A new method to evaluate the fracture toughness of thin films

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Abstract

A method based on microindentation is developed to determine the fracture toughness $K_I$ of a thin film bonded to a brittle substrate. Using an easily fabricated sample having a partially coated substrate, indentation on the uncoated portion of the substrate is used to generate a radial crack that propagates into and away from the coated region. Comparison of the lengths of the surface traces of the indentation crack into and away from the coated region is used to measure the toughening imparted by the coating. This test method avoids the complicating effects of delamination that often occur when the coating is indented directly. To extract quantitative results, a 3D finite element model of the system geometry is generated and a cohesive zone model is used to predict the complex equilibrium crack front. The model predicts the length of the crack penetration into the coating vs. the length of the crack growth away from the coating as a function of the elastic and toughness properties of the coating and the substrate, and the residual thermal stresses, which play an important contribution to the detailed crack growth. An approximate analytic model using energy balance ideas is developed to permit easy determination of the coating toughness from experimental data, and the model agrees well with full numerical results over a wide range of coating and substrate property values. The overall method is applied here to determine the toughness of a thin CVD diamond coating on a thick silicon single crystal substrate, and a coating toughness of $K_I = 8.4$ MPa m$^{1/2}$ is obtained using a previously measured biaxial tensile coating residual stress of 1 GPa.

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1. Introduction

Fracture toughness is an important property for brittle materials, and many methods for measuring the mode-I fracture toughness $K_I$ have been developed. The most commonly used specimens are single-edge notched beams tested in three- or four-point bending, double cantilever beams loaded in tension, double torsion specimens, short-rod specimens with a chevron notch, and compact-tension specimens. These techniques require comparatively large specimens having specific geometries and thus are not amenable for measuring the properties of thin films deposited on substrates. As a result, more approximate methods based on indentation, which is well suited to in situ testing of films and small volumes, have been devised to measure $K_I$.

Micro- and nano-indentation have been used to measure the fracture toughness of many brittle materials [1–14]. When brittle materials are indented with a sharp tip, radial cracks are generated during the unloading cycle. The length of these radial cracks is correlated with the fracture toughness by various semi-empirical relationships based on solutions of half-space fracture mechanics problems [15]. The fracture toughness so obtained often correlates well with the fracture toughness measured by more rigorous means but the absolute values are usually in some error. More importantly, applying the indentation technique to thin films can give misleading results due to effects that are not easily incorporated into the analysis, such as: (i) the effect of the substrate on the deformation fields, (ii) crack growth into the substrate, (iii) interface delamination, and (iv) the influence of residual stresses. When delamination...
occurs, a large fraction of the mechanical driving force can be consumed in creating the new delaminated surfaces and it is difficult to separate out the energy corresponding only to the cracking of the coating. For these reasons, the conventional fracture toughness measurement based on radial cracking cannot be used directly. Efforts have been made to deal with the effects of substrates and interface delamination [15,16]. For example, Li et al. [16] evaluated the $K_{\text{IC}}$ of thin films using non-indentation by directly measuring the energy released during the second-ring cracking after interfacial cracking. However, they assume that the effects of interfacial debonding are negligible. These methods may be successful on specific systems after much testing and complementary analysis, but are not sufficiently well-controlled to represent a reliable method.

In the present work, we propose a new method to determine $K_{\text{IC}}$ in thin films without the occurrence of the problems noted above. Instead of directly indenting the film, we propose preparation of a half-coated sample, easily created by masking within an existing deposition chamber, and the use of indentation to generate radial cracks that initiate in the uncoated substrate just adjacent to the coated substrate. The propagation of one radial crack into the film is then used to investigate the film fracture resistance without other complexities. The fracture toughness of the coating is obtained by comparing the lengths of the radial crack traces that propagate into and away from the coating, and interpreting this data using a detailed finite element analysis, and simple but accurate analytic model. The new method and analysis developed here are used to determine the toughness of polycrystalline CVD diamond films on single-crystal silicon substrates. The remainder of this paper is organized as follows. In Section 2, we present the experimental method and test sample. Section 3 describes the numerical modeling. The crack growth predictions, development and verification of analytic models, and comparison to the experiments are presented in Section 4. Section 5 summarizes our results.

