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# Interface roughening in wrinkly metal

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## Abstract

We report a study on the roughening process in wrinkly metal film (aluminium) for thicknesses ranging from 35 to 1000 nm. The spatial and temporal scaling behaviours have been investigated by using atomic force microscopy. We show that fast diffusion is a part of the buckling process on a viscoelastic substrate due to Grinfeld-type instability. Power spectral density analysis reveals that the roughness exponent  $\alpha$  is  $\sim 0.85$  for all thicknesses. This value is consistent with the fact that fast diffusion is the underlying process. The films exhibit slow temporal evolution, i.e.  $W \sim t^\beta$  ( $\beta = 0.15 \pm 0.02$ ,  $Z = 5.66 \pm 0.5$ ), as the film thickness increases. The wavelength or correlation length also changes as  $\xi = t^{0.19 \pm 0.04}$  ( $Z = 5.26 \pm 1$ ) with the thickness. Deposition through a  $400 \mu\text{m} \times 1 \text{cm}$  (35 nm thickness) window shows a very organized wrinkled pattern with chain-like island attachment parallel to a surface with a smaller length scale ( $400 \mu\text{m}$ ). We propose a model explaining why deposited atoms would move parallel to a surface with a shorter length scale to create ordered chain-like structures.

## 1. Introduction

Spontaneous formation of complex structures on different substrates has become a subject of intensive experimental and theoretical study. Growing clusters and films often evolve into interesting self-similar and fractal patterns. Most of the studies available are on solid substrates; few studies were performed on complex structure formed on stretchable substrates. Bowden *et al* [1] first demonstrated how complex structures can be formed in thin metal films by thermal contraction of the substrate. Depositing metal films on thermally expanded polydimethylsiloxane (PDMS) and subsequently cooling them created wavy structures on the surface as a stress relief mechanism. Complex wave structures on polymer substrates have been studied by several groups [2–4] due to the wide scientific interest and potential applications. While most of the focuses are on the buckling of thin metal films, none of the previous studies address detail of the underlying surface processes. In this paper, we apply scaling laws [5–9] in an effort to understand roughening process during wrinkle formation for metal films for wide ranges of thicknesses. The growing surfaces naturally evolve into self-similar structures and the surface roughness or the interface width can be described by a dynamic scaling form,

for a system of size  $L$  and at time  $t$ ,  $W(t, L) = L^\alpha F(t/L^{\alpha/\beta})$  with  $F(\infty) = \text{constant}$ , i.e.  $W(L) \sim L^\alpha$  as  $t \rightarrow \infty$  and  $F(x) \sim x^\beta$  for small  $x$  or  $W(t) \sim t^\beta$  before saturation. The correlation length parallel to the surface ( $\xi$ ) scales with time as  $\xi \sim t^{1/Z}$ , where  $Z$  is the dynamic exponent ( $Z = \alpha/\beta$ ). By measuring the correlation length  $\xi$ , for each film thickness studied, we have calculated the dynamic exponent  $Z$  and showed that the values we obtained for  $\alpha$  and  $\beta$  are consistent.

## 2. Experimental details

About 1 mm thick substrates of PDMS (Dow Corning) were prepared by mixing gel with cross-linker in a 3:1 weight ratio. The substrates were cured for 15 min at 100 °C; this was followed by 2 h of slow cooling to room temperature. Before the loading into the deposition chamber, the PDMS substrates were carefully examined with an optical microscope for any surface irregularities. Several AFM images were also taken to check for any kind of irregularity. The surfaces were found to be quite uniform. The depositions were completed approximately 5 h after the curing process at room temperature. A plastic piece with an about  $400 \mu\text{m} \times 1 \text{ cm}$  opening was used on one of the substrates for chain-like pattern formation. Films were deposited using a thermal evaporator at a rate of  $1 \text{ A s}^{-1}$  with a base pressure of  $\sim 10^{-7}$  Torr. We used aluminium as a representative metal in view of its wide application; however, similar patterns are confirmed also for gold.

