

Instability and Patterning of Thin Polymer Films

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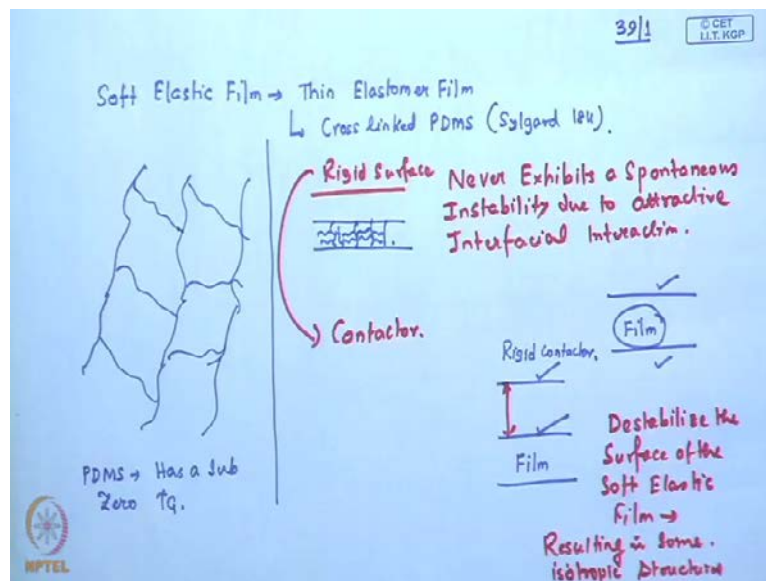
Indian Institute of Science Bangalore

Lecture No. # 39

Elastic Contact Instability and Lithography

Welcome, this is penalty wet lecture of this course series. Today what I do is I will introduce you to another class of instability. So we are talk about a instability, but what we have really discussed in great detail is the spontaneous deviating of an ultra thin discuss liquid like film. What I discuss today is the instability, what is known as the contact instability of a Soft Elastic Film, and also we talk about the elastic contact lithography.

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So, Soft Elastic Film essentially we are talking about a thin Elastomer Film, and we have actually talked about an Elastomer in pretty good detail while talking soft lithography. It is essentially a cross link PDMS is a perfect example of a Elastomer thin film.

So, we have argued that these as some unlike a polystyrene in a p m m a film which are essential the linear chain molecule, so they are in a frozen in glassy state at room

temperature. So, if you hit them beyond the glass transition temperature they behave like a liquid, but this Elastomeric film the soft elastic films, they are physically cross linked actually. So and these other is lot of flexibility within the matrix this we have already argued, and PDMS is as a material as a sub zero T G, so its typically liquid at room temperature, so we have already also argued how to cross link syngard film and it becomes flexible, etcetera, etcetera.

Typically, there are two parts so you can read the proportion of the cross linker to get the volatile the exact shear modules of the film and, this elastic modules you can find out from geometric analysis to the extent of what is the storage modules, what is the lose modules, what is the extent of velocity etcetera, the system is very simple.

So, firstly since this molecules are cross linked they are actually stiffer than a discuss film and therefore, even **if there if** you make a very thin film out of it due to interfacial attraction if they try to deform the physical cross link sort of opposes in addition to the surface tension forces this deformation. So this type of a system never exhibition spontaneous instability, due to attractive interfacial interaction it never exits, however what we seen that if you take such a film in close proximity to another rigid surface.

So, here is another rigid surface which we be referring as the Contactor, what happens is below a critical separation distance between the two, now instead of talking about the interaction between the two interfaces of the film as we have so for talked about, we are now talking about the interaction between the free surface of the film an another rigid Contactor.

So, this is the interaction we are now talking about, and when the separation distance is below a critical separation distance, what happens is there **is thus** this attractive interaction between these two destabilized the surface of this soft elastic film resulting in some isotropic structures. So, this is the system geometry we are talking about so you have an elastomeric thin film and you have a rigid Contactor.

So if you bring the Contactor into a close proximity, so below a critical separation distance there are these self organized structure formations on the film surface.

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Contact Instability of Thin Soft Elastic Films

- Surface of a soft thin elastic film when brought in contact proximity to another rigid flat surface or contactor, undergoes spontaneous roughening with the formation of a short-wave isotropic structure.
- The patterns form due to competition between attractive van der Waal's force and restoring elastic forces originating from the deformation of the surface

Instability sets in on Film Surface
Manifested by Random Patterns

Contactor

Elastomeric Film

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So, the pattern essentially form due to a competition between the attractive Van Der Waals force acting between the free surface of the film, and the Contactor so again the Van Der Waals force again the attractive Van Der Waals force is present but the forces is not active between the two interfaces of the film but it is between the Contactor and the film, and any instability is essentially opposed by surface tension as well as elastic forces originating from deformation of the matrix.

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Soft Elastic Film → Thin Elastomer Film
↳ Cross linked PDMS (Sylgard 184).

PDMS → Has a Sub Zero T_g.

Rigid Surface
Never Exhibits a Spontaneous Instability due to attractive Interfacial Interactin.

Contactor.
Contactor
Film
Which Structures are Short-wave Patterns

Rigid Contactor, Film
Destabilize the Surface of the Soft Elastic Film → Resulting in some isotropic structure

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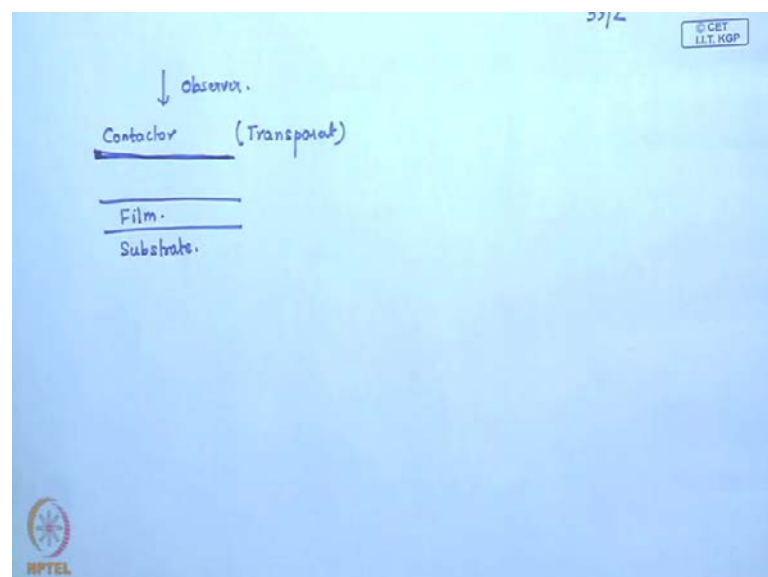
So, what happens is now you are looking at a system, so suppose the film involves like this, we will immediately see so this a Contactor let say which is coming in contact, the film become unstable makes an structures with sort of tend to go and touch the Contactor.

But, what happens is you have to understand that you are talking about a cross link films so film, so the moment some parts of the film sort of elongate towards the Contactor due to the instability, other parts try to pull it back because you have this physical cross links which are present. This is unlike a purely viscous system where you do not have a any restoring mechanism other than the surface tension, so once you can sort of overcome the stabilizing influence of surface tension there is nothing to hold it back really.

So, **this is so** a competition there or in other words there is much stronger competition here or much stronger restoring effect present within the film, in case of an elastic film which is in the form of the physical cross link of the molecule.

And these competition result in the structures which are Short Wave Structures, so the patterns have periodicity which are much smaller as compare to the long wave approximation we have already talked about in case of deviating of a thin film, so it much smaller than λ but that does not really happen here. There is a small we will just give a very brief theoretical understanding of that but before that let see what type of instability we are actually talking about.

