Discontinuous cracking of TiN film induced by adhesive interlayer under tension

Journal:	*Philosophical Magazine & Philosophical Magazine Letters*
Manuscript ID	TPHL-2018-0136.R2
Journal Selection:	Philosophical Magazine Letters
Date Submitted by the Author:	31-Jul-2019
Complete List of Authors:	Guo, Tao; University of Science and Technology Beijing, Department of Materials Physics and Chemistry Pang, Xiaolu; University of Science and Technology Beijing, Department of Materials Physics He, Jianying; Norwegian University of Science and Technology Qiao, Lijie; University of Science and Technology Beijing, Corrosion and Protection Center
Keywords:	coatings, cracking, interfacial segregation, plastic deformation, dislocation interactions
Keywords (user supplied):	buckling

Discontinuous cracking of TiN films on a steel substrate induced by an adhesive interlayer

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Abstract

The basic mechanisms governing the process of cracking of single-layer brittle films have been extensively explored through both simulations and experiments. However, the role that an adhesive interlayer plays in the cracking of the overlying brittle film remains unclear. By performing three-point bending experiments, we observed that the insertion of a 100 nm thick Ti interlayer changed the cracking behavior of TiN films from a continuous pattern to a discontinuous pattern. The slight change in the microstructure of the film and the increase in film thickness arising from the addition of the Ti interlayer are unlikely to cause the observed cracking morphology.

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The combination of the different interface between the Ti and the steel substrate and the fracture of the Ti interlayer are responsible for the transition in the TiN film cracking morphologies.

Keywords: TiN film; Ti interlayer; cracking morphology transition; interfaces; interlayer fracture.

1. Introduction

Brittle films prepared by magnetron sputtering are one of the most common surface modification methods for preventing damage to the underlying substrates without changing their surface components and structure [1]. However, the brittle films produced by this technique usually exhibit large residual compressive stress and poor adhesion, which results in film interfacial delamination [2-5]. A metallic interlayer, such as Cr, Ta or Ti, is usually applied to solve such problems [6-12]. Previous studies have reported that the insertion of such interlayers can significantly affect the cracking behavior of ductile Cu films [6, 8, 12, 13], even causing these films to undergo a ductile-to-brittle transition [8]. However, the effects of an adhesive interlayer on the cracking behavior of brittle films have received relatively little attention.

Generally, the cracking behavior of single-layer brittle films on ductile substrates under tension can be understood by a shear lag model, in which the normal stress in the substrate is transferred to the coating via a shear stress through the interface [7, 14-18]. The presence of a brittle interlayer will transfer the stress from the substrate to the film and change the microstructure and properties of the film, thereby affecting its cracking behavior [19-21]. The aim of the present study is to explore how the interlayer affects the cracking process of the overlying brittle film.

TiN films were prepared via magnetron sputtering on 50CrVA spring-steel substrates with or without a 100 nm Ti interlayer. The cracking behavior of sandwich specimens was studied using a homemade three-point bending device. The effect of the 100 nm thick Ti interlayer on the cracking behavior of the TiN film is here evaluated and relevant mechanisms proposed.

2. Experimental procedures

The dimensions of the 50CrVA spring-steel substrates were 15.5 mm × 0.9 mm × 6 mm. The TiN films were deposited on either the front 15.5 mm × 0.9 mm surface (the lateral surface) to study the cracking behavior or the 15.5 mm × 6 mm surface (the top surface) to evaluate the residual stress. Two sets of approximately 1 μm thick TiN films were deposited on substrates with and without a 100 nm thick Ti interlayer, as shown in Fig. 1. Both TiN films exhibit a fine columnar structure and the addition of a Ti interlayer increases the columnar width of the films. To consume the residual oxygen and nitrogen in the chamber, the Ti target was pre-sputtered for 10 min with the substrate sheltered prior to film deposition. The TiN films were deposited using the same parameters as those used in previous studies [22, 23]. The Ti interlayer was deposited using reactive RF-pulsed magnetron sputtering with a 2×10^{-3} Pa base pressure, a 300 W target power, a 0.3 Pa deposition pressure and a 0.25 Pa Ar pressure.