Film surface structures were obtained using an optical microscope and an atomic force microscope (DI) in tapping mode at room temperature. The tip radius was about 20 nm. Height information in the images was collected in  $512 \times 512$  arrays of pixels with various area scan lengths at room temperature. The surface was characterized by power spectrum density analysis (PSD). PSD analysis correlates the vertical amplitude with the spatial frequency ( $k$ ). In this case, the PSD in one dimension is given by

$$\left( \int h(x) e^{2\pi i k x} dx \right)^2 \quad (1)$$

here  $h(x)$  is the height profile along a scan direction ( $x$ ) of a given area. By calculating the amplitude of the Fourier coefficients, the power can be obtained for each line scan. The scaling behaviour can be found from the PSD [10]. In general, the self-affine surface exhibits a power law decay with a finite spatial frequency  $k$ :

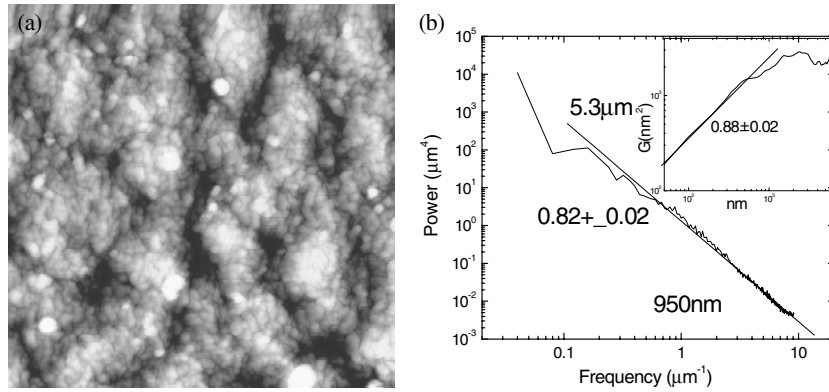
$$P(k) = C k^{-\gamma} \quad (2)$$

where  $C$  is a constant and  $\gamma$  is related to the roughness exponent  $\alpha$  by  $\alpha = (\gamma - d)/2$  where  $d$  is the dimension. We have also calculated the height–height correlation function for consistency in the exponent calculation. The height–height correlation function is given by  $G(\rho, t) = \langle [h(r_1, t) - h(r_2, t)]^2 \rangle$  with  $\rho = |r_2 - r_1|$ . This often exhibits scaling laws such as  $G(\rho, t) \sim \rho^\alpha$  at fixed time.

## 3. Results and discussion

The typical wavy structures on the surface revealed by atomic force microscopy are shown in figure 1. The image shows an early stage of coalescence of small clusters in the wrinkly pattern. The PSD (figure 1(b)) calculation shows the calculated roughness exponent to be  $0.82 \pm 0.02$ . The value is close to the value predicted by a fourth-order linear continuum equation given by [6]:

$$\frac{\partial h}{\partial t} = -K \nabla^4 h + \lambda \nabla^2 (\nabla h)^2 + \eta. \quad (3)$$

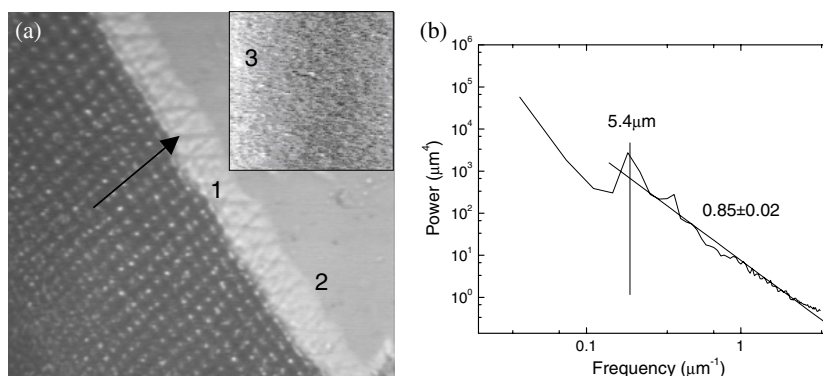


**Figure 1.** (a) AFM image of a wrinkly pattern ( $25 \mu\text{m} \times 25 \mu\text{m}$ ). Small metal clusters are visible, showing not just buckling. (b) Corresponding power spectrum of the surface. Inset: corresponding height–height correlation function.