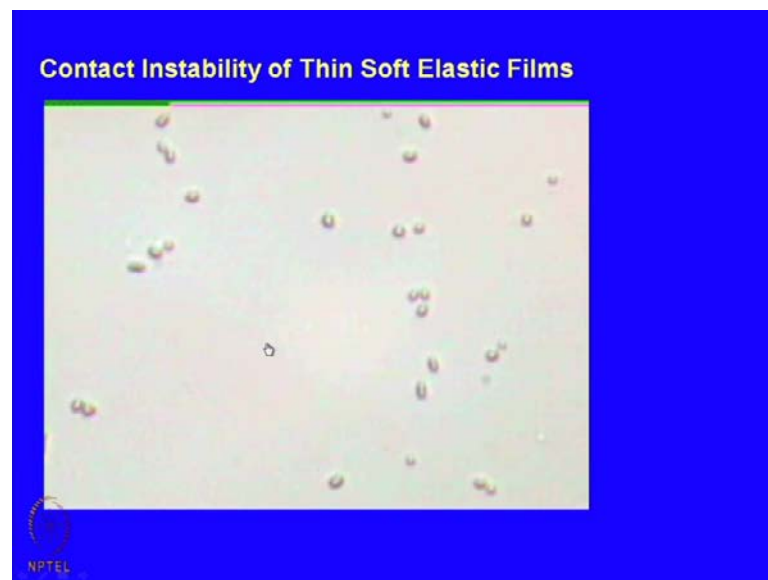
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So, now what you are seeing is actually from the top, again under a microscope so you are now seeing so here is the Contactor, here is the film and which is rigidly bounded to a substrate, and the Contactor of the film is approaching towards the Contactor.

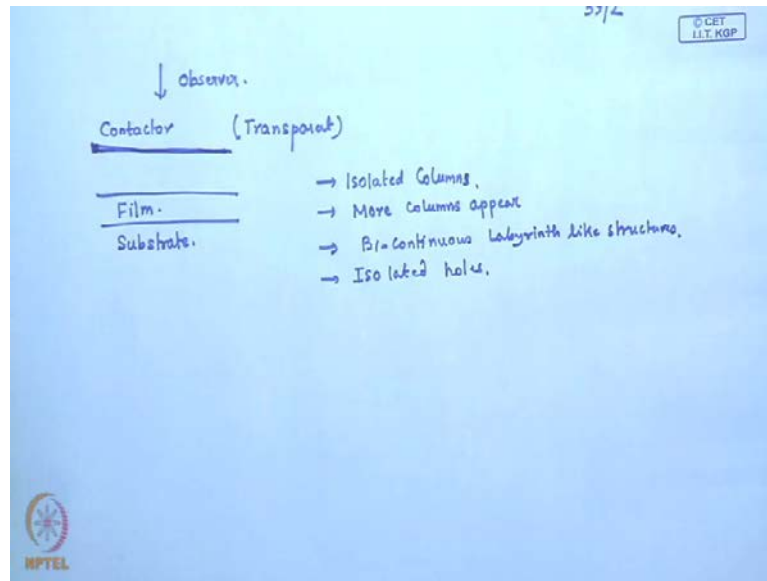
And you are seeing from here, so is possible to perform this type of experiments **if you on a** on a optical microscope platform, only requirement is in that case the Contactor as to be transparent, and you here you use a transparent Contactor and the film or the Contactor is progressively approaching the film see what happens.

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So, as the Contractor approaches the film you see **some** something, certain things are appearing and which is growing in number, so these are some insolated column so let us note down what are the things we see, first we see some isolated columns.

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Then we see that more columns appear, then if you further allow the progress you see that the columns are getting transformed into Bi Continuous Labyrinth so the next thing is you see some Bi Continuous Labyrinth like structures.

So here is the Bi Continuous Labyrinth like structures, and then if you allow a approach this Labyrinth get transformed into isolated holes which we will soon see here, now is a stage which compresses isolated holes that black dark spots, before the two come in complete contact. So here is something which is again very interesting.

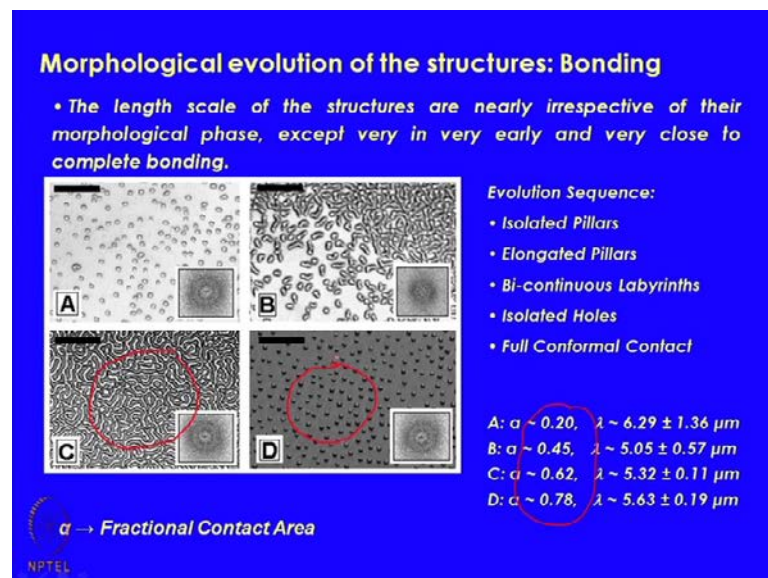
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So, let us start from the beginning again and watch the movie, so the Contactor is approaching from the top and what you see is, **the** first the appearance of the manifestation of instability is with the formation of some isolated columns.

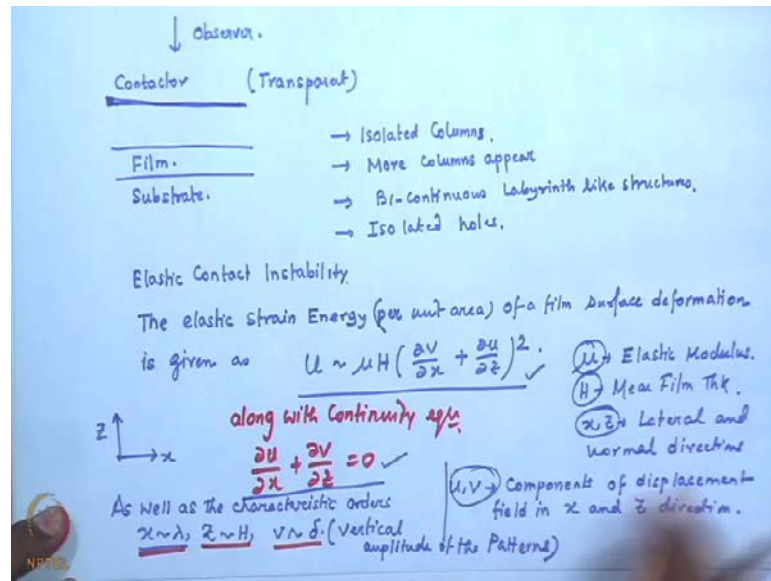
And then as it approaches further, more number of columns appear and then certainly use see whole lot of structure to all over the film surface the structure now transform to Bi Continuous Labyrinth like structures and further approach, this Labyrinth like structures get transformed into isolated holes, and before the Contactor in the film coming conformal absolute conformal contact. So, what happens here is that the structure originate as we have already pointed out due to a competition between the attractive Van Der Wall's forces.

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And the pre storing effects imparted by surface tension as well as the elastic deformation. So this is the Morphological pollution sequence you first the onset of a instability is with the formation of columns, than more number of columns appear the column state to be elongated and they gradually get transformed to this Labyrinth like structures, and then it is associated with an increase in the fractional area of contact, then you have these random Bi Continuous structure and then finally, these holes. What is interesting the basic physics of, so this is what is known as Elastic Contact Instability.

