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# Indentation of a hard film on a compliant substrate: film fracture mechanisms to accommodate substrate plasticity

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**Abstract** In this paper we discuss various contact damage modes in thin TiN films on steel substrate with increasing load. To understand the displacement at different points along the depth of the film, we have used a TiN–AlTiN multilayered film in which each layer acts as a strain marker and we have also calculated the stresses theoretically using an elastic model of spherical indentation of a bi-layer. The study has helped to understand the physics behind different fracture phenomena, such as confinement of columnar sliding to the middle of the film, the genesis of lateral cracks during unloading, etc. We also emphasize the co-existence and competition of different modes of fracture in the film, rather than a single mode, at a particular combination of film thickness, substrate hardness and load and describe the way different modes interact in the spatial domains when they do coexist.

## Introduction

Hard coatings of TiN and related transition metal nitrides are being used in cutting tools and machine elements to protect the substrate by conferring increasing hardness, wear resistance and corrosion resistance. We briefly introduce here past work on the nature of deformation and indentations in columnar TiN coatings and various analytical approaches to model the competition amongst the different modes of slip and fracture.

Traditionally, indentation studies of thin hard coatings have been preoccupied with the need to derive properties of the coating from a response that includes the substrate [1, 2]. In more recent times the availability of depth sensing nanoindenters has made this less of a critical issue. However, even with depth resolution being as small as 1–2 nm it is sometime not possible to obtain data that may be unequivocally related only to the coating's plastic properties. In this connection, it has been shown [3] recently that what is meant by hardness (or contact pressure) in TiN coatings on metallic substrates falls into two domains. In the low load domain we can measure a true hardness over a depth range that is limited by the surface roughness: the rougher the surface (and this is less of a problem with sputtered films than with arc-evaporated ones) the greater the penetration necessary to avoid artifacts that lead to unacceptable scatter. Within this domain, load displacement behaviour may indeed be fitted to an elastic–plastic expanding cavity model applied to a material with the Young's modulus and hardness (yield stress) of bulk TiN, barring some effects of elastic and plastic anisotropy that play minor roles is what is effectively a strongly textured film. Higher loads, on the other hand, lead to completely different types of behaviour. In the simpler of these classes of responses, typified by thin films on hard substrates, one may continue to relate the penetration

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to an elastic plastic response, but plasticity here refers to the sliding [1, 4–6] at a critical shear stress of TiN columns aligned parallel to the indenter axis, into the soft substrate. Thus, we can model the response of the composite system as one that is compatible i.e., plastic displacements are continuous across the interface. The weakness of TiN columnar boundaries arising out of structural disorder that develops during deposition is well known. However, the ensuing propensity to slide is not necessarily a drawback since the alternative, as we show later, is a potentially more deleterious sequence of cracking.

Thicker films and softer substrates have been shown to lead to a variety of fracture events beginning with the well known "nested" [4, 7], edge cracks [7, 8] that appear parallel to the line of intersection of the indenter face with the sample surface. For long, these have been assumed to correspond to mode-I type bending cracks that extend through the thickness analogous to the cone cracks that appear at the periphery of spherical indentation. Another mode of failure is the so called bending crack [8, 9] that is driven by a plate bending stress and which appears at the interface, most prominently near the indenter axis and extending a short distance towards the free surface. In addition, the most recent observations have revealed a completely new failure mechanism [7, 8] consisting of inclined cracks that lie within the indentation and propagate at a large angle to the loading axis. A characteristic feature of all this cracking (see Fig. 1a) is that they are driven by incompatibility, i.e., by deformation of the film that is driven by plasticity in the substrate. The combined deleterious role of increasing film-thickness and strain mismatch was confirmed by an analytical model of elastic deformation using integral transforms [10] in which substrate plasticity was simulated by an artificially low modulus [11].

Our work so far has implied that when the substrate is strong and the substrate plasticity is limited, a thin hard coating renders the system benign against environmental invasion and contact damage. If the application demands wear resistance, a thicker coating is needed but which at best offers weak protection. When the substrate is however weak and prone to corrosion as in the case of soft metals, even thin coatings are inadequate as even at the lightest of tractions the substrate plasticity drives catastrophic fracture often through the thickness of the films. Excluding thin coatings on hard substrates all other coatings exhibit co-existence of different fracture modes, some benign and some catastrophic. In this paper we attempt to understand the complementarity of different modes of fracture in TiN coatings of metallic substrates subjected to indentation. We do this by extending our combined treatment of analytical modeling with focused ion beam machined cross sectioning of indentations.

