Oscillating Cracks in Glassy Films on Silicon Substrates

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Abstract

An experimental study of cracks that develop during the annealing of hydrogen-rich amorphous silica films deposited on crystalline silicon substrates shows an interesting duality of behaviour. Cracks in films deposited on cubic (001) silicon surfaces tend to form in two principal directions, cube-edge directions such as [100] or diagonal directions [110] and, while the [100] cracks are straight, those at 45° in [110] directions have an exactly regular oscillating structure. Sections through the wafers show that the oscillating cracks have a complex three-dimensional structure that extends through the glassy film and into the underlying silicon substrate. While a complete theoretical treatment of the behaviour would be extremely complicated, a simple theory shows that an oscillating crack has a minimum energy per unit length for a particular wavelength and amplitude which depends upon the physical parameters of both film and substrate. When the energy at this minimum is compared with that of a straight crack, it is found that for certain parameter ranges it can be less than for a straight crack, so that the oscillating geometry is preferred. Because the elastic strain energy and surface energy of the crystalline substrate are anisotropic and the behaviour depends sensitively upon these parameters, it is not surprising that cracks in some directions are preferentially straight while those in other directions are oscillatory.

Keywords:

AMS Subject Classification:

1 Introduction

Electronic devices such as computer chips often make use of thin insulating films of glassy silica (SiO_2) deposited upon a crystalline silicon substrate. These films can develop cracks during fabrication or use, which may destroy the device. A study of such cracks is therefore of practical importance as well as being of considerable theoretical interest. As is described below, it is found that cracks in films deposited on cubic (001) surfaces of silicon may form in two principal directions, cube-edge directions such as [100] or diagonal directions [110] and, while the [100] cracks are straight, those at 45° in [110] directions have an exactly regular oscillating structure. While this effect is clearly related to the crystalline anisotropy of the silicon substrate, it is surprising that oscillatory cracks can apparently have a lower total energy per unit length than do straight cracks, at least under some conditions, and it is the purpose of the present study to examine the experimental evidence and then to provide a simple semi-quantitative theoretical explanation of the effect.

2 Experimental Study

The propagation of cracks has been studied extensively[1,2]. There has been work published on oscillating cracks in media such as rubber membranes under biaxial tension[3], in thin glass sheets subject to axial temperature gradients[4], and in crystalline silicon[5], but here we address the problem of oscillating cracks in a silica layer deposited on a crystalline silicon wafer.

The deposited silica layers used in this study were produced by the process of plasma-enhanced chemical vapour deposition (PECVD). Depositions were performed in an Oxford Instruments PlasmaLab 80 system using SiH_4 as a source of silicon and NO_2 as a source of oxygen. Samples were heated to 300°C during deposition and the RF power was held constant at 20W. The resulting silica layers contain as much as 30 atomic percent of hydrogen, which reduces the electrical insulating properties of the layer. To remove the hydrogen and improve the structural integrity of the deposited films they are normally subjected to a densification annealing process in which they are heated to a temperature as high as 1100°C for up to one hour.

This whole process results in a silica layer that is under high tensile stress, typically up to 1 GPa and it is this stress that can lead to cracking or delamination. While not all applications require such processing extremes, similar effects are also observed after processing at lower temperatures. Only recently has it been recognised that hydrogen plays an important role in this behaviour[6]. The silica films used in the present study were approximately 1.2μm thick and were typically annealed at about 1100°C. They exhibited a tensile stress when cooled to room temperature of about 750 MPa, this stress being evaluated from measurements of the curvature of the underlying substrate wafer. Subsequent cooling and heating of the samples induced only relatively small and reversible changes in stress.

After the development of cracks, their surface geometry was examined by optical microscopy and/or scanning electron microscopy (SEM), with selected samples also examined in cross-section using an FEI Quanta 200 ESEM Focussed Ion Beam (FIB) system. The composition of films, including hydrogen, was determined by heavy-ion elastic recoil detection (HI-ERD) using 200 MeV Au primary ions.