2. Experiments

Polycrystalline diamond coatings were deposited on polished [001] single-crystal 2.5 mm-thick silicon substrates within an AsTex (model HPM/M) microwave plasma chemical vapor deposition (CVD) reactor. To prepare the Si surface for deposition, the substrate was first cleaned in an ultrasonic container and then abraded with 0.5 μm diamond grit to increase nucleation. All films were grown with a substrate temperature of 800 °C, an input microwave power of 1200 W, and a gas composition of 0.5% CH$_4$ in H$_2$. Deposition times of 10 h produce 3 μm thick coatings. Additional experimental details concerning film growth are published elsewhere [17]. Of importance here is that before deposition the substrates were partially masked by a thin silicon plate of 0.3 mm in thickness to form a clean boundary, aligned along the (110) directions, between coated and uncoated areas of the sample. After deposition, the mask was removed, thus generating a sample consisting of a Si wafer that is only partially coated with a diamond film. To permit wafer curvature measurements of the intrinsic stress, films were also grown under identical conditions without employing the masking procedures [17]. Microindentation was performed with an Instron Indentation Instrument (Wilson model Tukon 2100) with a maximum force of 1 kg applied. The load was applied over 10 s, held for 10 s at the peak load, and then unloaded completely. Using a standard Vickers indenter aligned along the $\langle 110 \rangle$ directions in the Si substrate and situated at a preselected distance $s$ from the edge of the coating, radial cracks were initiated by indentation into the substrate. Crack propagation into the coating at attainable loads and without the indentation mark reaching the coating requires that the distance between the indent and the edge of the coating, $s$, be chosen appropriately. For the present CVD diamond coating and Si substrate, cracks reaching and growing into the coating were observed for $s < 25$ μm at $P = 4.5$ N and $s < 40$ μm at $P = 9.5$ N. For this indenter alignment, half-penny cracks propagate along $\{ 110 \}$ cleavage planes in the Si. Fig. 1 illustrates the geometry of the experimental set-up and resulting indentation cracks, along with some notation regarding the various lengths associated with the crack formation. The indentations and associated crack patterns were observed using both optical microscopy and scanning electron microscopy (SEM).

Fig. 2(a) and (b) shows an indentation crack formed in this test, demonstrating crack propagation into the coated region of the sample. Note that the crack growing away from the coated region (length $d$) is notably longer than the crack growing into the coating (length $a$). This demonstrates that the tough coating is sufficient to restrain the
crack growth significantly, relative to the substrate material. _This is the first main result of this paper._

For comparison, direct indentation of the diamond film shows clear evidence of delamination between the film and the substrate in regions adjacent to the indent, as shown in Fig. 2(c). The difficulty of interpreting the indentation data in the presence of such delamination is the primary reason for developing the new technique with masked samples.

Although Fig. 2(a) shows that the film is tough relative to the coating, detailed analysis must be performed to relate the ratio \( a/b \) to the actual coating fracture toughness because the surface observations provide no information on the full subsurface geometry of the radial crack. The analysis is described in the following section.

### 3. Finite element analysis

To predict the crack propagation and stable crack geometries in the coating/substrate system, and thus determine the fracture toughness of the coatings associated with the test geometry of Fig. 1 and crack pattern of Fig. 2(a), several 3D finite-element/cohesive-zone models were developed. Making use of symmetry, a 45°-wedge model was developed as illustrated in Fig. 3(a). The geometry and symmetry of this model corresponds to a situation in which there is a square patch of uncoated substrate of edge length \( 2s \) surrounded by coated substrate, with two perpendicular radial cracks. This geometry does not correspond precisely to the experimental situation but calculations of the crack length vs. applied loading without, and with, the coating provide

