13,14]. It is also reported that periodic diameter modulation was sensitive to additive impurity metals to Au catalyst [15]. Although the SiC nanowires of both type A and type B were grown on the same substrate, it is very likely that sort and/or amount of impurities involved in the catalytic Au droplets were different and as a result the wetting properties were affected differently. Further investigation is under way to find catalytic particles and analyze their compositions.

In conclusion, we have found multiaffinity in SiC nanowire growth. We speculate that the abrupt changes of the diameter are a source of the multiaffinity. It is very interesting that such complex fractality appears in nanometer scale. At present, details of the nonlinear behavior of α_q remain matter of investigation. Furthermore, dependency of the behavior of α_q on the growth conditions is also an open question. Recently, much effort has been focused on electron transport properties of various nanowires with smooth surface [16,17]. In addition, interesting dielectric properties have been found in fractal materials [18,19]. We expect the multiaffine nanowires will exhibit fascinating transport property and dielectric response.

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