The films of interest are thus of nonstoichiometric silica of composition SiO_x with x less than 2 and some included hydrogen, and develop large tensile stresses during the annealing process. If a crack develops in the film and penetrates a short distance into the substrate, then the total energy of the system is reduced by the relaxation of elastic tension in the film. But account must also be taken of the increase in energy of the substrate due to the elastic compression and shear energy around the crack as well as the surface energy of the crack itself. If the film tension is high enough, then cracking will occur because it reduces the total energy. Since the silicon substrate is crystalline, its elastic properties are anisotropic and there is a similar anisotropy in surface energy, with low-index faces having the least energy. These anisotropies account in a general way for the existence of preferred crack directions and crack surface orientations in the silicon substrate and also explain the transition between straight and oscillatory cracks, as will be shown later.

Figure 1 shows an optical micrograph of cracking in a 1.2 μm silica (actually $SiO_{1.2}N_{0.4}$) film grown on the (001) surface of a silicon wafer and subjected to thermal annealing at 1100°C. Cracks in [100] and [010] directions are quite straight, while cracks in [110] and [1$\overline{1}$0] directions have a regular oscillatory form.
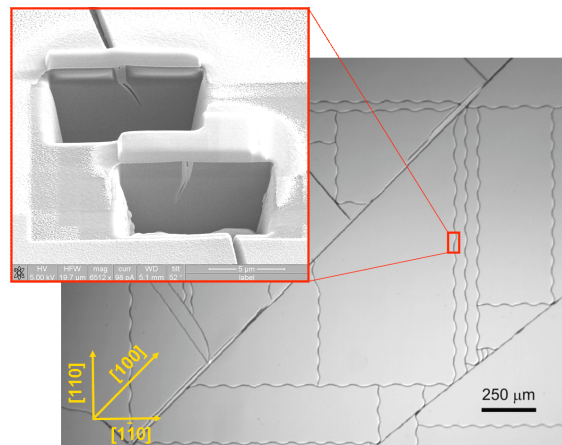

Figure 1. Optical micrograph of a typical cracking pattern in a silica layer grown by PECVD on the (001) surface of a silicon wafer after thermal annealing. Straight cracks run in [100] and [010] directions, while cracks in [110] and [1$\overline{1}$0] directions are oscillatory. The inset electron micrographs show the sloping nature of the cracks below the silicon surface. The lower of these two sections indicates how a sloping crack at less than the maximum angle resolves into two sections at energetically preferred slope angles.

When an oscillating crack is observed in cross-section, as shown in Figure 2, it is found that the crack plane is generally not normal to the surface. The slope angle of the crack is itself oscillatory so that the crack plane is always sloping inwards towards the axis of the crack alignment direction. This oblique angle is important in explaining the behaviour of oscillatory cracks, as will be shown. Even in straight [110] cracks on the (001) surface the crack plane is sloping rather than being normal to the surface, though in growth on some other crystal faces there can be straight cracks with crack plane normal to the surface.
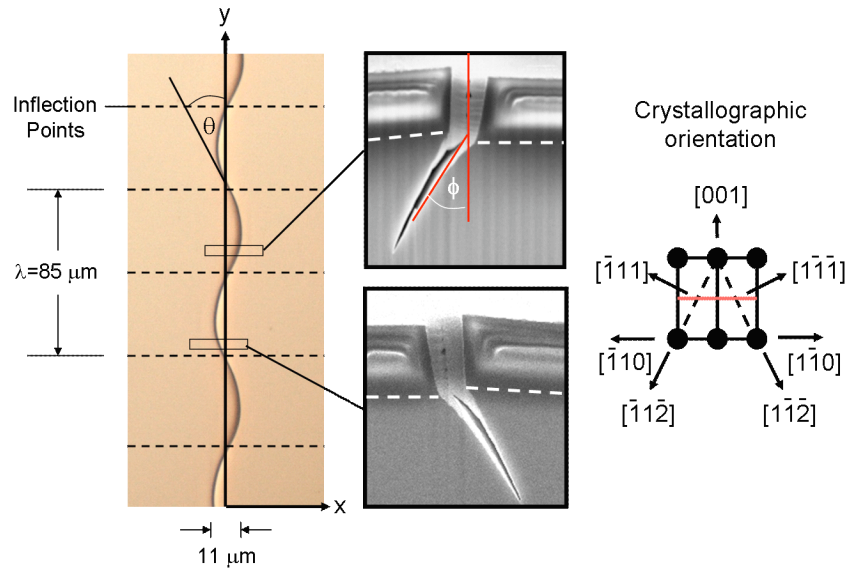
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Figure 2. Optical micrograph of an oscillating crack together with transverse sections as observed by electron microscopy after focussed ion beam sectioning, the crack cross-sections being partially filled with platinum to improve the image contrast. The interface between film and substrate in the sections is indicated by a dashed line. Note that the crack plane is always sloping inwards toward the axis of the track trajectory (y). Although not shown, the crack slope is nearly normal to the surface at the inflection points where the crack crosses the trajectory axis. Relevant crystal directions in the silicon substrate are also indicated.