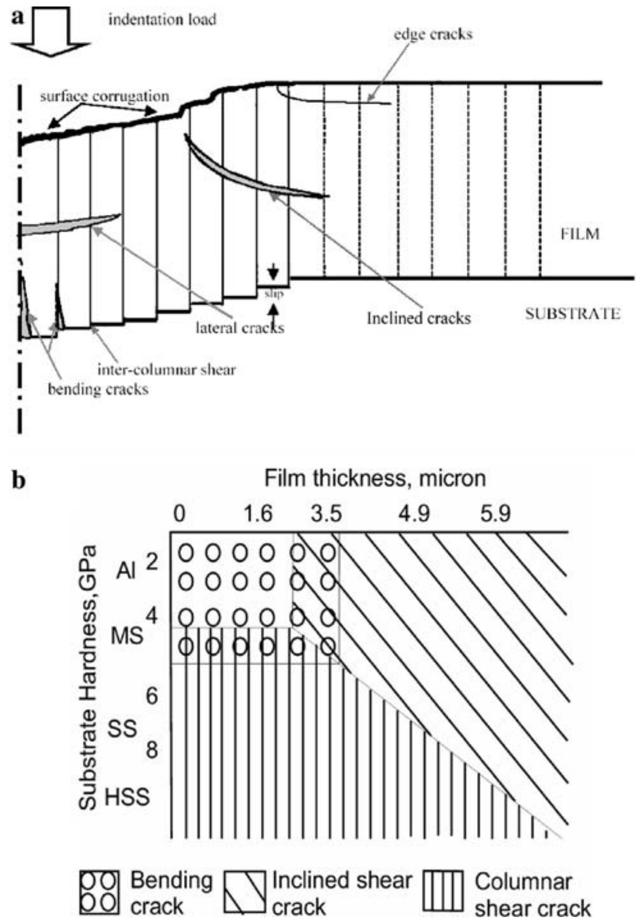


**Fig. 1** (**a**) Schematic diagram of possible fracture modes in a thin hard film deposited on a soft substrate (from Bhowmick et al. [7]). (**b**) Map of different failure modes as a function of film thickness and substrate hardness (MS – mild steel, SS – stainless steel, HSS – high speed steel) (from Math et al. [11])

## Experimental procedure

TiN films ranging from 1–10 microns in thickness were prepared by cathodic arc evaporation on to steel substrates. The substrates were first polished to an average surface roughness (*Ra*) of ~20 nm and then cleaned thoroughly with solvent and dried before being placed on a continuously rotating planetary holder inside the vacuum chamber. The substrates were heated using radiant heating to the deposition temperature of 350 °C. After the chamber was evacuated to a pressure of $1.3 \times 10^{-3}$ Pa ($10^{-5}$ Torr), the substrates were sputter-cleaned with Ar+ and coated with a thin interfacial layer of ~50 nm of Ti. Deposition of TiN was carried out in high purity nitrogen at a pressure of 2.6 Pa (20 mTorr). A negative bias voltage of 150 V was applied to the substrates during deposition. Multi-layers of TiN and AlTiN were deposited by using two targets of Ti and Al–Ti alternately and a substrate that remained stationary between each of them in sequence. Films displayed perfect <111> texture along the surface normal with column diameters of ~0.2–0.5 μm. Indentations were made with a Vickers pyramid at loads ranging from 1–10 N. Cross sections were made by focused ion beam machining at 2700 pA followed by fine polishing at 350 pA.

## Modeling

Calculations of stresses and crack driving forces were made using a model described earlier [10] and which is described here briefly. The stress solution for axi-symmetric, spherical contact of a half space is extended to a bi-layer by iteratively obtaining the instantaneous modulus as a function of ($\delta/t$) where $\delta$ is the penetration depth and '$t$' the specimen thickness. The biharmonic equation $\Delta\Delta\Phi(r,z) = 0$ (where $\Delta = \frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r} + \frac{\partial^2}{\partial z^2}$), which comes from stress equilibrium, has been reduced to an ordinary differential equation using the Hankel's transform technique. A stress function $\Phi^H(\xi, z) = (a_1 + b_1 z)e^{\xi z} + (c_1 + d_1 z)e^{-\xi z}$ has been taken to solve the ODE in Hankel's domain, $a_1$, $b_1$, $c_1$, $d_1$ being some constants and $\xi$ being a parameter. The form of the pressure distribution at the surface for a half space (i.e., $p \propto \delta^{3/2}$) is preserved. The remaining boundary conditions pertain to the continuity of stresses and displacements at the interface and the vanishing of stresses at an infinite distance from the contact. A schematic of the geometry of analysis and the relevant symbols is shown in Fig. 2. To simulate plasticity the substrate is assigned a modulus of 2 GPa [11].