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And very briefly the basic physic of Elastic Contact Instability can be understood, that the elastic strain energy per unit area of a film surface deformation is given as $U \sim \mu H \left(\frac{\partial v}{\partial x} + \frac{\partial u}{\partial z} \right)^2$, where μ is the Elastic Modulus, H is the Main Film Thickness x and z the Lateral and normal direction, u and v are the components of displacement field in x and z direction they are not velocity.

Now, this equation along with the continuity equation which is, as well as the characteristic orders which are x is the of the order of λ which is the natural line scale of instability, z is the order of H the film thickness and v is the order of δ which is the vertical amplitude of the pattern.

So, we have this equation which gives the elastic strain energy along with the continuity equation $\mu H x \left(\frac{\partial v}{\partial x} + \frac{\partial u}{\partial z} \right)^2$ and u, v are defined here, and the characteristic orders again as we did an order of magnitude analysis, so x is the order of λ there is out of H and these are of the order of δ which is the vertical amplitude.

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$$U \sim \mu H \delta^2 (\lambda^{-1} + \lambda H^{-2})^2$$

- The energy penalty for deformations increases both for very short ($\lambda \ll H$) and very long ($\lambda \gg H$) waves.
- Therefore, the minimum elastic Energy pattern, for which $\left(\frac{\partial U}{\partial \lambda}\right) = 0$ is possible only when $\lambda \approx H$

Detailed theoretical calculations show $\lambda = 2.96H$

$\lambda = 3H$

✓ **Contactor**
✓ **Film.**

Bonding ↓ ↑ Debonding

Film. → (Long wave)

Elastic Deformation of a Soft Solid Film ⇒ No transport of material by either convection or diffusion!

What gives is that U the strain energy; you get an expression like this. So, from this expression what you can see that the energy penalty for deformation, this derivation is beyond the scope of this lecture, so there is no need for you to go into the starvation. The energy penalty for deformation increases both for very short and very long waves.

So, because of the non-linearity of the equation so if λ is very short or λ is very long both waves U sort of increases, and the minimum elastic Energy pattern for which λ will be equal to 0, as a length scale is possible only when λ is of the order of H or the literal wave length of instability, natural wave length of instability is of the same order of H , and detail theoretical calculations show pre factor close to three, so a λ is roughly three times the film thickness and these as been verified in experiment.

What is a more interesting is, apart from this scaling this short waves scaling which make the structures really short wave as compared to the long wave structure absorbed in instability, there are couple more things which are important. Firstly what we need to understand that this form of instability is completely distinct from the instability of a purely viscous thin film we have discussed so far.

First difference we have already talk that this instability is triggered or engendered by an, inter surface attractive interaction, inter surface. So you have a, you have to have Contactor and then you have film, and the instability results due to an attractive

interaction between the Contactor and the film rather than a spontaneous instability as absorbed in a purely viscous liquid thin film and, the interfacial, the attraction or the interaction was between the two interfaces of the film, here the interaction is between the film major interface and the interface, the free interface of the film and the Contactor.

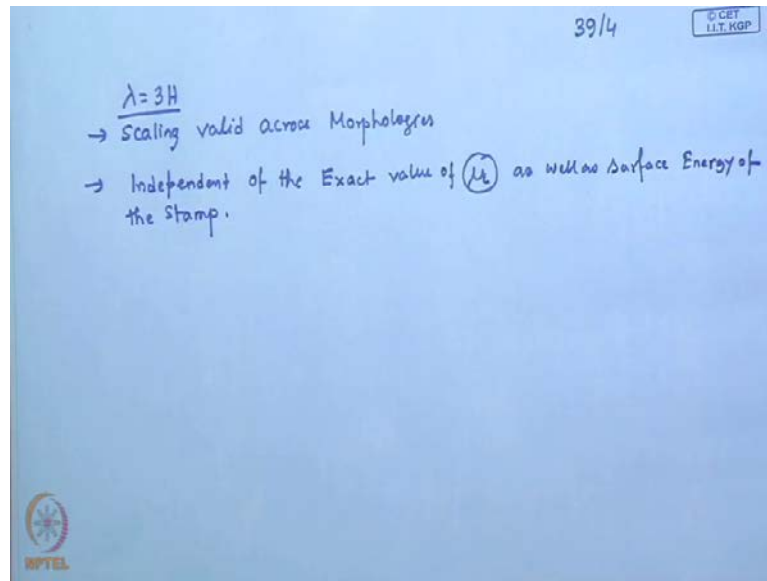
The second more important thing is, that the surface evolution in case of a deviating or an instability of a liquid thin film was associated with physical displacement of liquid from one part to the other, physically displacement of polymer by diffusion and convection. In contrast here, there is no physical flow of either by diffusion or convection of the material, and what you see is just an elastic deformation of a soft solid field, so it is a purely elastic deformation of a soft solid film, and there is no transport of material by either convection or diffusion.

The other important thing as for as the scaling so that the thing we understand of course, it is a short wave instability as compared to a long wave instability that is absorbed in case of a the instability of a spontaneous instability of a liquid film. The other interesting thing is, what we have seen in the video is the progressive Morphological evolution during the bounding and the de bounding stage, so if you perform the opposite experiment that is the Contactor comes in complete contact and then you withdraw it again, so the experiment we saw the other or the movie you saw was for the situation or the case of bounding, were the Contactor was progressively approaching the film.

But, you can also do a de bounding experiment and you see a similar Morphological evolution in the inverse order, so fast from conformal contact if you start de bounding you will first see the holes coming in then it will be followed by the Labyrinth, then the columns a b and then finally, detaches. There is of course, a bit of a (()) in terms of the vertical suppression distance.

So, the see the critical distance at which the patterns form while you are approaching, and the finally, when the two surfaces de bound completely there is a difference between that but that can be accurate to peening of the patterns to the surface. Other interesting thing is, this λ equal to $3H$ scaling or whatever scaling is there, remains valid almost across the difference morphological settings, so the columns the Labyrinth the holes, they all follow this λ equal to $3H$ scaling .

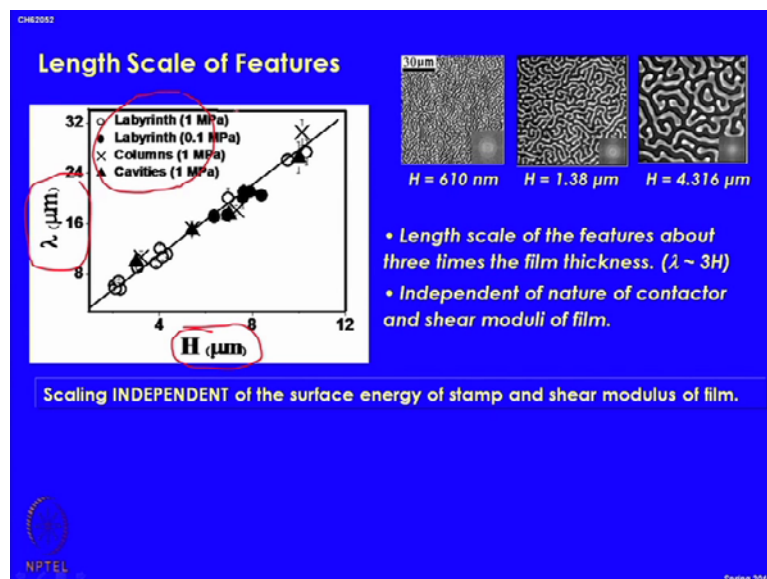
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Importantly this scaling relation valid across Morphologies, and very importantly this scaling relation is independent of the exact value of μ , exact shear modulus, as well as surface energy of the stamp. So here also you now have a potential flexibility.

So, you take films of two different thickness, the periodicity of the structures you will get will be different but the morphologically the structures will be roughly the same. And the other thing is you can vary the vertical separation distance and can create either holes or channel, holes sort pillars or Labyrinth.