During the sputter deposition of the Ti interlayer, a bias of -80 V was applied to the substrate. In addition, Si substrates (20 mm × 20 mm) were also used to study the effect of the Ti interlayer on the cross-sectional microstructures of the TiN films.

The cross-sectional microstructures of the TiN films with and without Ti interlayers were first characterized by scanning electron microscopy (SEM). The residual stresses in the films were determined with the XRD $\sin^2\psi$ method [19] and determined to be approximately -2.1 ± 0.5 GPa and -3.1 ± 0.9 GPa with and without a Ti interlayer, respectively. The TiN films deposited on the lateral surface were bent to a maximum strain of 6% at a strain rate of 2 x 10^{-2} s^{-1}, similar to the approach used in our previous studies [22, 23].

3. Experimental results

The crack density of the TiN films after bending was measured along the longitudinal direction, as shown schematically in Fig. 2a. The maximum strain on the top surface of the sample was measured with a strain gauge. Assuming that the externally applied moment is constant along the same longitude, the strain can be calculated as follows:

$$\varepsilon_x = \left(1 - \frac{2y}{h}\right)\varepsilon_{\text{top}} \tag{1}$$

where h is the substrate thickness and y is the distance from the upper edge. Based on Eq. (1), the strain at the sites observed under SEM can be obtained by inserting the distance to the edge. Fig. 2b shows that the crack spacing of the TiN films decreases markedly at low strain and then gradually saturates when the films are bent to the maximum strain of 6%. The insertion of the Ti interlayer not only decreases the saturated crack spacing ($L_{\text{without}} \approx 7.7$ μm and $L_{\text{with}} \approx 4.4$ μm) in the TiN film but also lowers the critical fracture strain ($\varepsilon_{\text{without}}$=1.6% and $\varepsilon_{\text{with}}$=0.8%).

Fig. 3 shows SEM images of the lateral surface of the TiN films deformed at a maximum strain of 6%. The cracks in the TiN film without a Ti interlayer are straight and continuous, whereas those with a Ti interlayer are discontinuous. The same phenomenon is also observed in both crack-initiation and crack-saturation stages. As previously mentioned, the normal stress in the substrate is transferred to the coating by the interface, meaning that the normal stress is transferred through the interlayer in this bilayer system. Therefore, the cracking morphology of a bare 100 nm Ti film (keeping the same deposition time as that used for the Ti interlayer) on the 50CrVA spring steel substrate was studied. Fig. 4a shows that the Ti layer forms many discontinuous and irregular cracks with a much smaller saturated crack spacing of $L \approx 0.33$ μm. Of particular interest, the critical strain for crack initiation in the Ti layer is approximately 2.7%, which is much larger than that for the TiN films, as shown in Fig. 4b.

4. Discussion

The experimental results presented above reveal that the presence of the Ti interlayer produced three effects on the cracking behavior of the overlying TiN films (i.e. the Ti interlayer): decreasing the critical fracture strain, shortening the saturated crack spacing, and altering the cracking morphology. The first two effects can be attributed to the relief of compressive stress in the TiN films with a Ti interlayer, which will accelerate the crack initiation and subsequently decrease the saturated crack spacing of the films [23] according to the shear lag model [14]. In the following section, we will focus on the discussion of how the Ti interlayer affects the cracking morphology of the TiN films.

The microstructure [25, 26], thickness [4, 27], and interfacial structure [28] of the films were reported to be able to alter the cracking morphologies of single brittle layers. For example, cracks in a crystalline $Ge_2Sb_2Te_5$ (GST) film deviated from straight paths as they propagated along the grain boundaries, whereas this phenomenon is not observed for cracks propagating in amorphous GST films [26]. A Cr film with columnar grains exhibits a network-type cracking with cracks regularly deviating from the direction perpendicular to the applied strain, whereas the cracks in a Cr film with equiaxed grains are straight [25]. In polyimide-supported Cr films, the cracking morphology transforms from network-type to straight cracking as the film thickness increases from 50 nm to 150 nm [4]. However, polyimide-supported Ti films exhibit straight cracks in thinner 8 nm films, whereas these films exhibit zig-zag cracks for thicker 50 nm films [27]. In addition, the change in interfacial structures also affects the film cracking morphologies. For instance, the annealing-induced crystallization of the amorphous interlayer at the interface changes the film morphology from zig-zag cracking to straight cracking [27]. Herein, the effects of the microstructure, thickness and interfacial structure of the films on the resultant cracking morphologies are individually considered.