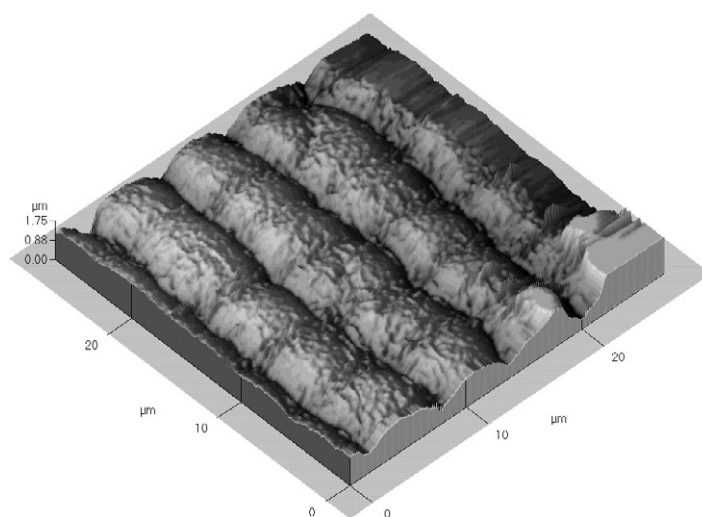
The fourth-order term arises from the surface diffusion of atoms caused by the curvature dependent chemical potential. The  $\lambda$  term is due to the lateral growth of the surface and follows the KPZ equation [9]:

$$\frac{\partial h}{\partial t} = v\nabla^2 h + (\lambda/2)(\nabla h)^2 + \eta(r, t). \quad (4)$$

This equation is expected to describe the large length scale dynamics of the growing interface in the absence of a specific conservation law. The first term in the right-hand side is the surface tension term, as it tends to smooth the surface through evaporation condensation processes. The nonlinear term accounts for growth along the local normal direction and  $\eta(r, t)$  is the Gaussian white noise of constant strength which accounts for microscopic fluctuations. The value predicted for the roughness exponent for this case is 1. This is higher than the value obtained from the PSD calculation. The reason could be the existence of several competing mechanisms at the surface. Smoothing effects, such as surface diffusion, could only lower the exponent. From AFM images, we observe clustering of atoms at lower thickness and labyrinth patterns at higher thickness. This is a clear indication of surface diffusion. The surface diffusion in this case is stress driven. Surface instability due to stress was studied by Grinfeld [11]. It has been shown that a planar surface is unstable under a certain critical value. This nonlinear instability induces mass transport and hence surface diffusion. However, we believe the shadowing effect cannot be ignored, since buckling of the films creates hills and valleys. As more atoms arrive at the surface, hills will receive more atoms than the valleys. Unlike most of the deposition processes, where shadowing can occur due to deposition noise [12] or angular distribution of atoms [13], on an elastic substrate shadowing is induced by the change in local nature of the substrate. Therefore, shadowing combined with a smoothing mechanism could result in a roughness exponent ( $\alpha$ ) less than 1. In fact, our calculated growth exponent ( $\beta$ ) value of  $0.15 \pm 0.02$  is consistent with the value obtained for a model with shadowing growth with a low sticking coefficient [14]. The PSD calculation shows that there are two cut-off lengths. 950 nm corresponds to the small clusters that are visible all over the surface (figure 1(a)). The coalescence of these clusters becomes correlated, causing saturation, and finally large features are formed due to this coalescence. The large features are represented by a  $5.3 \mu\text{m}$  length scale. The wrinkle-like patterns formed at this scale can be attributed to the difference in elastic constants of the film and the substrate. The thickness of the film is also an important factor in this case as these wrinkle patterns change their shape with increasing thickness (figures 1, 5 and 7).