3 Simple Theory

Some simple considerations explain why oscillating cracks can be preferred in some directions but straight cracks in others, and a slightly more formal theory gives semi-quantitative predictions of wavelength and amplitude that are in general agreement with experiment. A crack driven by the tensile strain in the deposited layer will propagate in such a way as to minimise its energy per unit length. We begin with the fact that slight anisotropy in the crystal substrate causes the crack to be inclined at an angle to the surface that is not 90° and also fixes its depth in such a way as to minimise the total elastic and surface energy. If the crack is oscillating rather than straight, then two things happen. The first is that, because the crack plane is observed to always slope inwards towards the axial direction of the crack propagation, its area per unit crack length is less than that for a straight crack. Provided the crystal anisotropy is not too large, this results in a reduction of crack surface energy. The second effect is that elastic relaxation in the film and the coupled elastic compression of the substrate normally extend for a distance on either side of the crack about equal to the crack depth. When the crack is oscillating and the wavelength is not extremely long compared with the crack depth, then the elastic relaxation fields from neighbouring peaks begin to overlap and this reduces the magnitude of the negative elastic energy driving the crack. Taking both effects into account, there is a small range of amplitudes and wavelengths for the crack in which its energy per unit length is smaller than that of a straight crack, so that an oscillating crack is energetically preferred. This result depends sensitively upon the anisotropy of the crystalline substrate, so that straight cracks are preferred in some directions and oscillating cracks

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in others.

It is helpful to set out this argument in slightly more formal terms, since it then gives some semi-quantitative predictions. We assume that the substrate is semi-infinite in thickness so that it does not bend, that the depth of the crack measured obliquely and including the film thickness is L, and that the crack direction oscillates with amplitude a and wavelength λ. If the crack shape is $y = a \sin(2\pi x/\lambda)$, then the surface length of the crack per unit distance in its axial direction is

$$F = \frac{1}{\lambda}\int_0^\lambda \left[1 + \left(\frac{dy}{dx}\right)^2\right]^{1/2} dx \approx 1 + \left(\frac{\pi a}{\lambda}\right)^2 . \tag{1}$$

Because of the crack slope, its length at the base has a replaced by $a - L \sin \phi$, where ϕ is the angle between the crack plane and the normal to the surface . The area of the crack per unit axial length is thus

$$S \approx L\left[1 + \delta\left(\frac{\pi a}{\lambda}\right)^2\right] , \tag{2}$$

where

$$\delta \approx \frac{1}{2}\left[1 + \left(1 - \frac{L}{a}\sin\phi\right)^2\right] \leq 1 . \tag{3}$$

The average surface energy of the crack per unit length along the crack is then

$$E_1 = \frac{\gamma_1 E_S S}{F} , \tag{4}$$

where $\gamma_1 E_S$ is the surface energy per unit area and $\gamma_1 \approx 1$ is an allowance for crystalline anisotropy.