Fig. 1 shows that the presence of a Ti interlayer simply increases the columnar width, and there are no essential differences in the microstructures of the two sets of TiN films. In addition, the cracking morphology of the 1.1 μm thick TiN without Ti interlayer is straight and continuous, as shown in Fig. 5. Therefore, the changes in the microstructures of the TiN films and the increase in the film system thickness arising from the addition of a Ti interlayer are unlikely to cause the cracking morphology to transform in the present study. The influence of the Ti interlayer on the cracking of the TiN film can be divided into two steps with respect to the applied strain. When the applied strain is less than 2.7% (the critical strain of the Ti layer), the normal stress in the substrate transferred to the film and the resultant cracking are mainly controlled by the Ti/steel interface. Fig. 4a shows that the normal stress transferred from the substrate through this interface, causing discontinuous and irregular cracking in the Ti film. Therefore, we attribute the discontinuous cracking of the TiN film at the crack initiation stage to different stress transfers through the Ti/steel interface, as shown in Fig. 3d.

Once the Ti interlayer fractures (the applied strain exceeds 2.7%), the cracking process of the overlying TiN film will be subjected to two effects. The first is tensile stress relief, which reduces the stress transfer to the overlying TiN film; hence, a higher strain is required for further cracking [15]. In addition, the formation of cracks also causes stress redistribution in the uncracked Ti segments [12, 15], thereby affecting the stress transfer to the TiN film. The other effect is a stress concentration generated in the TiN film above the Ti interlayer; this stress concentration is located near the cracks in the Ti layer [12]. As the applied strain increases, an increasing amount of stress transfers to the TiN film through the uncracked Ti segment, and the stress concentration becomes more substantial due to the increased crack opening of the Ti interlayer, which will eventually cause the TiN film to fracture. The above discussions indicate that the cracking process of the TiN film is highly dependent on the fracture of the Ti interlayer. The discontinuous cracking of the TiN film will be promoted once the underlying Ti interlayer fractures. In addition, as indicated in Figs. 2b and 4b, the saturated cracking spacing of the Ti film is much smaller than that of the TiN film; therefore, the cracking of the Ti interlayer also contributes to decreasing the saturated crack spacing of the TiN film.

5. Conclusions

In the present study, the influence of a 100 nm thick Ti interlayer on the cracking behavior of a 1 μm TiN film adhered to a Si substrate was experimentally investigated. The results showed that the Ti interlayer contributed to an increase in the columnar width and a relief of the compressive stress. In three-point bending tests, the cracking morphology of the TiN films changed from a continuous pattern to a discontinuous pattern due to the presence of the Ti interlayer. The slight change in the microstructure of the TiN film and the increase in film thickness due to the addition of Ti interlayer were unlikely to cause the cracking morphologies to transform. The different interface between the Ti and the steel substrate and the cracking of the Ti interlayer are attributed to altering the stress transfer across the interface to the TiN film, leading to the resultant transition in cracking morphology. The relief of compressive stress and the cracking of the Ti interlayer created a stress concentration that decreased the saturated cracking spacing of the TiN film.

Acknowledgements

This work was supported by Beijing Nova Program (Z171100001117075), the National Natural Science Foundation of China (51771025, 51431004).