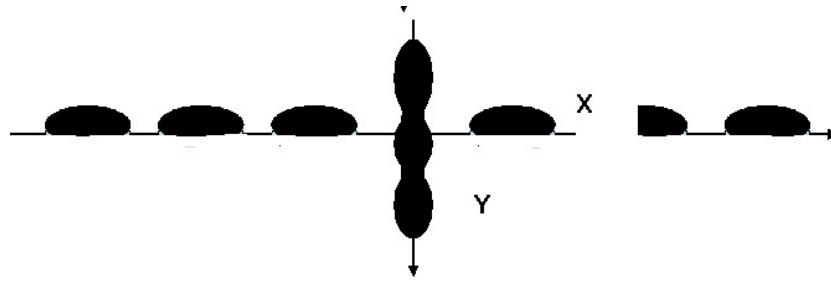
**Figure 2.** (a) Optical micrograph of the ordered domain. Region 2 is the PDMS surface. Region 1 is shown to show that there is no delamination of the thin film. Region 3 (inset of (a)) is a  $3 \mu\text{m} \times 3 \mu\text{m}$  AFM image of the PDMS surface before deposition. The AFM image shows that the surface is well cured and devoid of any large defects. (b) Power spectrum of figure 3 with a linear fit.



**Figure 3.** 3D image of the ordered domain.

The ordered structures that formed when films were deposited with a  $400 \mu\text{m} \times 1 \text{ cm}$  window on the substrate are shown in figures 2 and 3. The AFM image shows chain-like surface structure aligned along the shortest direction. The PSD shows the exponent to be  $0.85 \pm 0.02$ . The situation can still be described by the fourth-order nonlinear equation [6]. The equation gives us a general idea of the surface process but does not answer the question of why atoms diffuse only the shorter length scale, i.e. parallel to a surface with a  $400 \mu\text{m}$  scale rather than 1 cm. Most of the studies available address problems in terms of MBE growth [15]. Even though the short range attraction [16] or Grinfeld-type [11] instability can be present during the initial growth process, it does not explain why atoms flow only parallel to the shorter direction ( $Y$ ) to form this chain-like structure.

We propose a model where the final equilibrium structure is approximated as a prolate spheroid or egg shape. Using this model, from the theory of elasticity [17], we can show that



**Figure 4.** Schematic diagram of a prolate spheroid shape. The elastic energy minimum for the surface is along the smallest length scale ( $400 \mu\text{m}$ ) direction; therefore, diffusion is favourable parallel to a surface with a  $400 \mu\text{m}$  length scale, creating a chain-like structure. In the  $x$  direction (length:  $\sim 1 \text{ cm}$ ), no atomic diffusion due to step edges being too far for any interactions. Also the volume would be much higher compared to the  $y$  direction.

the minimum energy surface is the one with the smallest volume.

$$U = \frac{1}{2} u_{ik} \sigma_{ik} V = \frac{1}{3} (\sigma_{xx} + \sigma_{yy}) (u_{xx} + u_{yy}) V \Rightarrow U = \frac{2}{3} \mu (a - b)^2 V / R^2$$

$$u_{xx} + u_{yy} = (a - b) / R \quad (5)$$

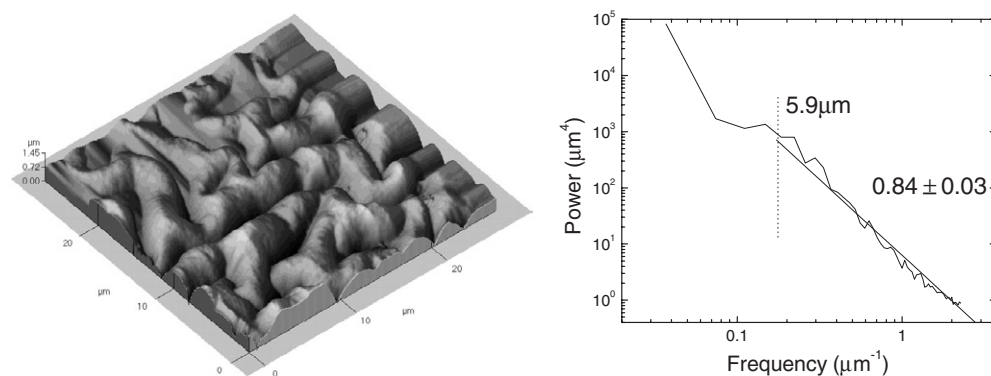
where  $U$  is the elastic energy,  $\sigma_{ik}$  is the elastic tensor,  $\mu$  is the modulus of rigidity of the material. The semi-axes ( $a$ ,  $b$ ) and  $R$  are thickness dependent. In general,  $b$  would be approximately of the order of the film thickness and therefore  $a \gg b$ . The value of  $R$  can be estimated from the number of spheres within  $400 \mu\text{m}$  by adding all the small radii. The value in this case would be of the same order of magnitude as  $a$ . By substituting  $a/R \sim 1$ , we can write our final equation as