## Results and discussion

We now examine the individual cracking modes by assuming that they are non-interacting, i.e., we apply the stresses obtained by the elastic analysis separately to the various failure mechanisms. This is a good approximation for nucleation but evidently becomes less valid as multiple cracking ensues. In the terminology of this article, $\tau_{rz}$ is the shear stress that promotes columnar sliding, $\tau_{\max}$ is the maximum shear stress which is used to calculate $K_{\mathrm{II}}$, the stress intensity factor for inclined cracks, while $\sigma_{rr}$ is the radial stress that drives bending/edge cracks whose axis lies normal to the radial direction.

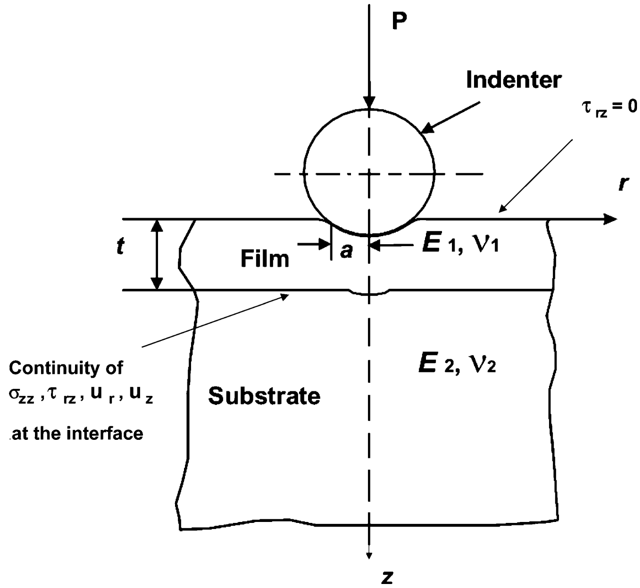


**Fig. 2** Schematic diagram of a film-substrate system indented by a spherical indenter with the boundary conditions at the free surface and at the interface. $t$ is the thickness of the film and $a$ is the contact radius (from Math et al. [11])

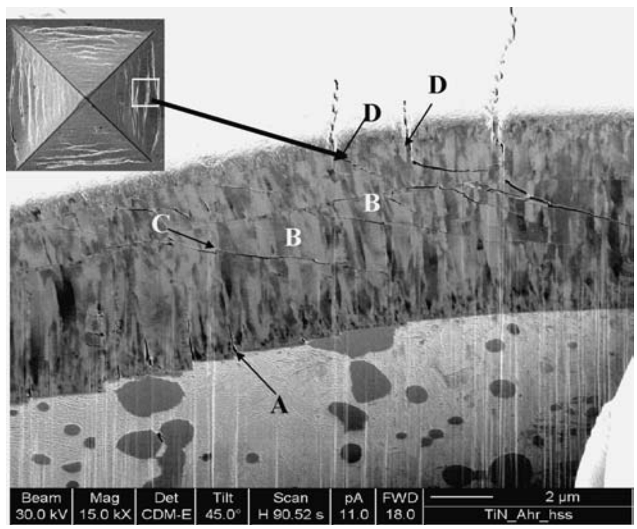


**Fig. 3** Existence of columnar shear (A) and inclined cracks (B) in a TiN film deposited on a HSS substrate, load = 10 N. Notice the step in an inclined crack (C). Edge cracks (D) are shallow and turn parallel to the surface at a depth of 1–2 μm

### Surface edge cracks

We have shown earlier [7] that edge cracks (Fig. 1a) nucleate only beyond a certain critical load. It may be seen from Fig. 3 that they do not extend significantly below the surface but instead branch almost parallel to the free surface after ~1–2 μm. This feature may be understood readily from the contours of one principal stress shown in Fig. 4. When the substrate is stiff ($E_{sub}$ = 200 GPa) $\sigma_1$ close to the surface and outside the contact is tensile and drops rapidly with increasing '$z$' and even becomes compressive as one approaches the interface. Figure 4b shows the trajectory of $\sigma_1$. Notice the tendency for the plane of maximum tensile stress to curve away from the vertical direction as is well known from the geometry of the Hertzian cone crack. When the substrate becomes softer the propensity for edge cracking increases, as seen from the magnitude of the stress contours shown in Fig. 4c, but in addition the depth at which the stress turns compressive also reduces. Bearing in mind that the films are under substantial compressive residual stress (in several GPa), clearly the zone of significant tension reduces dramatically as the substrate softens, even as the magnitude of the maximum tensile stress rises. These cracks curve at right angle and can interact with other modes leading to surface spalling (Fig. 4e). However they do not extend through the film thickness under any circumstances.