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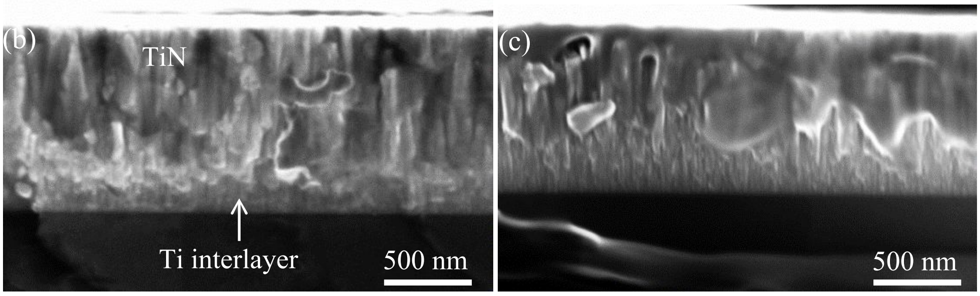

Fig. 1. The SEM cross-sectional microstructures of TiN films: (a) with Ti interlayer; (b) without Ti interlayer

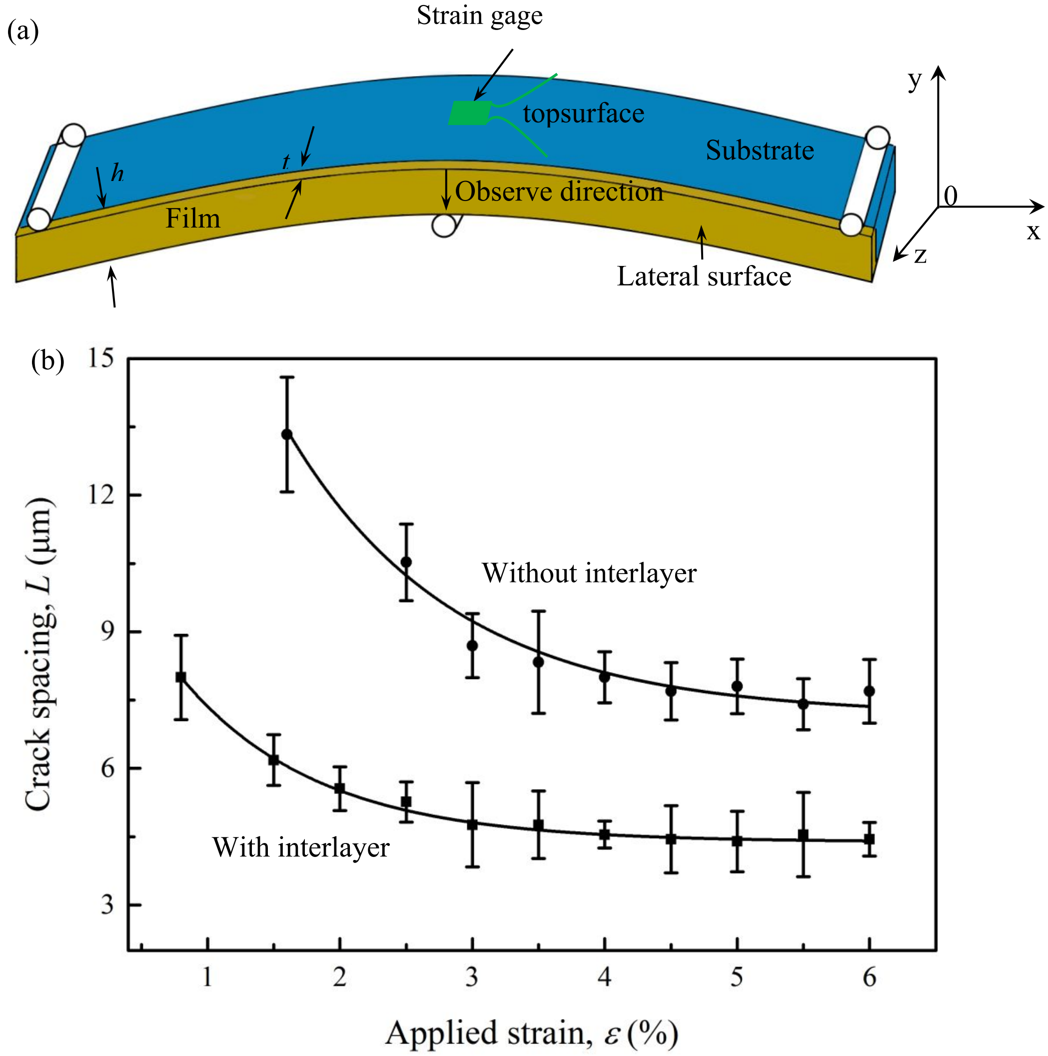

Fig. 2 (a) Schematics of the sample in three-point bending. The film is on the lateral surface, labelled by yellow color. (b) Crack spacing as a function of the applied tensile strain for TiN film with and without Ti interlayer under three-point bending. Three specimens were used for each point.