$$U \approx \frac{2}{3} \mu V. \quad (6)$$

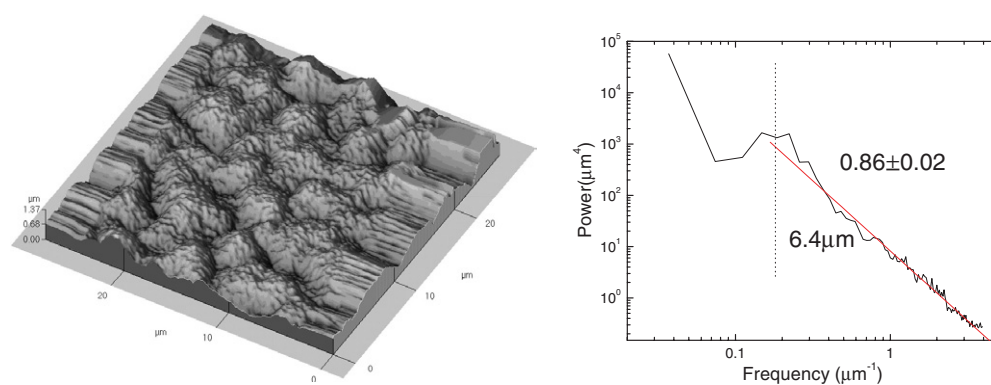
According to equation (6), the surface energy will tend to arrange the surface with the smallest possible volume or parallel to the  $400 \mu\text{m}$  width (figure 4,  $Y$  axis). The length of the  $400 \mu\text{m}$  wide line was about  $1 \text{ cm}$  ( $X$  axis). Now we can correlate this observation with equation (3). The fourth-order term in equation (3) is a curvature dependent chemical potential. By restricting the deposition process, we have created curvature anisotropy. The anisotropy is higher in the direction of smallest volume; therefore, diffusion is high in this direction.

Another possibility is the existence of dipole interaction between the two large step edges due to the tilt dependence of the surface. Obviously, closer steps will interact ( $400 \mu\text{m}$ ) better and faster. A surface can develop a small tilt relative to the substrate due to the shadowing caused by the opening or the angular arrival of the atoms from the source. If two step edges of different height interact via fields  $\Phi_{1(k)}$ ,  $\Phi_{2(h)}$ , the typical force would be proportional to  $(\Phi_1 \times \Phi_2) \tan \theta$ . Where  $\theta$  is the tilt angle with respect to  $L$ , and  $k$  and  $h$  are corresponding heights. Ideally,  $h = k = \text{film thickness}$ , in which case the force becomes zero and diffusion of atoms due to dipoles stops but random diffusion or diffusion due to Grinfeld-type instability may still be present.

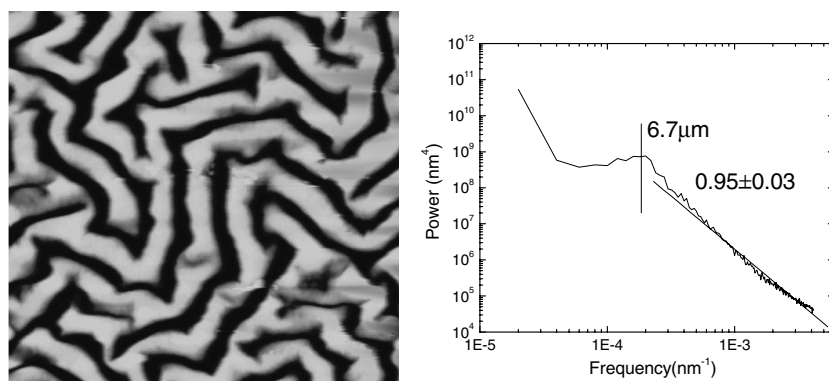
Figures 5–7 show the surface morphology and the corresponding power spectra at three different thicknesses. They are  $100$ ,  $350$  and  $1000 \text{ nm}$  respectively. At  $1000 \text{ nm}$ , a definite transition from an island-like sinusoidal pattern to a totally labyrinthine pattern is observed. The optical micrograph shows that there is no delamination. Therefore, the buckled structures represent an equilibrium situation with relaxation of the in-plane strain and the stresses associated with bending. The equilibrium equation describing the vertical displacement of the compressed film can be found in the literature [18].