## Sub-surface deformation

It has been shown earlier [8] that for every substrate, depending on the yield stress (hardness), there is a transition TiN film thickness below which the dominant mode of stress accommodation is columnar sliding while transgranular inclined cracking is the principal mode of failure at higher thicknesses. The transition occurs for a high speed steel substrate at ~10 μm. Thus, as shown in Fig. 3, both modes co-exist in the present films. The question naturally arises: why is plasticity in the substrate not completely accommodated by columnar sliding in the coating, i.e., what is the driving force for transgranular cracks? To understand the complementarity of different failure modes, we look in detail at the stress distribution across the film thickness. We first note strain distribution by observing the layer displacement in multi-layered films of TiN–AlTiN, in which individual layers of TiN and AlTiN serve as strain markers. Figure 5 shows that columnar sliding initiates, not at the free surface, but at some depth below. In addition, the sliding is sometimes not carried through to the interface which, accordingly, displays no shear whatsoever. Figure 5b shows that in one case the columnar shear is initiated below the surface, increases to a maximum as it moves toward the substrate, and then disappears before reaching the interface. In order to understand the rationale for this behaviour it is useful to examine the contours of $\tau_{rz}$ with increasing load. It may be seen from Fig. 6 that $\tau_{rz}$ builds up to a maximum with increasing depth. Thus the greatest tendency for sliding lies somewhere in the middle of the coating. Thus, shear commences in a manner analogus to the propagation of a dislocation loop, but then may not propagate if the available stress drops too rapidly as one approaches the interface. Instead, the displacement is elastically accommodated as shown in Fig. 5b in which the relative offset of the layers gradually disappears as the interface approaches. The relative displacement may be accommodated by other forms of failure. When the substrate is strong and non-compliant and the scope for transgranular shear cracking is thus minimized, bending cracks at the film substrate interface (high tensile stress normal to the indentation) can accommodate the relative displacements between columns (Fig. 5b) by reducing the residual compressive stress which directly affect the columnar strengths. At a critical strain, one can readily envisage that cracking may ensue as shown in Fig. 5a. Indeed, this behaviour is exactly analogous to the behaviour of dislocation pile-ups in classical plasticity wherein brittle materials can initiate slip which is unable to cross a grain boundary or other barrier and, instead, nucleates a crack after the mechanisms proposed by Stroh or Cottrell.

At the surface there is little sliding but instead, the layers appear to bend. This feature prompted us to examine whether plasticity could be responsible for shape change. Figure 7 shows the second stress invariant $J_2$ which is related to the von Mises yield criterion. It may be seen that $\sqrt{J_2}/k$, where k is the yield shear strength of the film ($k = \sigma_y/\sqrt{3}$, where $\sigma_y$ (the yield tensile strength of the film) = 15 GPa, which is consistent with reported value [2]), is high at the surface, decreases just below the surface upto a certain depth, depending on the load and then again rises with depth (Fig. 7a) for a compliant substrate. The fact that $\tau_{rz}$, responsible for columnar shear is also close to zero at the near surface region (Fig. 6) explains why there is permanent bending but no columnar shear in the near surface region as seen in Fig. 5.

We now address the issue of simultaneous columnar shear and inclined cracking. Since the latter is a product of incompatible flow, (in our elastic stress calculation, we have taken compatibility at the interface as a boundary condition, which becomes invalid when the substrate becomes plastic. The incompatibility in displacements of hard film and soft substrate causes the film to deform more) it is natural to ask why cracking should occur if columnar sliding can simultaneously take place. A possible answer lies in the fact that the sliding stress very likely varies from column to