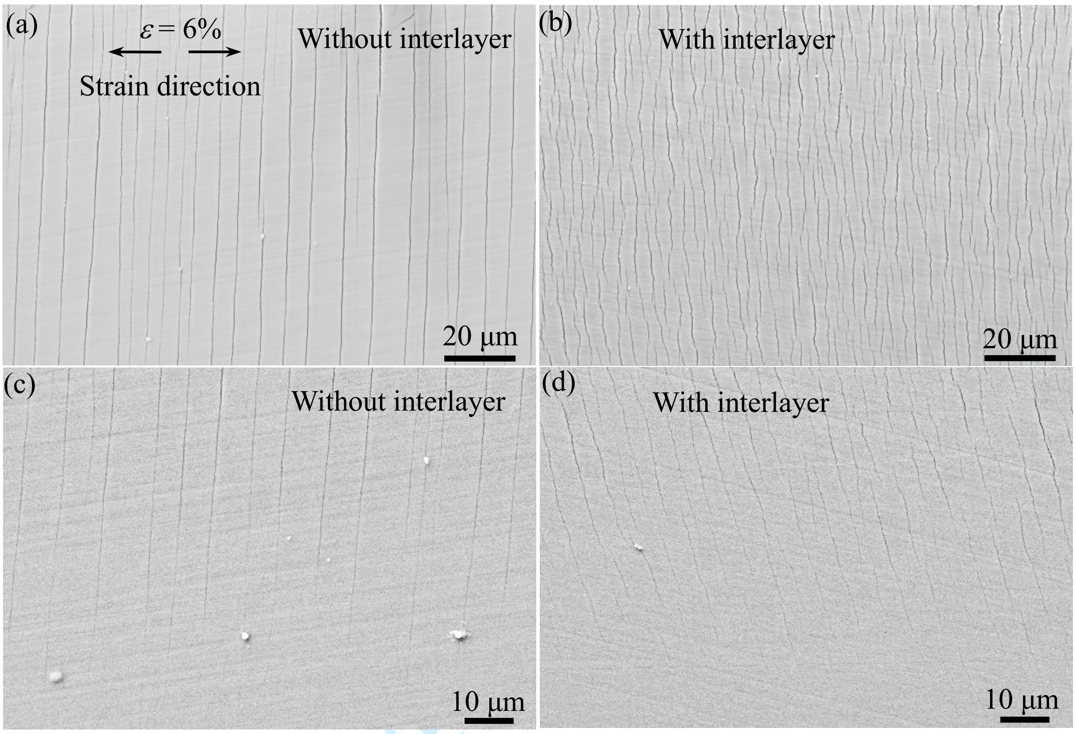

Fig. 3. SEM micrographs of TiN films (about 1 μm) strained to approximately 6% under three-point bending: (a) and (c) without Ti interlayer showing straight and continuous cracks, (b) and (d) with Ti interlayer showing discontinuous cracks. (a) and (b) are micrographs at the strain of 6%, (b) and (d) are micrographs when the cracks start to appear.