**Figure 5.** AFM image of 100 nm thick film with a power spectrum (LHS).

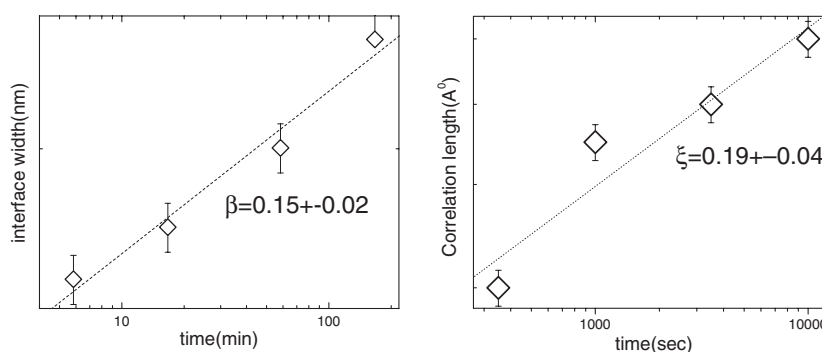


**Figure 6.** AFM image of 350 nm thick film with a power spectrum.  
(This figure is in colour only in the electronic version)



**Figure 7.** AFM image for 1  $\mu\text{m}$  thick film and a corresponding power spectrum. Image size: 50  $\mu\text{m}$   $\times$  50  $\mu\text{m}$ .

From the correlation length at each thickness, we have calculated the dynamic exponent  $Z$  (figure 8(b)). Our value,  $\sim 5.2$ , is within the range of the values obtained from the equation  $Z = \alpha/\beta$  (figure 8(a)). The value calculated from  $\alpha$  and  $\beta$  is  $\sim 5.6$ . This value is a little



**Figure 8.** (a) Calculation of the dynamic exponent from the interface width. (b) Calculation of the dynamic exponent  $Z$  from the correlation length ( $\xi$ ).

higher than the predicted theoretical value. This is not really surprising because of the slow evolution of the films with time. The slow evolution of the interfaces causes breakdown of the dynamic exponent  $Z$ . It is possible that the film surfaces are exhibiting asymptotic behaviour similar to that shown by vapour deposited silver film on quartz [19], where the roughness exponent is found to be  $\sim 0.82$  regardless of the film thickness within the 10–100 nm range. The power spectrum calculation also shows that in the high frequency range the power did not shift to higher or lower amplitude ranges. This is suggestive of initiation of the interface pinning occurring at a lower length scale. As the roughness evolves very slowly, the stress due to the increased thickness is absorbed in the elastomer layer. Due to this ‘damping’-type behaviour of the PDMS, we observe a higher value of the dynamic exponent for the wrinkled metal surface.

#### 4. Conclusion

In conclusion, we have shown that wrinkle pattern can form not only by buckling but also by diffusion. We have calculated the roughness exponent to show that the underlying surface process is diffusion mediated and wrinkling of metal can be described by a continuum theory. Inconsistencies in the observed values of roughness and growth exponents are attributed to shadowing combined with a smoothing mechanism. While long range interaction of the order of  $\mu\text{m}$  is one of the least addressed topics in the literature, on the basis of the diffusion process, a model can be proposed based on step edge interaction and minimal surface elastic energy. There are several questions yet to be answered, in terms of the direction and range of the interaction in the  $\mu\text{m}$  range, the exact nature of the pinning and the incorporation of this effect in the existing theory. Better understanding of these processes and, especially, the curvature dependent chemical potential will guide us to an understanding of self-assembly of nanostructures on stretchable substrates.

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