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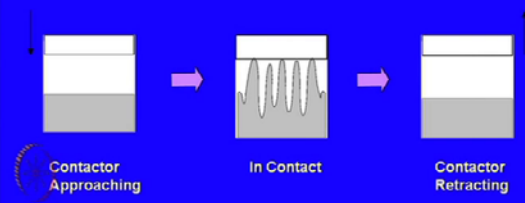


So, these are interesting aspects from the sun point of patterning, you can see that with this films of different thickness if you go an in focus lambda plot they fall, roughly fall on the same line irrespective of the Morphology, irrespective of the share modules of the film, as well as the nature of contact.

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Permanent Patterning

- Patterns are engendered due to elastic deformation, and hence disappear once the contactor is withdrawn
- Features are made by exposure to UV-ozone treatment.
- UV irradiation at 185 nm produces ozone from atmospheric oxygen.
- Atomic oxygen produced at 254 nm irradiation.
- This atomic species reacts with the siloxane group, leaving behind a stiff surface layer consisting of oxides of silicon
- The oxide layer of high modulus, prevents film relaxation..

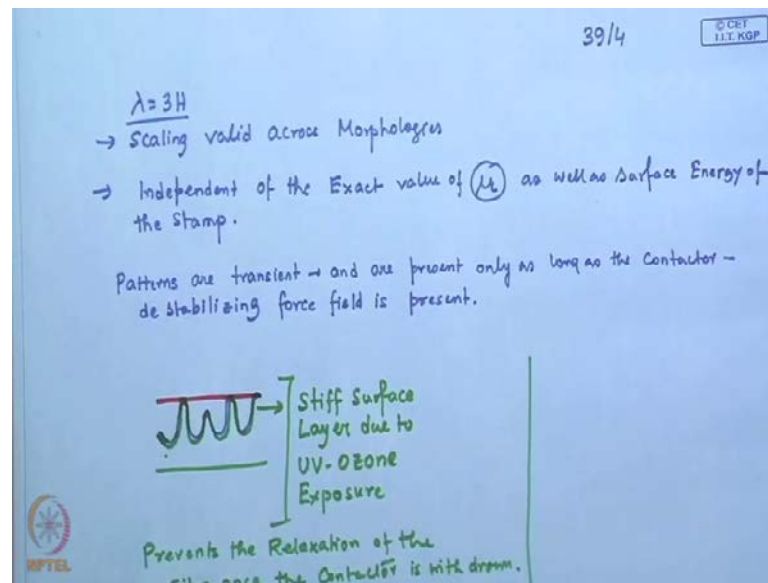


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The other important thing to release is that, these structures unlike the structures formed in deviating where the energy sort of dissipate due to viscous dissipation, and there is a permanent deformation on the film surface, so you take a flat film it deforms permanently resulting in holes or droplets or whatever.

In contrast here since the nature of that deformation on the film surfaces is predominantly or completely elastic, so the structures or temporarily or they are not permanent. So the moment the Contactor is withdrawn, the starch the elastic or the flexible crosslink's, the bound present or the flexible crosslink's present within the polymer matrix, restores the surface of the film, elastic film back to its original flack Morphology.

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So therefore, the patterns are transient and are present only as long as the Contactor which essentially signifies the de stabilizing force field is present. Once the Contactor is withdrawn the patterns again restore back to its original flat morphology. So, if you want to use this elastic contact instable induced instability as a viable pattern in technique, one of the critical requirements will be to make these patterns permanent, and we are talking essentially of using poly- di- methyl siloxane basic system.

So, what can be done or what has been done or what is typically done is, so this is how it is the Contactor approaching the film, you have a flat film so you while as long as it is in contact proximity or in contact you have the structures but once the contracted is retracted the structures go away.

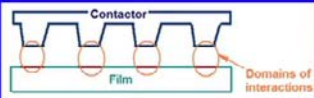
So, what is done is that, if during this stage while the Contactor is in contact proximity with the surface, if UV is exposed if the hole assembly is exposed to UV light, what happens is the ultra violet radiation produces **at moron than** molecular atomic oxygen, by dissociating first the atmospheric oxygen at a wave length of 184.9 nano meter, and these molecular this atomic oxygen again recombines with the oxygen that is available in the atmosphere to form ozone. That is why this UV exposure a thing which we have already talked while discussing about tailoring the surface energy of the PDMS film in terms of a whole lot of lithographic applications CFL mimic etcetera.

Then again also we discussed it but let me quickly repeat that, so this UV exposure what it does is that, it breaks down the atmospheric oxygen into atomic oxygen the at 184 nano meter is radiation, this atomic oxygen recombines with the molecular oxygen to form ozone but these ozone also gets dissociated by the UV at the 253.8 nano meter or 254 nano meter radiation. So, what you have is within the chamber, you have a whole lot of atomic oxygen's which are present, these atomic oxygen reacts with the siloxane film surface and it sort of forms higher oxides of silica on the surface of the film.

So here in presence of a Contactor you had a deformed film surface, and then you expose it to UV ozone so it forms a very stiff surface layer and **this stiff surface layer** so this stiff surface layer now prevents the relaxation of the film once the Contactor is withdrawn. So, thereby you can make permanent patterns based on elastic contact instability of course, the making the structure permanent is associated with some chemical modification of the surface.

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Elastic Contact Lithography



- Use a *patterned contactor or stamp*.
- *Novel intermediate structures obtained.*

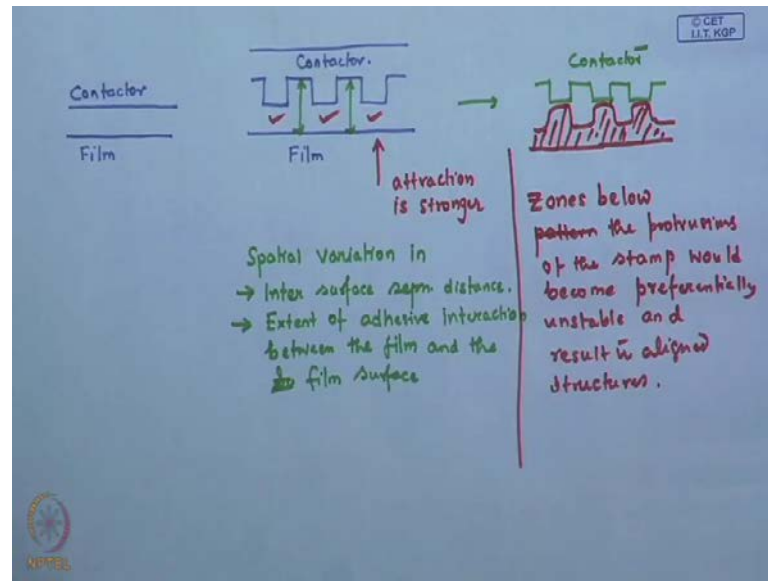
• *The structures form based on the commensuration of the periodicity of the stamp and the natural length scale of instability of the film.*

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Now, again if you look into the nature of the structures, the even if you are able to make these permanent patterns by UV exposure etcetera, the critical issue is that you still cannot use this Labyrinth or hole or columns or whatever, for any practical application because, they lack long rang order. So, what was consider that if this elastic contact instability as to be used as a viable pattern in technique, the structures a raising out of elastic contact instability have to be alight.