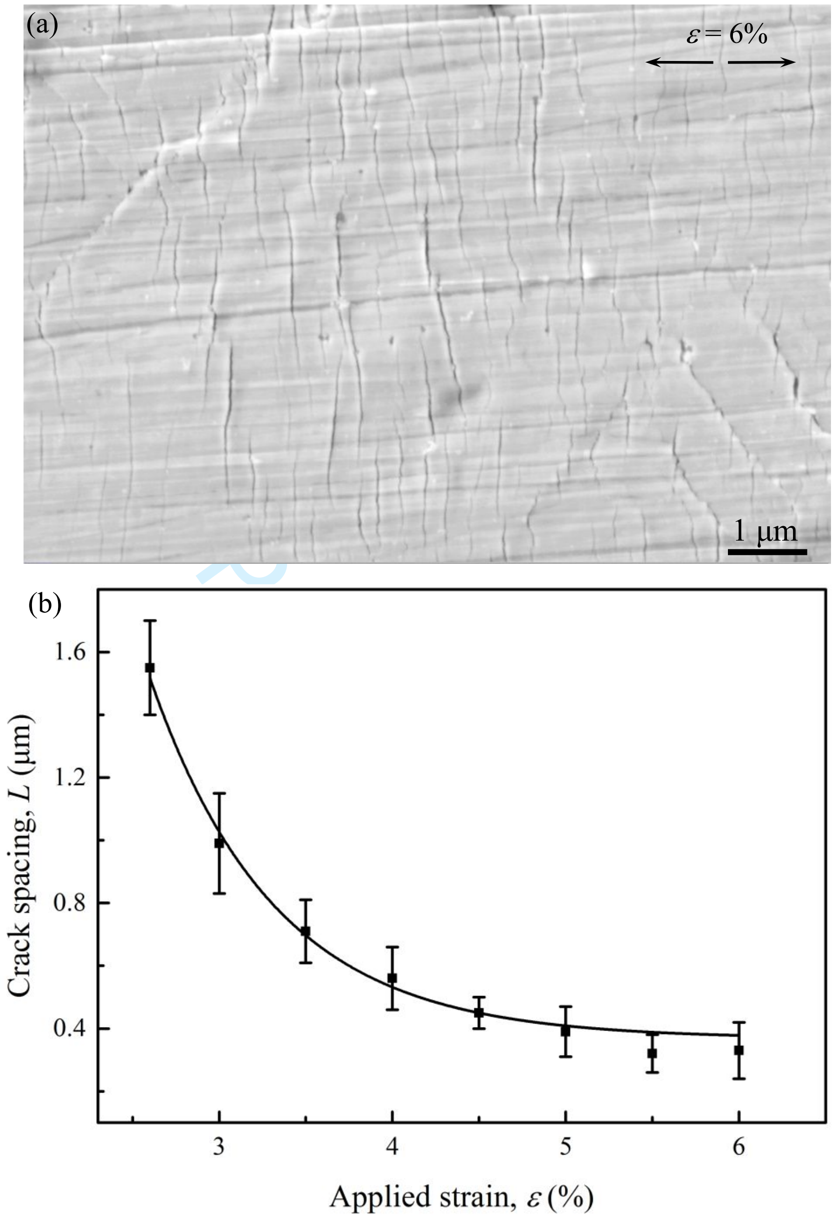

Fig. 4. (a) SEM micrograph of bare 100 nm Ti film surface strained to approximately 6% under three-point bending. (b) Crack spacing as a function of the applied tensile strain.

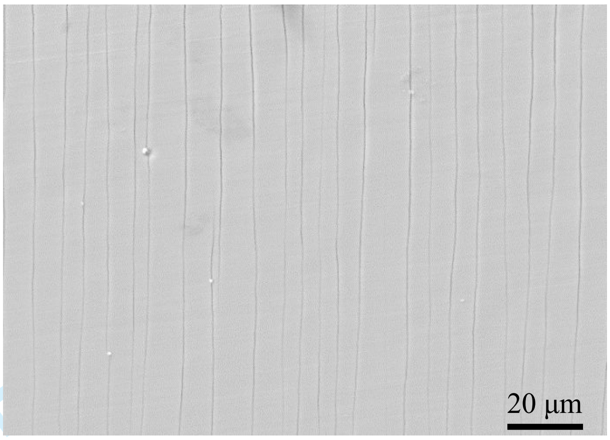

Fig. 5 SEM micrograph of TiN film (about 1.1 μm) strained to approximately 6% under three-point bending, showing straight and continuous cracks.