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![Fig. 2. SEM photographs of microindentation with \( P = 4.5 \) N: (a) indentation on the substrate close to the coating; (b) magnified view of (a); (c) indentation directly on diamond film/silicon substrate, showing delamination.](image)

![Fig. 3. Schematics of (a) 45°-wedge and (b) half-infinite plate finite element models with cohesive zones, and (c) the traction–displacement relationship used for the cohesive zone law.](image)
the necessary crack lengths \( b \) and \( a \), respectively. For comparison, a half-infinite plate model was also constructed, as shown in Fig. 3(b). For this model, there is only one radial crack but the sample configuration is the same as in the experiments. Comparison of results from the two models demonstrates the insensitivity of the coating cracking to the precise geometry away from the coating (see below).

In the models, the interface between the coating and the substrate is assumed to be perfectly bonded. The \( x-z \) surface at \( y = 0 \) is the plane of crack growth in this geometry, and a cohesive zone model is used on this plane to permit crack growth. Specifically, an initial small-radius, free surface region is surrounded by a region in which special 2D rod elements perpendicular to the crack plane are employed as cohesive elements to permit crack growth. The rod element constitutive behavior is described by the relationship between the normal traction \( T \) and corresponding displacement jump \( \delta \) across the interface given by [18]

\[
T = \frac{\sigma_{\text{max}}}{\delta_n} \delta_1^{1-(\delta/\delta_n)},
\]

where \( \sigma_{\text{max}} \) is the maximum interfacial normal tensile strength and \( \delta_n \) is the characteristic critical opening length, as shown in Fig. 3(c). Notice that here the tangential traction across this surface is ignored in the cohesive law. For such a cohesive zone law, the normal work of fracture \( G_c \) is the area under the traction–separation law,

\[
G_c = c \sigma_{\text{max}} \delta_n,
\]

which is related to the fracture toughness \( K_{\text{IC}} \) of the separating material (coating and/or substrate) as

\[
G_c = \frac{K_{\text{IC}}^2 (1 - v^2)}{E},
\]

where \( E \) and \( v \) are the Young modulus and Poisson ratio, respectively. Below, we will use \( K_c \) to denote the fracture toughness \( K_{\text{IC}} \) for the coating and \( K_s \) to denote that of the substrate.

In using a cohesive zone model, the relevant length parameter is not \( \delta_n \) but rather the characteristic cohesive zone length

\[
\delta_c = \frac{E G_c}{\sigma_{\text{max}}^2}.
\]

To properly model crack growth in a finite-element/cohesive-zone model, the numerical mesh must be several times smaller than the characteristic cohesive zone length \( \delta_c \). In this work, we take \( \delta_c = 7.4 m \), where \( m \) is the mesh size in the direction of crack propagation, for both the coating and the substrate materials. Choosing \( \delta_c \) to be the same for both materials allows the toughnesses and stiffnesses to be varied as desired without adaptive meshing or concern for inadvertently loss of computational accuracy. For fracture problems, where one is concerned with the propagation of an existing crack front, only the critical strain energy is required and hence the maximum separation stress \( \sigma_{\text{max}} \) and the characteristic length \( \delta_c \) can be adjusted as needed, within a wide range, to preserve the desired fracture energy and satisfy the need for computational accuracy and consistency. Given a mesh size \( m \), work of fracture \( G_c \), and elastic properties \( E \) and \( v \), the cohesive zone parameters can be calculated using Eqs. (2)–(4). Below, we will apply the model to a diamond film on Si using a mesh size of \( m = 1 \mu m \), and will use the parameter values shown in Table 1 [19].

The discretized FE models contain about 25,000 nodes and 23,000 elements with very fine meshes in the region over which the crack perimeter exists. The indentation is simulated using MSC–Nastran (Version 7.7), making use of as far as possible eight-node, six-side solid elements. In the simulation, the crack is driven by applying uniform forces perpendicular to the crack surface on the nodes within some small radius around the origin of the indent crack. Note that the indentation process is not simulated completely; we insert an initial starting crack without any plastic indentation and then apply forces that simulate the indentation load and drive the crack opening. In both models, the maximum cracked area is less than 0.01% of the cross-section of the models.