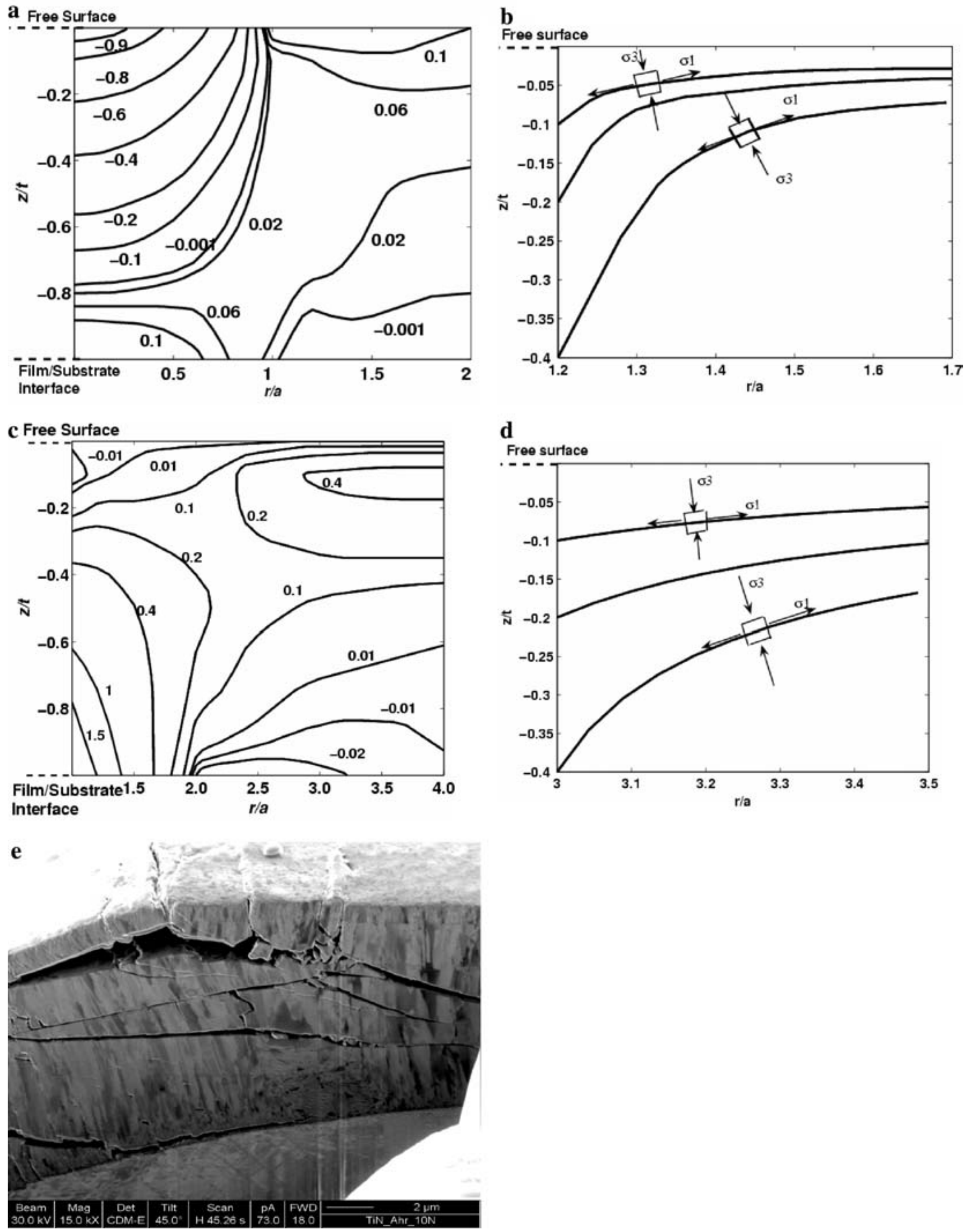


**Fig. 4** (**a**) Principal stress ($\sigma_1/|p_0|$) contour in the film, $p_0$ being the pressure exerted by the indenter at the center of indentation, $E_{\text{sub}}$ = 200 GPa, $E_{\text{film}}$ = 400 GPa, load = 1 N, (Young's modulus of indenter = 1140 GPa, Poisson's ratio = 0.07). Positive $z$ direction is taken upwards (from Math et al. [11]). (**b**) Principal stress ($\sigma_1$) trajectory in the film. $E_{\text{sub}}$ = 200 GPa, $E_{\text{film}}$ = 400 GPa, load = 1 N, (Young's modulus of indenter = 1140 GPa, Poisson's ratio = 0.07). Positive $z$ direction is taken upwards. (**c**) Principal stress ($\sigma_1/|p_0|$) contour in the film, $p_0$ being the pressure exerted by the indenter at the center of indentation, $E_{\text{sub}}$ = 2 GPa, $E_{\text{film}}$ = 400 GPa, load = 1 N, (Young's modulus of indenter = 1140 GPa, Poisson's ratio = 0.07). Positive $z$ direction is taken upwards (from Math et al. [11]). (**d**). Principal stress ($\sigma_1$) trajectory in the film. $E_{\text{sub}}$ = 2 GPa, $E_{\text{film}}$ = 400 GPa, load = 1 N, (Young's modulus of indenter = 1140 GPa, Poisson's ratio = 0.07). Positive $z$ direction is taken upwards. (**e**) Surface spalling in a TiN film deposited on a SS substrate, load = 10 N

