Spontaneously Emergent Ripples —Pattern Formation by Ion Beam Sputter-Etching and Wind—

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Spontaneously emergent ripples on fused silica surface under homogeneous oblique incidence of Ar ions accelerated up to 4.25 keV are reintroduced together with increasing recent related studies in theories and experiments for variety of combinations of ions and materials. The features of the ripple formation are discussed with an interest in similarity with ripples formed on sand by the wind.

Key words: Ripples, Ion Beam Sputter-Etching, Wind, Pattern Formation

1. Introduction

Spontaneously emergent ripples are often found in nature such as sand ripples formed by wind and ripples of clouds formed between cold dry upper atmospheric layer and warm moist lower layer. Recently, this spontaneous formation of ripples is drawing attention as new means of "nanotechnology" to overcome the minimum size limitation of artificial micro-fabrication and the maximum processible area limitation which poses impediment on reduction of process cost (Valbusa *et al.*, 2002). Here, the latter ripple formation is reintroduced with an interest in similarity with the sand ripple by wind.

2. An Example of Experimental Evidence of Spontaneous Formation of Ripples by Off-normal Low Energy Ion Beam Sputter-Etching

Low energy ion beam sputter-etching of flat, smooth and homogeneous solid targets often leads to spontaneous formation of characteristic erosion patterns as shown in Fig. 1 (Motohiro and Taga, 1987). In Fig. 1, at off-normal ion beam incidence $\eta = 45^{\circ}$ with respect to the surface normal, ripples emerged with their wave vectors in parallel with the ion beam azimuth. The foreboding sign of the ripple formation can be also found at $\eta = 22.5^{\circ}$ and even 0° indicating the ripple did not emerge abruptly at $\eta = 45^{\circ}$. Figure 2 displays SEM images of the ripples at different Ar⁺ ion doses J $[C/cm^{2}]$ in different magnifications. In contrast with the cases at $\eta = 0^{\circ}$ and 22.5°, it is indicated that the ripples emergedhomogeneously even at the initial stage and coarsened with J keeping the similar form. The average wavelengths of the ripples λ measured from the images in Fig. 2 were plotted against J as shown in Fig. 3. It was found that λ increased in the proportion of the 1.68th power of J. This tendency has been repeatedly reported in the later works until recently. At an appropriate J, the sample surface showed a rainbow colors indicating the ripples functioning as an optical grating in the visible wavelength range.

3. Spontaneous Formation of Sand Ripples by Wind

The physics of formation of sand ripples had been paid attention to by very limited number of researchers until recently. The essential features of the sand ripple formation can be reproduced by a simple cellular automaton model in which the following two events (A) and (B) take place repeatedly. (A) A grain of sand in a cell at a height H measured from the average level of the sand surface was driven laterally down the wind to a cell at a distance L which is determined in proportion to the height H of the initial cell and the strength of the wind U. (B) The sands in the cells with relatively higher sand level move to the neighboring cells with lower sand level at a rate in proportion to the height difference Δ H and diffusivity of the sand D (Nishimori and Ouchi, 1993).

Figure 4 gives two examples of reproduced simulation of sand ripple formation by present authors in 1994 based on the above Nishimori's model. Obviously, Fig. 4(a) is a typical result in which the promotive event of the ripple formation (A) is overwhelmed by the planarization event (B), and Fig. 4(b) vice versa.

4. Basic Understanding of Ripple Formation Mechanism by Ion Beam Sputter-Etching

A continuum theory proposed by Bradley and Harper (1988) has been the most popularly cited one to explain the essential aspect of this ripple formation by ion beam sputter-etching. Figure 5 tries to explain the essence of the BH-theory intuitively.