Residual stresses in the coating/substrate system can strongly influence the crack growth, i.e. the lengths \( a \) and \( b \), and thus must be considered in deriving a value of \( K_c \) for the coating from experiments. The diamond films used here have a residual biaxial tensile stress of 1 GPa, while the substrate is in biaxial compression to a degree determined by the coating/substrate modulus and thickness ratios [17]. The intrinsic tensile stress in these films is

<table>
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<th>Table 1</th>
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<td>Some parameters used in the finite element models</td>
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<tr>
<td>Coating</td>
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<tr>
<td>Substrate</td>
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generally attributed to grain boundary formation [17]. This tensile stress in the coating naturally drives crack growth. However, compression in the substrate restraints the substrate crack growth, particularly into the depth of the substrate. In the present case where the substrate is 2.5 mm in thickness, finite element results and analytic estimates show that the compressive residual stress in the substrate is below 5 MPa, which is negligible. The existence of a free coating edge in our special sample geometry also modifies the residual stress fields relative to the uniform values that would prevail in the interior of a large system. Thus, the shape of the resulting crack is strongly distorted by these competing factors. To study the effect of the residual stress on the crack lengths and within the numerical model, we generated tensile biaxial residual stresses of up to 1.2 GPa in the coating by introducing a thermal mismatch and tuning the temperature to obtain the desired stress level.

Several subtle details of the FE calculations are worthy of discussion. First, at the coating/substrate interface, two nodes are employed, one for each material on either side of the interface. This permits the elements on either side of the interface to deform according to the correct constitutive rules of the material in the element while maintaining continuity of displacements across the interface. Second, in the 45°-wedge model, along the z-axis (x = y = 0), the intersection of the symmetry plane at 45° and the cohesive zone along the x-z crack plane leads to indeterminant boundary conditions. Since the crack must open along the crack plane and since in the full problem this line can move due to the intersection of perpendicular radial cracks, we artificially sever the cohesive zone rods along these boundary nodes. Finally, the desired cohesive zone law cannot be directly applied on the Nastran elements because the traction becomes zero at large displacement and this causes problems in the Nastran code. To fix this purely software-related problem, we simply strain each rod element and then apply an equivalent force on the end of the rod so that the total force on the element is zero but the rod element remains stressed. We then modify the stress–strain curve so that the crack opening still follows the desired cohesive law. Such sleight-of-hand might not be needed for other FEM software packages.

Below we will show results for various equilibrium crack perimeters. The crack tip is taken as the point where the normal displacement δ is equal to 6δn, at which point the normal stress on the separated surface is essentially zero.

4. Results and analysis

4.1. Experimental and numerical results

Fig. 2(a) shows the indentation on the silicon substrate near the diamond coating. Two radial cracks are generated, one of which propagates into and away from the coating while the other grows in the substrate parallel to the coating. Careful imaging shows no evidence of debonding between the coating and substrate, even at the higher loads where the crack propagates into the coating. The crack length in the coating, a, is always shorter than the cracks in the substrate, b. The indentation crack sizes at different loads and distances s are presented in Table 2.

FE simulations were performed for cracks in pure silicon with no coating and for the coating/substrate system with a variety of coating properties. In all cases, the initial crack grows roughly radially with increasing applied load, but the crack front does not maintain a half-penny shape, even when a uniform force or strain is applied in the indent area. This is attributed to a free surface effect wherein the stress intensity factor in a subsurface layer drops off rapidly [20]. For the coating/substrate system, the crack propagation is strongly affected by the coating. Fig. 4 shows the normal stress distribution in the cohesive zone for a pure silicon substrate (using $K_c = 0.83$ MPa m$^{1/2}$) and for a coating/substrate system with a coating fracture toughness of $K_c = 10$ MPa m$^{1/2}$, as calculated for both the wedge and half-space models. Fig. 5 shows several crack front shapes for different values of coating fracture toughness and modulus at the same applied stress and it is evident that the tough coating impedes crack growth to some degree in all cases. Fig. 5 also shows that the crack growth in the uncoated portion of the substrate is only slightly affected by the presence of the coating. The crack front under the indenter does differ between the two models, but the predicted surface crack lengths a and b are nearly identical, especially for low fracture toughness of the coating.