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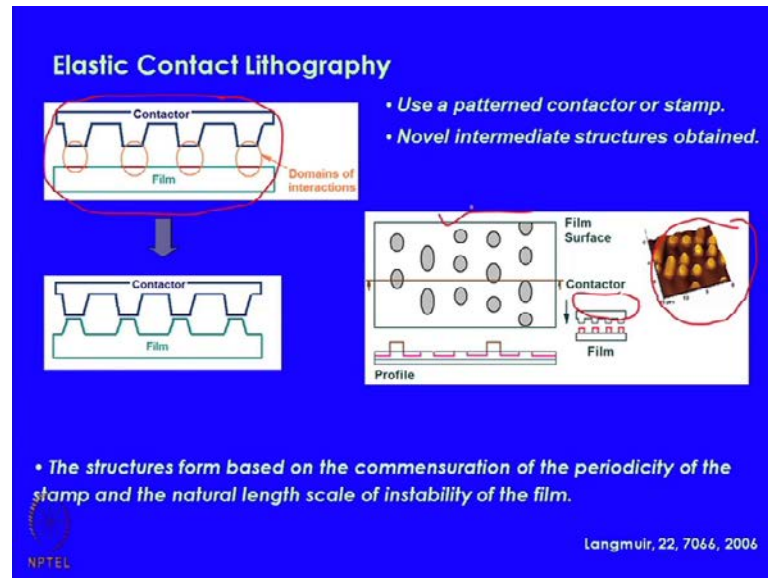


And the simplest approach that was taken was, so this is the regular setting, so you have an elastic film which I am writing now is a film you understand it is an elastic film, this is a Contactor. So, the attraction between the Contactor and the film the adhesive or the attractive interaction which is responsible for the manifestation of the hole instability is uniform between these two now in a regular case. So, instead of that if a topographically structured Contactor was taken, then what would happen? You have a film, what will happen is because of the topographic contrast on the **on the** stamp surface, over these zones the attraction is stronger as compared to these zones which are where the separation distance is higher.

So, one can talk about a simple grating and instead of using a flat Contactor one can approach the film with a grating let us say, and because of the picture height or the topographic contrast on the surface of the Contactor, now you have a spatial variation in inter surface separation distance, which results in a spatial variation in the extent of adhesive interaction between the film and the surface. So what is expected? What was expected was that by using a pattern Contactor it might be possible that the zones below the stamp protrusions will preferentially become unstable and therefore, result protrusion of the stamp preferentially unstable and result in align structures.

So, this is what was expected, and then if one can make these structures permanent at this level probably, one can get align structures using or utilizing the concept of elastic contact instability.

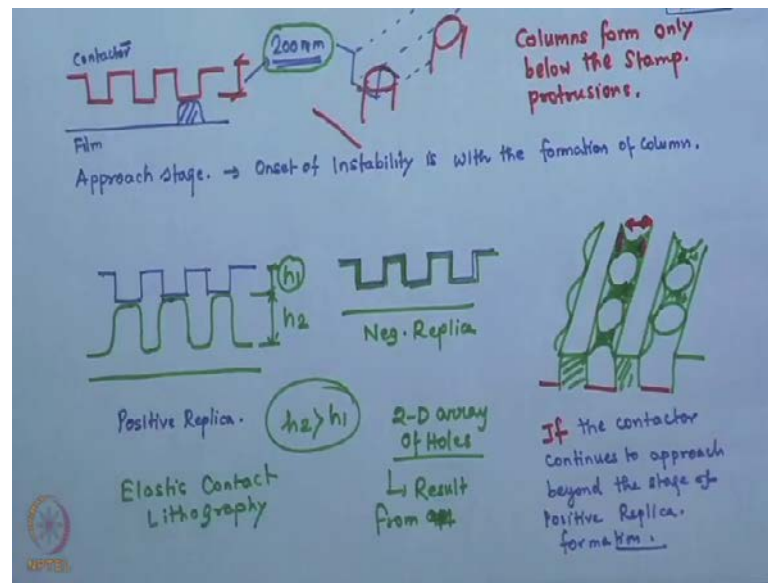
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So, this is what this systematically is shown here. And let us now try to see what exactly happens, so this you if these this type of a Contactor a pattern Contactor approaches the film, it is expected that it forms, it would, we expected or people expected, people working on this area expected that probably positive replica of the stamp pattern will form on the film surface. Let us say what exactly happen, so here is a flat elastic film to which a simple grating Contactor is approaching and we will have a carton to over this zone to show you what exactly happens.

So, as the film approaches first far away pre nothing happens, so it now comes closer and below that critical separation distance so there is now an active interaction between the Contactor and the film surface, and you see that the onset of instability here also onset of the page.

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So, we are looking at the approach stage. So the onset of instability is again with the formation of columns, why I use the what again, because of the fact that when we remember that, when we used a flat Contactor then also the instability, the onset of the instability was with the formation of isolated columns but what is the difference here, the key difference here is that because of the presence of the stamp protrusions or a topographically patterns stamp, the columns that appear are not random but they are sort of aligned, aligned and all the columns form at some zone are the other which is below a stamp protrusion, it may not be forming at all locations.

So, let us say this is a stamp protrusion which you are looking at and this is the film surface so this is how it goes, and somewhere of the other so the column can form here or here or something like that, so it forms only below the stamp protrusions. So the columns forms protrusions therefore, the columns get aligned.

You go for further approach what happens is because, you have some columns which are probably spanning between the film surface and the stamp protrusion, so the stamp comes down further so what is happen the what happens this columns sort of elongate along the strips or the stamp strips, as well as the reduced inter surface distance results in enhanced attraction between the stamp protrusions and the film surface therefore, more number of columns appear, and eventually one get's to a positive replica of the stamp.

So a question to ask is looking at this type of a structure how does one ascertain whether it is a positive or negative replica because, a problem is the duty ratio is one here. So you are seeing atomic force microscope images in this particular case, so at every stage what was done that the pattern were frozen by UV ozone exposure so that the stiff surface layer appears and therefore, these structures can be made permanent, and then one investigates them.

And so what happens is that critical question one can ask you, how do you determine looking at a structure like this whether it is a positive replica or a negative replica? So let us say you know the geometry of the Contactor again some atomic force microscope what you can find out, for this is particular case if I remember correctly the height of the grating was 200 nano meter.

But, if you look at so you have essentially reached this stage where a positive replica a replica forms, and we expect it to be a positive replica but then you are looking at this structure and you see it is a grating but how do you negate, so this would be a positive replica but one can also argue that well it is not that you have simply emboss the film which is also possible, and you have formed a negative replica, so this would be a negative replica.

Since the periodicity are same and the duty ratio is one, it is very difficult to identify whether it is a positive replica or negative replica but if you do an AFM scan and find out that the feature height and was found out here, that let us say the feature height was h_2 and the stamp feature height was h_1 or the Contactor feature height, it was seen that h_2 was higher than h_1 .

So, using 200 nano meters, using stamp with 200 nano meter feature height it was possible to obtain around 400 nano high structures, which is never possible if a negative replica is forming therefore, it has to be a positive replica. So, this way one can of course, use a this the concept of elastic instability to generate order structures, and also the advantage is unlike a soft way, soft lithography based technique where you like embossing, where you get a negative replica you can actually, create our aspect ratio structures but there was more to it.

So, since beyond the stage where a positive replica is formed if the Contactor was approach further what happens is, you have to understand that these structures if they not frozen, they are some tall strips of polymer of the soft elastomeric material.

And if you press them from the top what will happen, there will be some sort of bulging instability along the periphery of this polymers, so it is something like this is the morphology, and then you have adjacent stripes which are now exhibiting this bulging instability, and this happens only at a stage if the Contactor continues to approach beyond the stage of positive replica formation.

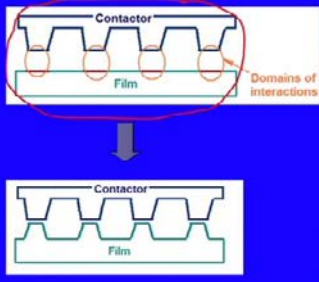
So, beyond the stage of positive replica formation it continues to approach and you see some, these types of bulging instabilities along the length of each of the positive replicated strips. So now what happens since now the additional thing that arises now these two bulged portion from these adjacent regions sort of start interacting with each other.