column. Indeed the spacing of steps at the interfaces is typically ~ μm whereas the individual column diameter is 0.2–0.5 μm. This difference in strength can arise both, from the structure of the boundary (these are essentially tilt boundaries with a common [111]) as well as its local orientation. For example columnar

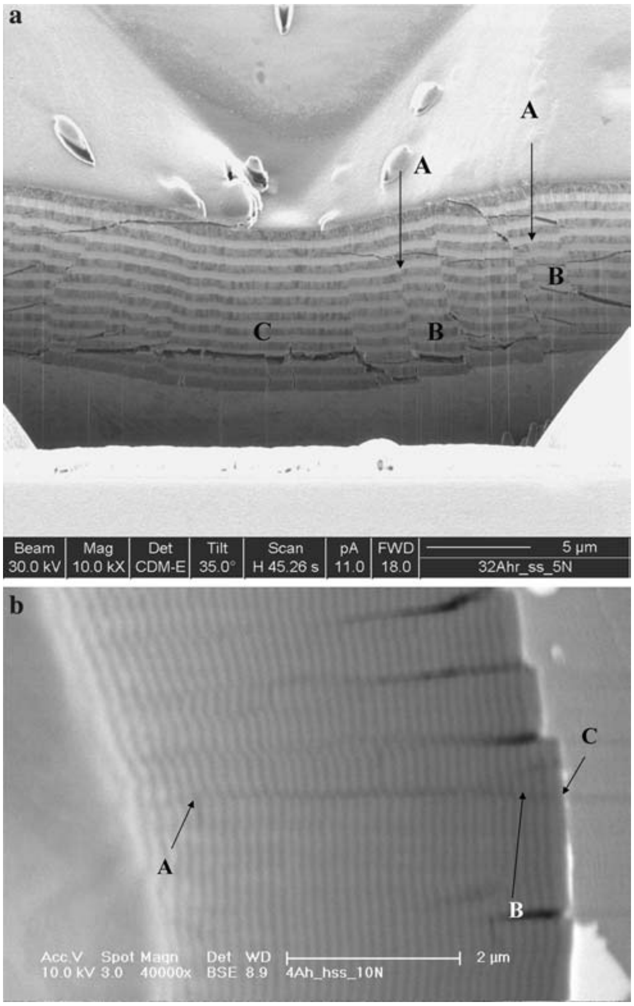


**Fig. 5** (**a**) Columnar shear in a TiN–AlTiN thin film on SS substrate, which starts at subsurface at A and ends in a crack further towards the interface (B), load = 5 N. Lateral crack (C) is also observed in the film. (b) Columnar shear in a TiN–AlTiN thin film on HSS substrate, load = 10 N. Shear begins at A and vanishes at B. There is no step at the interface (C)

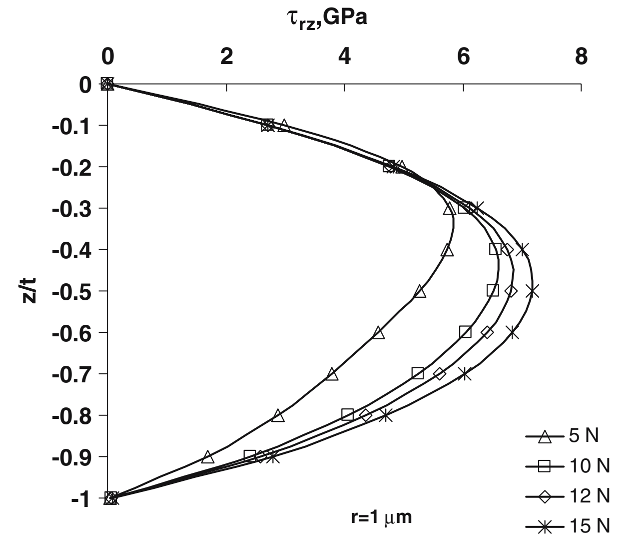


**Fig. 6** $\tau_{rz}$ in a 10 μm thick film as a function of depth at a radial distance = 1 μm, $E_{\mathrm{sub}}$ = 2 GPa, $E_{\mathrm{film}}$ = 400 GPa