<table>
<thead>
<tr>
<th>Load P (N)</th>
<th>Crack in substrate b</th>
<th>Crack in coating A</th>
<th>Distance to coating s</th>
<th>Impression diagonal d</th>
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<tbody>
<tr>
<td>4.5</td>
<td>51.2</td>
<td>34.2</td>
<td>20.7</td>
<td>26.8</td>
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<tr>
<td>4.5</td>
<td>50</td>
<td>36.4</td>
<td>19.6</td>
<td>26.3</td>
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<td>6</td>
<td>57.3</td>
<td>40.7</td>
<td>23.6</td>
<td>30.5</td>
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<tr>
<td>6</td>
<td>55.9</td>
<td>40.1</td>
<td>22.7</td>
<td>30.9</td>
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<tr>
<td>6</td>
<td>61.0</td>
<td>52.4</td>
<td>22.0</td>
<td>31.7</td>
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<tr>
<td>6</td>
<td>59.8</td>
<td>53.7</td>
<td>22.0</td>
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<tr>
<td>6</td>
<td>64.6</td>
<td>50.0</td>
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<tr>
<td>6</td>
<td>63.4</td>
<td>42.7</td>
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<tr>
<td>8</td>
<td>73.2</td>
<td>56.1</td>
<td>30.5</td>
<td>39.0</td>
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<tr>
<td>8</td>
<td>74.4</td>
<td>58.5</td>
<td>25.6</td>
<td>39.0</td>
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<tr>
<td>8</td>
<td>80.5</td>
<td>67.1</td>
<td>23.2</td>
<td>39.0</td>
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<tr>
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<td>78.0</td>
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<td>9.5</td>
<td>87.8</td>
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Fig. 6 shows the length of crack growth in the coating, as a function of the length of crack growth, in the uncoated substrate. For a given coating toughness, $a$ is nearly linear with $b$ when the crack is large enough to be away from the coating edge ($a > s$). The numerical data provide a direct means by which the fracture toughness of the coating can be evaluated: the measured values of $a$ and $b$ can simply be placed on Fig. 6 and the coating toughness can be estimated directly.

Fig. 7 shows the crack length relationship $a$ vs. $b$ in the presence of various tensile residual stress levels in the coating for a fixed coating toughness. The $a$ vs. $b$ relationship remains linear and is nearly parallel to the result found for the case of zero residual stress. At a fixed $b$, the residual tensile stress simply increases the coating crack length $a$, as expected. Thus, increasing tensile residual stress in the coating has an effect similar to a reduced coating fracture toughness. This is not surprising since cracking of the coating releases the stored energy associated with the residual stress field.

Figs. 6 and 7 demonstrate that direct measurement of the lengths $a$ and $b$ over a range of indentation loads (range of lengths $b$) provides a means of assessing coating toughness, given information only on the substrate constitutive properties, the coating elastic properties, and the residual stresses. These other properties can be determined with good accuracy by a host of existing experimental methods. In particular, coating residual stresses can be determined independently from measurements of wafer curvature or X-ray diffraction. Thus, numerical generation of Fig. 6 and/or Fig. 7 for a given system permits direct determination of coating toughness.
model represents an extension of the standard indentation-load/crack-length relationship that is typically used to derive toughness from indentation on uncoated surfaces.