The elastic strain energy is rather more difficult to calculate because of the strain overlap. In the substrate the principal strain decay is exponential with distance from the crack and has a characteristic length equal to the crack depth L. This strain variation is then imposed upon the film at the interface. There is a related strain variation in the film with decay length equal to the film thickness, and this is imposed upon the substrate at the interface but, because the film thickness is much smaller than the crack depth, this has much less influence, as can be seen from Figure 1 where the film crack walls essentially extend those of the substrate crack without much slope change. If $E_E < 0$ is the strain energy per unit length for a straight crack, taking account of both the negative contribution of strain relaxation in the film and the positive contribution of strain imposed on the substrate, then an oscillating crack with very small amplitude and very large wavelength will have strain energy $\gamma_2 E_E$ per unit crack length, where $\gamma_2 \approx 1$ is an allowance for anisotropy. When the wavelength λ is no longer very large compared with the crack depth L, however, the elastic fields overlap and eventually reduce to partial relaxation within the amplitude region a together with more complete relaxation in a region of characteristic length L on either side of the oscillation envelope. The reason that relaxation within the oscillation envelope a is only partial is that the elastic strain fields from neighbouring wavelengths of the oscillation overlap and this reduces the relaxation energy.

This behaviour can be approximated to give an expression for the average elastic energy per unit length of crack of the form

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$$E_2 = \frac{\gamma_2 E_E}{F}\left[1 + (F-1)e^{-\beta L/\lambda} + \frac{a}{L}e^{-\lambda/\alpha L}\right], \qquad (5)$$

where α and β are numerical parameters of order π. If the wavelength λ of the crack oscillation is large compared to the crack depth L, then (5) gives the result $E_2 \approx \gamma_2 E_E$, while if the wavelength is very short (5) gives $E_2 \approx \gamma_2 E_E(1+a/L)/F$, in agreement with the requirements of the theory.

The total average energy of the crack per unit length along its path is then $E=E_1+E_2$ and the preferred crack pattern can be determined by minimising E with respect to the oscillation parameters a and λ. This minimisation can really only be done numerically, and so numerical values must be assigned to the parameters E_S, E_E, ϕ, γ_1, γ_2, α and β. While this is a large array of parameters, most can be eliminated if we adopt simple scaling conventions. For simplicity we ignore crystal anisotropy and take $\gamma_1 = \gamma_2 = 1$ and set $\phi = 36°$ to agree with observations. The elastic energy E_E can be assigned the value -1 and the crack depth L the value 1, since units are irrelevant to the minimisation. The variables are then a/L and λ/L. The values of the parameters E_E, α and β can then be varied to examine their influence.

TABLE 1

E_S	α	β	a/L	λ/L	E	$E_{straight}$
0.9	1	2π	1.02	21.2	-0.1025	-0.1
0.9	0.5	2π	1.03	21.3	-0.1025	-0.1
0.9	π	2π	0.61	4.4	-0.1037	-0.1
0.9	1	π	1.00	10.6	-0.1095	-0.1
0.5	1	2π	1.10	53.8	-0.5004	-0.5

Numerical minimisation of the total energy $E=E_1+E_2$ for a variety of parameter values gives the results shown in Table 1. It can be seen that in all cases listed the energy per unit crack length E is slightly below the straight crack energy $E_{straight}$, though the difference is less than about 10%. This means that anisotropy in the elastic parameters or in the surface energy, as reflected in the parameters γ_1 and γ_2, could easily overcome this advantage and lead to a straight crack being energetically preferred. The predicted values of the relative oscillation amplitude a/L are all close to 1, which is smaller than the observed value by about a factor 5. The predicted ratio λ/a ranges between about 7 and 50 for extreme parameter values and thus is not inconsistent with the observed value near 8. The range of parameter values for which an oscillatory crack is favoured shows that the theory is robust and should provide a foundation upon which a true quantitative treatment can be based.

4 Extensions of the Theory

This theoretical approach allows semi-quantitative predictions to be made about some other aspects of crack behaviour. Consider first the dependence of oscillation wavelength and amplitude on the thickness d of the silica film. The simplest case is that in which the substrate is of effectively infinite thickness, which means in practice that it is thick enough that the stress in the film does not result in any appreciable substrate curvature. All the parameters describing the elastic and surface properties of