So, the inter surface attractive Van Der Waals interaction now set seem between these two bulged portion, so this is the film which is a positive replica as formed, and now there is an inter surface attractive interaction. So, what happens is these results bridging of these bulged portions forming bride like this and therefore, this is an unique thing which is not seen in any exiting of soft lithography technique, that these bulgier sort of bridge up in this direction also resulting in an, so this sort of bridge up resulting in an array of holes, so this a perfectly ordered 2 D array of holes that result from approach or contacting an elastic film with a single one de patterned stamp.

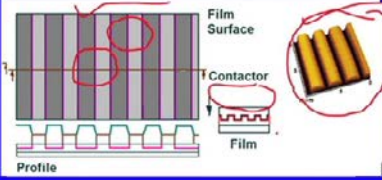
So, this is really unique feature of this method, this structure can be made permanent and this whole approach which is patented technology now is known as Elastic Contact Lithography.

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Elastic Contact Lithography



- Use a patterned contactor or stamp.
- Novel intermediate structures obtained.



- The structures form based on the commensuration of the periodicity of the stamp and the natural length scale of instability of the film.

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There are some additional critical issues of further approach will eventually lead to a perfect negative replica of the stamp which is nothing but simple embossing.

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Elastic Contact Lithography

Stamp Used: Simple Striped
Stripe width: $1.5 \mu\text{m}$
Periodicity: $3.0 \mu\text{m}$
Stripe Height: 200 nm



$\lambda \sim 3H$



- Height = $451 \pm 18.82 \text{ nm}$
- Height = $427 \pm 7.2 \text{ nm}$
- Height = $309 \pm 18.82 \text{ nm}$
- Height = $217 \pm 7.2 \text{ nm}$
- Height = $155 \pm 7 \text{ nm}$

- The structures form based on the commensuration of the periodicity of the stamp and the natural length scale of instability of the film.

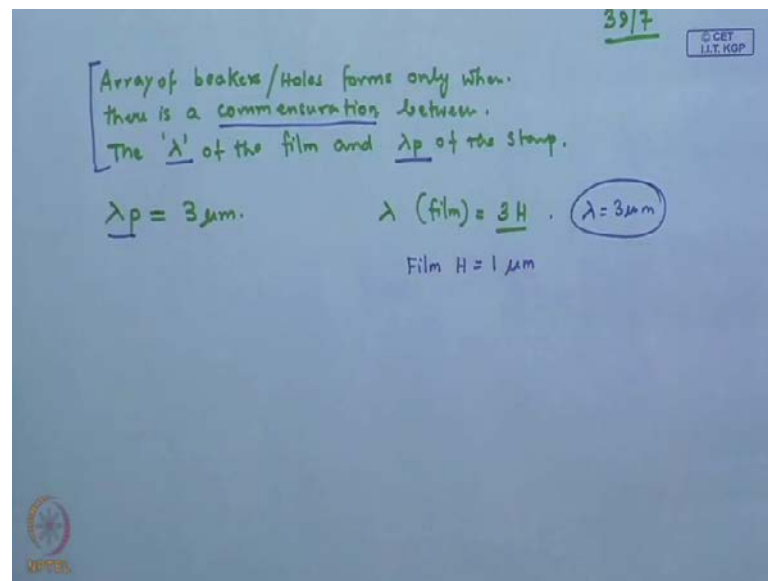
[What it means, for a $3 \mu\text{m}$ periodicity stamp, such structures can be obtained only if the film thickness is $1 \mu\text{m}$]
(for $1 \mu\text{m}$ film, length scale of instability is $3 \times 1 \mu\text{m} = 3 \mu\text{m}$)

Langmuir, 22, 7066, 2006

There are some critical approaches, so using a single stamp a simple grating stamp like this, by wearing the inter surface separation distance now you have the potential of generating a variety of structures so align columns, positive replica with higher feature height as compared the stamp, array of two be hole etcetera.

The important thing to note is that simply by taking a stamp and an elastic film will not give you this structure. This structure people are working on the theoretical aspect there is some complex bifurcation in the energy di energy land space which relates to this formation of a to be structure but the issue is fully, not fully result t a. What is important is that these order array of decades of tiny holes,

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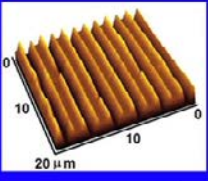
This forms only when there is a commensuration between the lambda of the film and lambda p of the stamp, what does it mean? Suppose you have taken a film, a stamp with lambda p equal to 3 micron. What is lambda of the film? If you use a flat Contactor it is going to be 3 H, this short wave instability for realistic contact Lithography a Elastic instability.

So, if you take a film now then only lambda becomes 3 micron, so statement what this statement means; that there is a commensuration between lambda and lambda p, implies that this type of a order to be structure forms, from a simple one de patterns stamp only when lambda p and lambda matches. So, in this particular case these structure resulted in a film which had thickness of roughly 1 micron using a stamp which was of the, which had a thickness which had a periodicity of 3 microns.

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Elastic Contact Lithography

- The structures form based on the commensuration of the periodicity of the stamp and the natural length scale of instability of the film.
- If there is a mismatch between the length scale of patterns and periodicity of stamp, various other structures form.



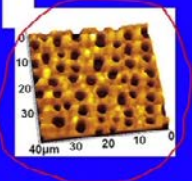
Stamp Periodicity: $3\ \mu\text{m}$
Film thickness: $480\ \text{nm}$

Double Periodic Structures



Stamp Periodicity: $3\ \mu\text{m}$
Film thickness: $4.2\ \mu\text{m}$

Missing Stripes



Stamp Periodicity: $3\ \mu\text{m}$
Film thickness: $1.3\ \mu\text{m}$

Disordered array of holes

M. Gonuguntla, R. Mukherjee, A. Sharma: *Langmuir*, 22, 7066, 2006.
Journal of Nanoscience & Nanotech., 7, 1744, 2007.

What happens is if there is a mismatch between these two length scale, you do not get a this type of a beautiful orders structures for example, here is a case where the film thickness was 1.3 micron and the stamp periodicity was three microns, so you see you are still close to that formation of 2 D holes but the alignment is not good, it is slightly distorted.

However, if there is a significant difference between λ and λ_p for example, here the film thickness is 500 nano meter and the periodicity λ_p is 3 micron.

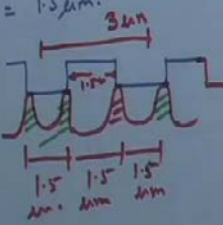
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Array of beakers/Holes forms only when there is a commensuration between. The ' λ ' of the film and λ_p of the stamp.

$\lambda_p = 3\ \mu\text{m}$ $\lambda(\text{film}) = \frac{3H}{2}$ $\lambda = 3\ \mu\text{m}$
 Film $H = 1\ \mu\text{m}$

$\lambda_p = 3\ \mu\text{m}$
 $\lambda = 1.5\ \mu\text{m}$



$\lambda = 1.5\ \mu\text{m}$
 $\lambda_p = 3\ \mu\text{m}$

An Elastic Film, even in the presence of a Confining Pattern (stamp) \Rightarrow Rather faithfully follows the Natural Instability Length Scale.