A crack front at equilibrium, independent of how it is formed, has a stress intensity factor at all points along the crack front that is equal to the fracture toughness. This makes prediction of the crack front shape difficult for most geometries and inhomogeneous elastic problems. However, if we compare cracks with coating properties different from and identical to those of the substrate, for which the relative crack growth is small, as generated at the same applied loading, an energy balance should be fairly accurate. The indent crack is driven by work done by the indenter system on the sample. In the half-coated sample, the work done by the indenter is the same as that done on the uncoated sample. Although some work goes into creating the indentation impression and generating other near-indent damage, such work is common to the uncoated and half-coated samples. Hence, the differential work in creating the indent cracks in the two cases (coating different from, and coating identical to, the substrate) should be zero. Thus,

\[ A_c G_c + A_s G_s = A_{sc} G_s + A_{ss} G_s, \]  

where \( G_c \) and \( G_s \) are the strain energy release rates for the coating and substrate, \( A_c \) and \( A_s \) are the true cracked coating and substrate areas, and \( A_{sc} \) and \( A_{ss} \) are the cracked areas when the coating properties are identical to those of the substrate. Using Eq. (3) and solving Eq. (5) for the coating fracture toughness \( K_c \), we have

\[ K_c = K_s \left( \frac{E_c (1 - v_c^2)}{E_s (1 - v_s^2)} \right)^{1/2} \frac{A_{ss} - A_s + A_{sc}}{A_c}, \]  

where \( E_c \) and \( E_s \) are the Young moduli of the coating and substrate, respectively. We have verified that Eq. (6) agrees well with the finite element results, using the simulated crack areas such as those shown in Figs. 4 and 5, for various combinations of substrate and coating properties and at different load levels.

Proceeding further, we know the areas \( A_c = t(a - s) \) and \( A_{sc} = t(b - s) \), where \( t \) is the thickness of the coating, but the areas \( A_s \) and \( A_{ss} \) involving subsurface cracking are unknown functions of \( a, b, t, \) and \( s. \) Analyzing the finite element results, we have found that the quantity \( [(A_{ss} - A_s + A_{sc})/A_c]^{1/2} \) is proportional to the quantity \( (\phi b - a)/t + B, \) where \( \phi \) is a dimensionless factor and \( B \) is a constant. Introducing a proportionality constant \( \lambda, \) Eq. (6) can be rewritten, after some manipulation, as

\[ K_c = K_s \left[ 1 + \lambda \frac{(\phi b - a)}{t} \right] \frac{E_c (1 - v_c^2)}{E_s (1 - v_s^2)}, \]  

4.2. Analytic model

We now develop an approximate analysis that supplements the numerical results. This analysis provides both insight and analytic formulae for use in general coating/substrate systems, thereby avoiding the development of purely numerical models. Such an analytic toughness from experimental data. This is the second main result of this paper.
where $\lambda$ and $\phi$ are geometry terms obtained from the FEM. If Eq. (7) is solved for $a$ in terms of $b$, Eq. (7) predicts $a$ to be linear in $b$, consistent with the FEM results, and the parameter $\phi$ is the slope of $a$ vs. $b$. Therefore, $\phi$ can be determined directly from the experimental values of $a$ and $b$ without using the FEM results. The parameter $\lambda$ has no simple means of determination but the finite element results show that the single value $\lambda = 0.45$ is accurate for a wide range of coating and substrate properties for coating thicknesses of 3 and 6 $\mu$m and at different loading levels. To demonstrate that Eq. (7) is accurate with this one value of $\lambda$, Fig. 8 shows the calculated coating fracture toughness using Eq. (7) against the actual $K_c$ values input into the numerical calculations, where the slope $\phi$ is taken from the numerical “data” of Fig. 6 and many other similar FEM calculations.