film and substrate are assumed to be given, so that film thickness is the one input parameter. Dimensional analysis shows that the elastic moduli of the film and substrate must appear as a ratio and, while the dimensionless initial elastic strain in the film is also certainly relevant, this is assumed to be invariant. It then follows that crack depth and width, and oscillation amplitude and wavelength, must all be proportional to film thickness d.
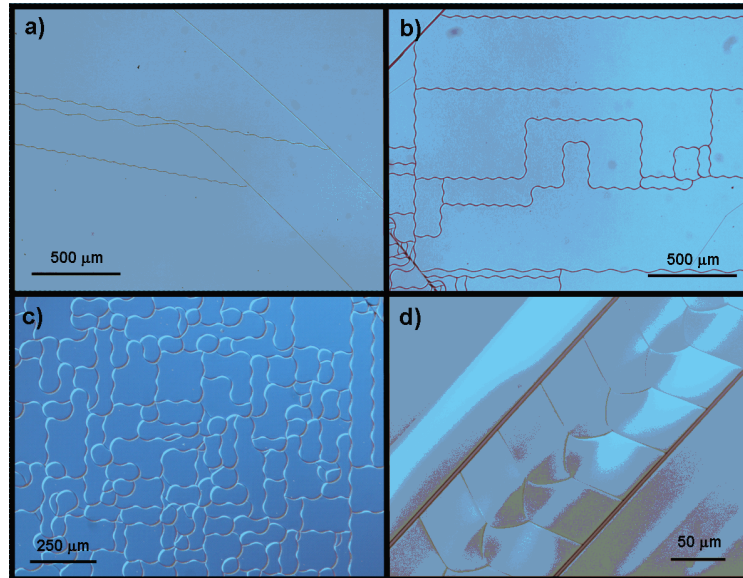

Figure 3. Examples of crack interactions in $SiO_{1.2}N_{0.4}$ films on (100) silicon. Panels (a) and (b) show simple interactions, while (c) and (d) show two different types of quasi-regular crack arrays, described as (c) "chain mail" and (d) "snake skin" respectively. Panels (a)–(c) are from 1200 nm films annealed for 1 hour at 1100°C, whilst panel (d) is from an 855 nm film annealed for 1 hr at 600°C.

When the substrate thickness D is not effectively infinite, this introduces another length parameter, which appears in experiment as a curvature of the substrate caused by the film tension. The simple proportionality described above then no longer applies and oscillation wavelength and amplitude are proportional to $d.f(d/D)$ where f is an unknown function. Consideration of the physical effect of substrate curvature shows that $f(d/D)$ is constant for small values of d/D and then decreases as d/D increases because the substrate curvature reduces the tensile strain in the film and applies compressive strain to the substrate near the interface. In the limit $d/D \gg 1$ the film dominates the system and can relax completely. There is then no substrate curvature and a high compressive strain through the whole substrate thickness so that no cracking occurs. The whole system is, however, made more complex if the film composition is not uniform throughout its thickness, as could well occur in thick films because of the deposition and annealing process. This would then introduce a further length parameter associated with the non-uniformity in the film and render the scaling conclusions above invalid.

The theoretical approach outlined here also gives some understanding of the interaction between cracks. In general terms, the elastic relaxation field around an existing crack makes it energetically unfavourable for another crack to nucleate within a distance from the existing crack rather greater than $2L$, so that it is possible for an ordered arrangement such as those shown in Figure 3 to develop under the influence of a slight gradient in one or more parameters. The slope of the crack also influences

the way in which it is deflected when it approaches another crack. Detailed analysis of these patterns is outside the scope of the present paper but is the subject of an ongoing experimental and theoretical study.

5 Conclusions

This theory developed here involves extreme simplification of the true situation and so can be expected to give semi-quantitative, results. The crystalline anisotropy of the elastic relaxation in the substrate has been ignored in formulating E_E, as also has been the anisotropy of surface energy in E_S. Also ignored has been the fact that when the mean crack slope is less than its maximum value ϕ, a crack is observed to resolve into two sections with slopes 0 and ϕ respectively, as shown in the lower cross-section in Figure 1, to give an average slope that varies nearly sinusoidally, as assumed for E_S in the model. This has the effect of reducing the effect of any surface energy anisotropy and so tends to support the approximations of the theory. Surface energy anisotropy cannot be realistically included, however, without also including elastic anisotropy, which is an extremely complicated matter for oblique cracks, so this has not been attempted here.

More importantly, an approach of this kind offers hope of a simple understanding of the physics underlying the complex and beautiful crack patterns sometimes found in these films and their crystalline substrates..

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