boundaries are not always vertical and occasionally terminate when one grain over-grows its neighbour. If one looks at the shear stress $\tau_{rz}$ at a single point within the film (Fig. 6), then as the indentation depth increases, $\tau_{rz}$ rises to a maximum (at $r = 0.75$ a) and then falls as expected given that it must vanish for symmetry reasons when $r = 0$ (at the indenter axis). With increase in contact area a fixed point in film space moves closer to the $\tau_{rz} = 0$ axis. Thus, if a column does not fully slide from surface to interface where $\tau_{rz}$ is a maximum, it will clearly not do so with further increase in load since the shear stress remains almost constant at that point as it effectively moves closer to the $r = 0$ plane, thereby leading the way for other cracks to nucleate. Note: such competition is expected only near the transition thickness at which columnar shear gives way to inclined cracks and when $\tau_{rz}$ becomes large enough to initiate columnar shear but does not reach its critical value throughout the film thickness. Figure 6 shows that for a TiN film with 10 μm thickness, the value of shear stress through out the depth of the film is well below the critical value of 4 GPa [2] for column shear ($\tau_{\mathrm{critical}}$) except at the middle. As we have shown in earlier work [8], films of less than 8 microns on high speed steel only display columnar shear, while a film that is as thin as 1 micron on a soft aluminium displays inclined and bending fracture with little in the way of column sliding.

It is also clear from observations of the interactions between sliding and transgranular cracking that the inclined cracks arise during loading and not due to residual stresses that accumulate during the unloading cycle. For example, Fig. 3 shows that a column boundary that has slipped has also produced an offset in a transgranular crack proving that the latter preceded the former during the loading cycle. In addition, it is found that bending cracks at the interface are frequently associated with columnar steps (Fig. 8). This feature is consistent with the fact that a bending crack relieves the compressive residual stress that exists in these films and which is known to increase the sliding stress through a Coulombic type effect. Alternatively, the reverse may also hold: a step created through sliding will act as a stress concentration that promotes the nucleation of a bending crack.