Suppose ions A–F coming into a target material along the dotted lines deposit energy in droplet-like regions a–f inside the target. If these energy deposited regions are located near the surface like X and Y portions of the concave region, the surface materials are eroded away. However, in the convex region, the V and W portions are located away from the energy deposited region d and f, therefore, surface erosion does not take place. This is a promoting event (A)



Fig. 1. Scanning electron microscope (SEM) images and corresponding schematic diagrams of erosion patterns caused by normal ($\eta = 0^{\circ}$) and off-normal ($\eta = 22.5^{\circ}, 45^{\circ}, 67.5^{\circ}$) 4.25 keV Ar⁺ ion beam sputter-etching of fused silica targets with initially smooth planar surfaces. The angles of the observed facets in the schematic diagrams were obtained by tilting the sample under SEM observation.



Fig. 2. Coarsening of ripples at $\eta = 45^{\circ}$ with Ar⁺ ion doses J: (a) 0.36 C/cm²; (b) 0.23 C/cm²; (c) 0.14 C/cm²; (d) 0.09 C/cm². The white arrows indicate the azimuthal directions of the ion beam incidence. The black bar scales represent lengths of (a) 17.8 μ m, (b) 8.99 μ m, (c) 4.72 μ m, (d) 1.78 μ m, respectively.

for the ripple growth. However, since ion beam irradiated surfaces get high temperature and it promotes surface diffusion of atoms, reduction of concavity and convexity of the surface takes place. This is planarization event (B) for the ripple growth. Therefore, the ripple formation by ion beam sputter-etching shares essential features with the sand ripple formation although redoposition of sputtered materials are not taken into consideration.

5. Recent and Future Trend

Figure 6 shows the numbers of annual publications on the ripple formation by ion beam sputter-etching. They are increasing rabidly after 1994. Besides Ar^+ -SiO₂ system (Keller *et al.*, 2009), variety of combinations of ions and target materials such as metals, semiconductors and ionic crystals have been studied. Prevalence of scanning probe microscopy may have contributed to this as well as practical necessity in micro-fabrications and "nano-technology". For example, in fabrication of artificial nano-scale structures,



Fig. 3. Plots of measured average wavelength of the ripples λ against Ar^+ ion dose J.

focused ion beam machining plays a key role. However, spontaneous ripple formation can be an obstacle for fabrication of intended fine structures. Therefore, it is important to know the condition to reduce the spontaneous ripple for-



Fig. 4. Reproduction of sand ripples by computer simulation based on the Nishimori's model. (a) Corresponds to the case with week wind U and high diffusivity D, whereas (b) vice versa. The displayed area was divided into small cells. Darkness of the cells is given in proportion to the height H of the cell.



Fig. 5. Schematic diagram of the ion beam sputter-etching of a concave and a convex region of the target material.



Fig. 6. Recent rapid increase in number of annual publications on the ripple formation by ion beam sputter-etcing.

mation under the compromise between the promoting event for ripple formation (A) and the planarization event (B) as Fig. 4(a) in the case of sand ripples. On the other hand, it is often the case that the artificial periodic nanostructure can be precisely formed using nano-technology in a small area but cannot be extended to a large area in a costeffective manner. These periodic nanostructures have varieties of functions such as optical gratings and liquid crystal alignment if they can be extended in a macroscopic area. Spontaneous formation of periodic structure can offer costeffective substitutes for artificial precise nanostructure in large area with some irregularity but without losing their essential functions.

Theoretical works are continuously published pointing out incompleteness of the BH-theory and complexity of the phenomena (e.g. Munoz-Garcia *et al.*, 2008). Continuous publication of basic researches such as published in *Phys. Rev.* (PR) and *Phys. Rev. Lett.* (PRL) which are indicated in black blocks in Fig. 6 teaches that the phenomena involve attractive physics which will keep stimulating appetite of physical scientists.

The promotive event (A) for ripple formation is, in a sense, an autocatalytic phenomenon and the planarization event (B) is a kind of negative feedback caused by the

autocatalytically enhanced inhomogeneous structures. As shown in the sand ripple formation by wind and the ripple formation by off-normal low energy ion beam sputteretching, the inspection of similarities and differences both in the autocatalytic process (A) and the negative feedback process (B) involved in spontaneously emergent ripples often found in nature as well as in artificial systems offers varieties of problems to solve and this may cause those trends shown in Fig. 6.

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