To include the effects of residual stress, we make a similar energy balance argument as follows. The work done by the indenter, as described earlier, is now augmented by work done on or by the residual stresses in the system. When a compressive residual stress exists in the coated system, some of the work of the indenter goes into relieving the residual coating compression. When a tensile residual stress exists in the coated system, the tensile residual stress provides work that acts along with the indenter to drive the crack. Since the energy stored in the coating system is predominantly in the coating itself, for a thick substrate, the differential work in creating the indent cracks in the two cases (coating different from, and coating identical to, the substrate) is equal to the work done by the coating residual stresses.

$$ (A_c G_c + A_g G_g) - (A_{c0} G_c + A_{g0} G_g) = \pm G_{rc} A_c, $$

where $G_{rc}$ is the stored energy due to residual stresses in the coating and the $+$ and $-$ signs refer to tensile and compressive residual stresses, respectively. To obtain $G_{rc}$, we consider work of Lawn and Fuller [21] which showed that for a residual stress $\sigma_r$ distributed uniformly over a depth $t \ll b$, the stress intensity at the surface of a penny crack is $K_r = 2\psi_\lambda \sigma_r \sqrt{r}$ where $\psi_\lambda$ is a geometry factor on the order of unity ($\psi_\lambda = 2/\pi^{1/2}$ for a penny crack). In the present case, the residual stress exists in a thin coating layer with different elastic properties than the substrate and the cracks are not exactly half-penny cracks. As a result, the factor $\psi_\lambda$ differs, but our finite element results show that $\psi_\lambda = \psi_\lambda^{(2)}(1-\nu_c^2)/E_c(1-\nu_c^2)$ with good accuracy for different residual stress levels with $\psi_\lambda = 0.95$ (close to $2/\pi^{1/2}$). The Lawn and Fuller solution for stress intensity is converted into an estimate for the desired required work suggests $G_{rc} = K_r^2(1-\nu_c^2)/E_c$. Substituting this result into Eq. (9) and solving for the coating energy release rate yields $G_{rc} = G_c(A_{rc} + A_u - A_d) / A_t \pm G_{rc}$.

Using Eqs. (6) and (7), and Lawn’s result for $K_r$, and solving for the coating fracture toughness, we obtain our final result

$$ K_c = \left\{ K_s^2 \left[ 1 + \lambda \frac{(\phi b - a)}{t} \right] \left[ \frac{E_c(1-\nu_c^2)}{E_s(1-\nu_s^2)} \right]^{1/2} \right\}^{1/2}, $$

where the dimensionless factors $\lambda$ and $\psi_\lambda$ are 0.45 and 0.95, respectively, and the $+$ and $-$ sign are for tensile and compressive residual coating stresses, respectively. Although the factor $\psi_\lambda$ varies slightly with residual stress (compare the slopes of the results with and without residual stress in Fig. 7), Eq. (9) otherwise shows that the tensile residual stress acts similar to a decrease in coating fracture toughness, as noted in the FEM results.

To demonstrate the accuracy of Eq. (9), Fig. 8 shows the predictions of Eq. (9) against the input coating toughness for a range of coating properties (both toughness and elastic properties) and residual stress levels: the agreement is excellent in all cases. Note that the coating materials used in Fig. 8 correspond elastically to materials ranging from silicon to titanium nitride to diamond, with coating toughness as an input variable, and for a wide range of residual stresses. Although silicon was the only substrate material used in making these comparisons, the general validity of Eq. (9) across a wide range of coating/substrate systems is clearly demonstrated in Fig. 8. Thus, using the linearity of $a$ vs. $b$ found in the FEM data and an approximate energy balance argument, we have generated an analytic expression, Eq. (9), that can be used to estimate coating toughness. This is the third main result of this paper.
4.3. Toughness of CVD diamond coatings on silicon

We now use the numerical and analytical models to derive the CVD diamond coating toughness from our experimental data. The properties of the coating and substrate used in the calculations are listed in Table 3; these differ slightly from some of the values used in the FE models above due to our use of early inaccurate literature values at the time many of the FE calculations were performed. The CVD diamond/Si system has a significant residual stress and for the growth conditions used here the residual stress is 1000 MPa, as measured previously by one of the authors using the wafer curvature technique [17].

Table 3

| Parameters for microwave-enhanced CVD-diamond/silicon system |
|------------------|------------------|------------------|
|                  | $E$ (GPa) | $\nu$ | $K_{ic}$ (MPa m$^{1/2}$) | Reference |
| CVD coating      | 891       | 0.07  | $\ldots$            | [28,29]    |
| Si substrate     | 185.6     | 0.223 | 0.7$^a$              | [30]       |

$^a$ Silicon {1 1 1}, our microindentation experiment yielded the same value.