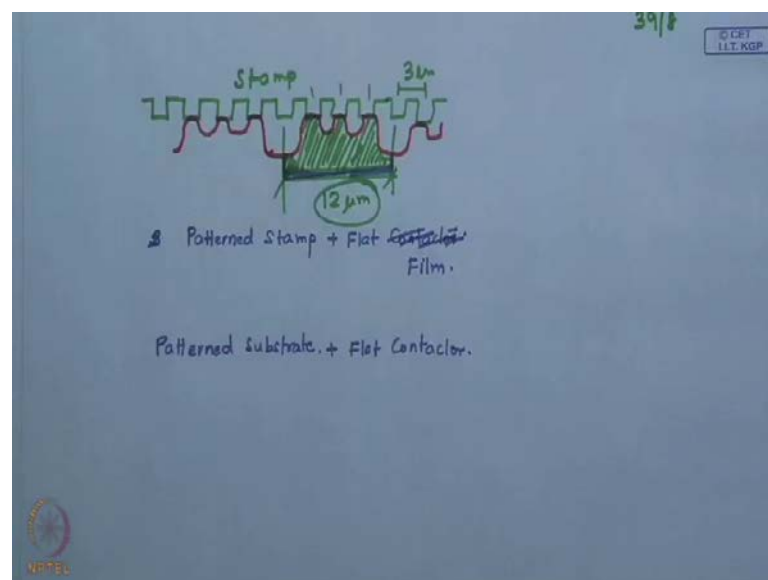
$\lambda = 13\ \mu\text{m}$
 $\lambda_p = 3\ \mu\text{m}$

So, in this case λ_p is 3 micron and λ equal to 1.5 micron so it is roughly half, so what you see it that the double periodic structure emerges or in other words this is the type of structure that forms, so **here the and** if you check at the periodicity of this peaks it still is 1.5 micron. So, the signature of the **that emerge the** information that emerges, out so this is 3 micron the stamp periodicity so this gap is roughly 1.5 micron.

So, if you now look at the periodicity of this double periodic structures what has formed over here, so would be the periodicity it is roughly of the order 1.5 micron. So the take home message is that, an elastic film even in the presence of a confining stamp or a pattern stamp, rather faithfully follows the natural instability length scale. Similarly, if you take a scenario like this film thickness is 4.2 micron stamp periodicity is 3 micron it does not match so film thickness is much higher, so here what will happen the λ are corresponding to the film thickness is of the order of 13 micron and λ_p is 3 micron.

So see what type of structure has appeared, you see that you get a strange type of a structure where you have a three strips and then one strip is missing.

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So, three strips pipes under or the stamp protrusion would essentially mean, so this is let us say the stamp and what you see is a very interesting structure this, this, this and this one is missing, then again you see these this, this the next one is again missing, so it continues like this. What it implies, now the periodicity of the stamp is 3 micron so 1 2

and 3 so from here to here it is actually, 12 micron and you see the natural periodicity is roughly 13 micron, so here as if this whole individual structure now behaves as one single elastic instability structure so that it still obeys the natural length scale of instability under this type of situation.

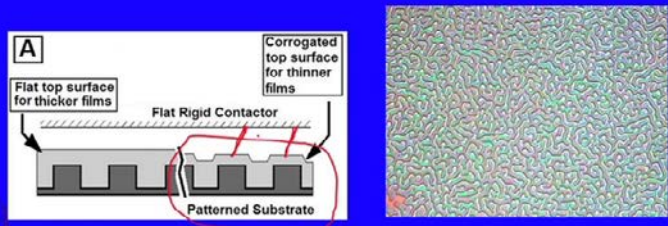
So, this in an natural the method of elastic contact lithography, the other approach that had been taken was to instead of using a combination, so here the classical sense we had a pattern stamp and a flat Contactor, the other combination was to use a patterns substrate and **sorry** a patterns stamp and a flat film was the condition we talked about, the other alternative would be to use a pattern substrate and a flat Contactor. So, in the previous lecture we talked about using a topographically pattern substrate for swain coating or something, so the moment you take a topographically pattern substrate you now get a film which is undulating top surfaces.

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Elastic Contact Lithography on Patterned Substrates

- Surface of a soft, thin elastic film becomes unstable with formation of random isotropic labyrinth structures when brought in contact proximity to another rigid flat surface.
- The patterns form at a length scale (λ) ~ three times the film thickness. ($\lambda \sim 3H$)

A new strategy to align and order these structures by using a patterned substrate.

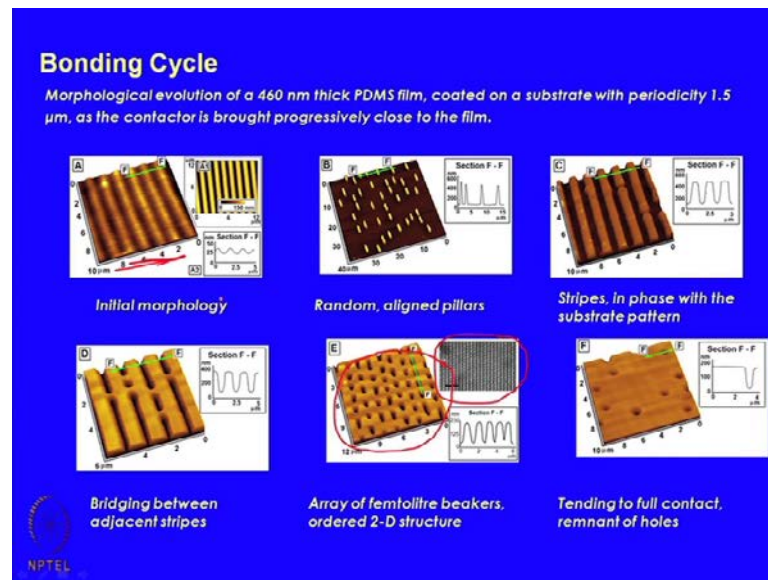


The diagram, labeled 'A', shows a cross-section of the lithography process. It features a 'Patterned Substrate' with rectangular features. A 'Flat Rigid Contactor' is shown in contact with the top surface of the substrate. The top surface of the film is divided into two regions: a 'Flat top surface for thicker films' on the left and a 'Corroged top surface for thinner films' on the right, which is highlighted with a red circle. To the right of the diagram is a micrograph showing a complex, interconnected, labyrinthine pattern of the film's top surface.

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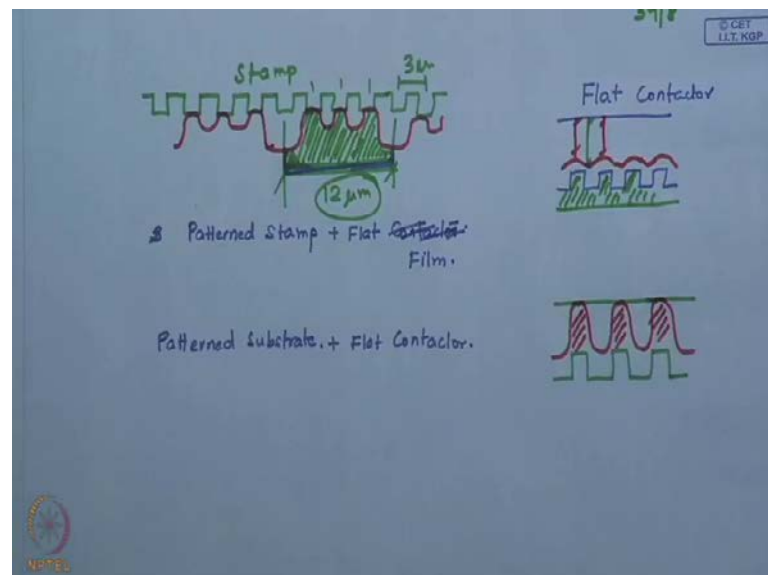
So, same thing happens here, if you now use a patterns substrate an on to which you coat your elastic film you have undulating a film with an undulating top surface and therefore, the inter surface separation distance over here, over these locations about the undulation is lower as compared to other areas, so this is expected to give a sort of a some sort of directionality preferential directionality.

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And if you now do an approach sequence, you see that the morphological evolution sequence is almost the same, you again see the array of holes appearing in this case and this optical micro graph shows how well organized and large area these structures are. So this is the morphology of the initial film.