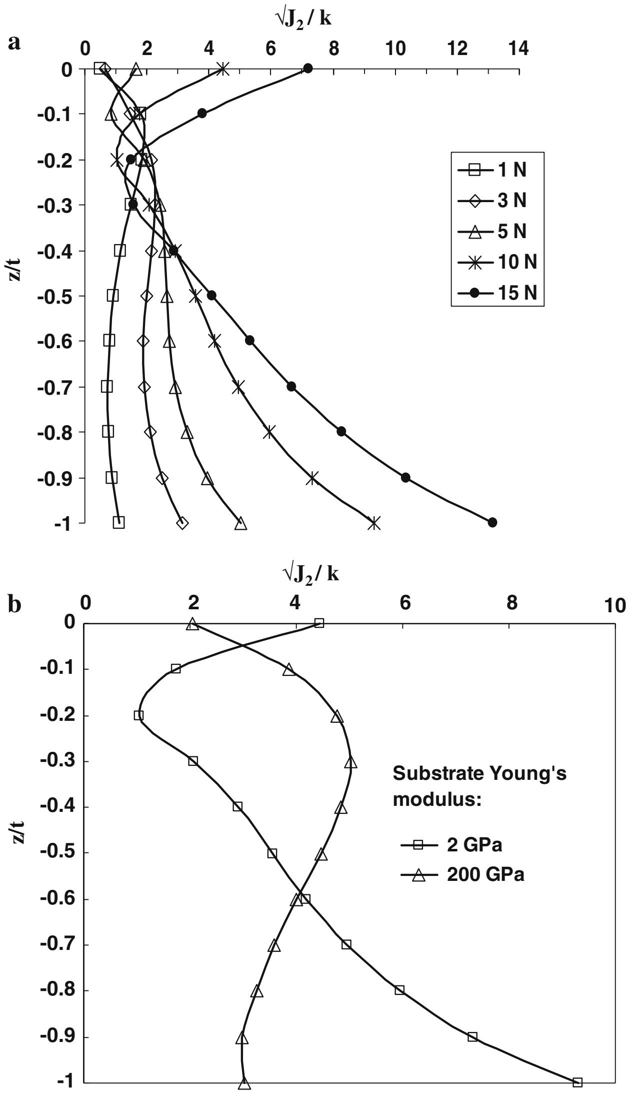


**Fig. 7** (**a**) $\sqrt{J_2}/k$ in a 10 μm thick film as a function of depth at a radial distance = 1 μm, $E_{sub}$ = 2 GPa, $E_{film}$ = 400 GPa. (**b**) $\sqrt{J_2}/k$ in a 10 μm thick film as a function of depth at a radial distance = 1 μm for $E_{sub}$ = 2 GPa and 200 GPa. $E_{film}$ = 400 G-Pa, load = 1 N

## Lateral cracks

We also know that when the substrate is compliant there are lateral cracks normal to the indentation axis, which exist in a zone directly below the indenter. Figure 5a shows such a crack about three quarters of the way through the film thickness. Such cracks are never seen when the substrate is stiff. We believe that such cracks occur due to residual tensile strain, which develop during unloading. Figure 7b shows that at a load of 10 N, $\sqrt{J_2}/k$ for a compliant substrate is significantly higher than that for a stiff substrate near the interface. The sharp gradient in the $\sqrt{J_2}/k$ with depth gives rise to

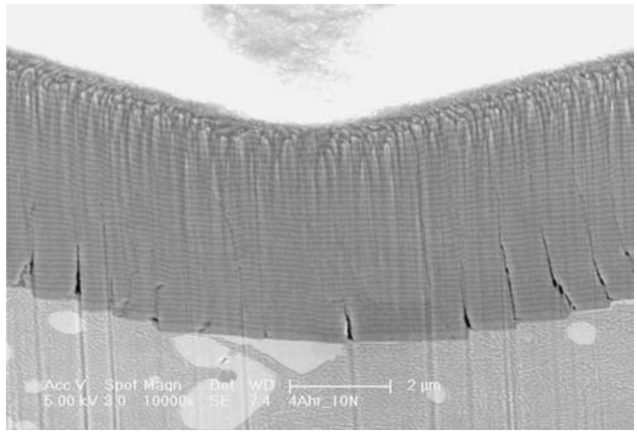


**Fig. 8** Bending cracks at the interface associated with columnar steps in a TiN–AlTiN thin film on HSS substrate, load = 10 N

a tensile stress on unloading, which is likely to lead to a lateral crack.

## Conclusions

The examination of the cross sections of indentations in columnar TiN–AlTiN multi-layers on steel has established the following features of cracking and deformation:

1. Columnar sliding begins in the middle of the film as substantiated by the maximum in the calculated shear stress acting along the column boundaries.
2. At the transition thickness at which columnar sliding gives way to transgranular fracture, it is possible for partial sliding to terminate in a crack if the average stress along the column does not exceed the critical sliding stress.
3. Edge cracks, the most prominent surface features in such indentations, do not propagate through the film as predicted from the rapid decay with depth in radial tensile stress near the indentation periphery.
4. Analytical elastic modeling, in which a low modulus simulates the effect of substrate plasticity, is a simple and useful tool to understand the behaviour of coatings with varying thickness and substrate yield strength.

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## References

1. Bhowmick S, Xie ZH, Hoffman M, Jayaram V, Biswas SK (2004) J Mater Res 19:2616

2. Bhowmick S, Jayaram V, Biswas SK (2005) Acta Mater 53:2459
3. Gunda R, Biswas SK, Bhowmick S, Jayaram V (2005) J Am Ceram Soc 88(7):1831
4. Ma KJ, Bloyce A, Bell T (1995) Surf Coat Technol 76–77:297
5. Ma KJ, Bloyce A, Andrievski RA, Kalinnikov GV (1997) Surf Coat Technol 94–95:322
6. Shiwa M, Weppelmann ER, Bendeli A, Swain MV, Munz D, Kishi T (1994) Surf Coat Technol 68–69:98
7. Bhowmick S, Kale AN, Jayaram V, Biswas SK (2003) Thin Solid Films 436:250
8. Bhowmick S, Bhide R, Hoffman M, Jayaram V, Biswas SK (2005) Philos Mag 85(25):2927
9. Soloukhin VA, Brokken-Zijp JCM, With GD (2003) J Mater Res 18(2):507
10. Math S, Jayaram V, Biswas SK (2006) J Mater Res 21(3):774
11. Math S, Jayaram V, Biswas SK (2006) J Mater Res 21(3):783