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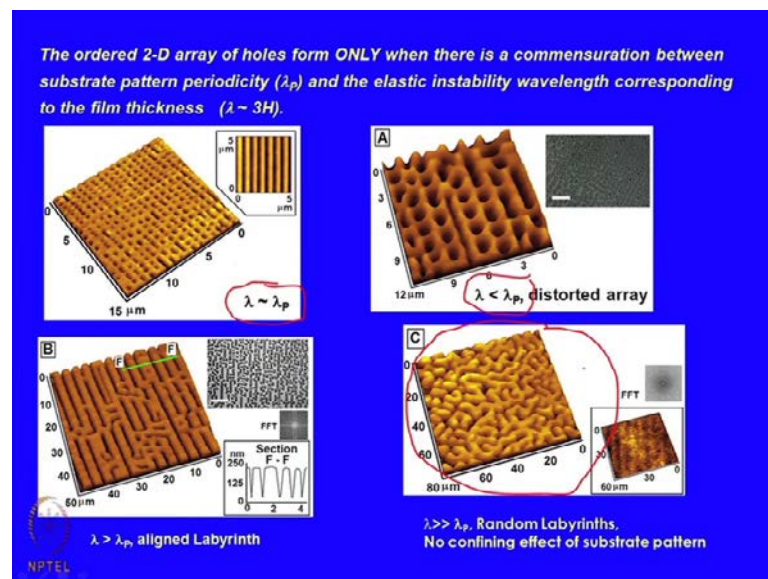
And you need to understand that here you are using a flat Contactor, and a film with undulating top surface so these zones you have preferential attraction as compared to the

other zones and therefore, it result in the formation of a again align structure using elastic instability.

So, this is a level where do you see, so this is interesting in the sense here the effect of the substrate sort of bleeds shoe the film and resulting in first sort of replica which morphologically resembles the sub steady self, this is the substrate this is what happens when you the columns forms because, of a adhesive attraction. So as the stamp sort of approaches one would expect first that so this is the substrate, and first the columns would form like this again this, I am not very sure whether it is a positive replica.

But when you get to this stage, the height of the features is much higher than the original height of the substrate patterns and then a subsequent approach of the flat Contactor again starts to the bridging, and eventually you get to this array of holes and final, before final conformal contact is achieve that resulting completely flat surface. Here also this commensuration between lambda and lambda p is important

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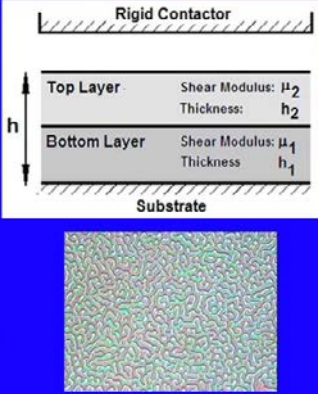
So if there is commensuration, you get an aligned perfectly aligned array of this 2 D holes, slightly lambda slightly lower than lambda p de sorted array lambda higher than lambda p you tend to back to the align structures but labyrinth but they sort of align because of the presence of the subsurface substrate pattern substrate or align substrate.

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Pattern Miniaturization with Elastic Bilayers

$M = \mu_1 / \mu_2$ $H = h_2 / h_1$

- For an elastic bilayer system, the length scale of instability depends on the thickness ratio (H) and the shear moduli ratio (M).
- For having a softer film on top of a harder film, the ratio reduces and passes thru' a minima.
- For harder film on top, the opposite trend is observed. ($R_F > 3$)
- A co-operative instability evolves at the interface.



Rigid Contactor

Top Layer Shear Modulus: μ_2
Thickness: h_2

Bottom Layer Shear Modulus: μ_1
Thickness: h_1

Substrate

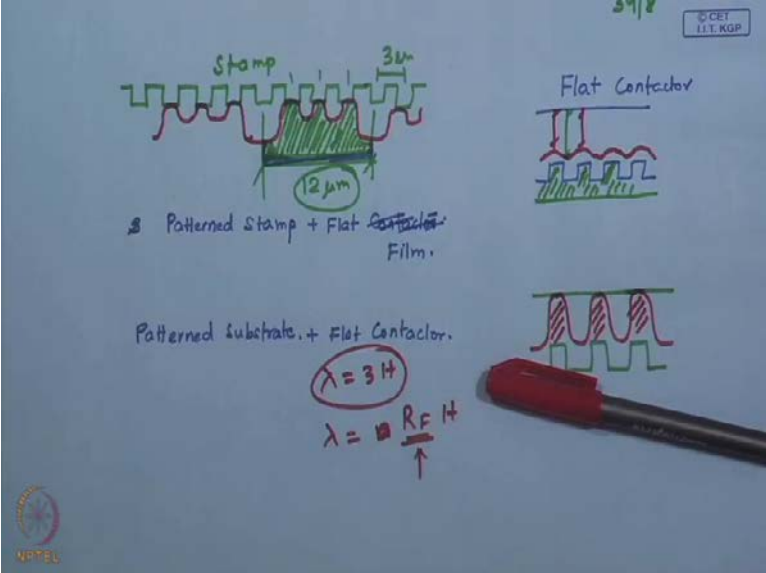
Morphology remains same

Rabibrata Mukherjee, R. Pangule, A. Sharma and G. Tomar *Adv. Funct. Mater.* 17, 2356, 2007

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And when the film thickness is much higher it completely sort of disobeys the presence of the substrate pattern, and you get these labyrinth. The other interesting aspect is that, you can tailor this lambda equal to three h scaling by using an elastic bi layers where this scaling ratio.

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Stamp 3μm 12μm

Flat Contactor

Patterned Stamp + Flat Substrate Film.

Patterned Substrate + Flat Contactor.

$\lambda = 3H$

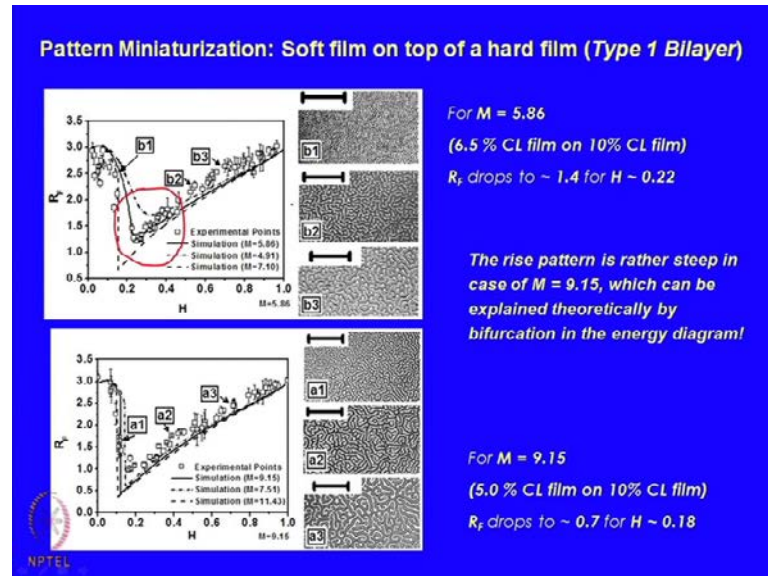
$\lambda = R_F H$

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Now becomes r m is function, lambda is an a function of R F into H, and this R F tailored between any values between 0.5 and 8 depending on the configuration of the elastic bi layer, and which becomes a function of the ration of the share module I of the

two layers as well as the thickness of the two layers, and in principle becomes possible to achieve a much reduced R F.

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So, you can now start off with a film which is much thicker and can achieve smaller features, recently this concept of the elastic bi layer has also when extended with an ECL to generate sub micron literals will structures with elastic films.

So I think a I will stop this lecture, so in this lecture I gave you a glimpse of a new kind of an instability which is elastic contact instability, which is a absorb in elastic films in contact proximity to Contactor or a stamp. And this is induced by inter surface adhesive attraction and is opposed by the elastic restore in forces, and we have use we have shown that how this form of instability, and we use for a, as a viable pattern in technique by combining the concepts of again top down method like using topographically patterns stamp or a substrate.

The uniqueness of elastic contact lithography ECL is that, due to complex bipartition in the energy scenario, it is one of the rare methods which allows you the formation of a ordered 2 D structures using a simple same one D pattern stamp. So I think I will stop here, thank you.