Fig. 9 shows the measured $a$ vs. $b$ for a number of indentation cracks at various loads and offset distances $s$, along with FEM predictions for two values of the coating toughness. The linear slope in the experimental data is consistent with that in the analysis, while the coating toughness clearly lies between 8 and 10 MPa m$^{1/2}$. Furthermore, as predicted by the finite element model, the distance $s$ from indent to the coating edge has a little effect on the slope $\phi$ or magnitude of the toughening. Using the measured value for the slope $\phi$ of $a$ vs. $b$, the fracture toughness of the CVD diamond film is calculated via Eq. (9) for each data point shown in Fig. 9, and the values are shown in Fig. 10 vs. the indentation load used. The mean derived value of the coating toughness $K_c$ is 8.4 MPa m$^{1/2}$. We estimate uncertainties due to fitting of the slope to yield uncertainties in $K_c$ of $\pm0.5$ MPa m$^{1/2}$, consistent with the variations seen in Fig. 10. Our derived value is independent of the loading conditions and indent position $s$, as required physically; this provides further confidence in the accuracy of the predictions.

Other data on CVD diamond exists. ASTM-style compact-tension tests on 100–400 µm thick free-standing CVD diamond yield fracture toughness values of 5.3–7.3 MPa m$^{1/2}$ [22] and 8.0–9.2 MPa m$^{1/2}$ [23]. Hehn et al. [24] give values of approximately 8 ± 1 MPa m$^{1/2}$ using the ball-on-ring test. However, such thick diamond films have a rather different microstructure than the thin diamond coatings deposited here. $K_c$ for CVD diamond thin films on substrates has been measured by many workers using standard indentation techniques, with their associated difficulties, yielding values ranging from 4.8 to 10.9 MPa m$^{1/2}$, with the values larger than natural diamond being attributed to the grain boundaries [16,25–30]. However, none of the previous works considered the effects of residual stresses. Our results without residual stresses yield a comparable value (4.8 MPa m$^{1/2}$) without significant scatter or difficulties in accurate interpretation. Our numerical

Fig. 9. Experimental coating crack length $a$ vs. substrate crack length $b$ for the CVD Diamond/Si system along with the finite element predictions for coating toughnesses of 8 and 10 MPa m$^{1/2}$ at a residual stress of 1000 MPa, with other material parameters $K_c = 0.7$ MPa m$^{1/2}$, $E_c = 185.6$ GPa, and $E_s = 891$ GPa. A coating toughness of about 9 MPa m$^{1/2}$ can immediately be deduced.

Fig. 10. Coating fracture toughness vs. indentation load as calculated using the approximate formula of Eq. (9) and the experimental data on $a$ vs. $b$, including the coating residual stress of 1000 MPa ($K_c = 0.7$ MPa m$^{1/2}$, $E_c = 185.6$ GPa, $E_s = 891$ GPa). The predicted coating toughness is essentially a constant over a wide range of crack lengths or loads, as physically expected. Dotted lines show the variation in predicted toughness associated with variations of ±100 MPa in the coating residual stress.
results show, however, that the residual stress significantly reduces the apparent fracture toughness and our analysis indicates that the true toughness of the microwave-assisted CVD diamond thin-film is about 8.4 MPa m$^{-1/2}$. The effects of residual stress, and its dependence on processing/deposition conditions, may be one reason for the wide range of previously reported fracture toughness values.

4.4. Application to other coating systems

Existing literature does not use the half-coated sample configuration proposed here. Rather, indentation is typically done directly onto a coated substrate. If delamination does not occur during such indentation, our method of analysis can be applicable, under certain key assumptions. First, as mentioned earlier, the 45°-wedge finite element model represents a configuration of indentation onto a small square patch of uncoated material surrounded by coated substrate. Therefore, following upon the arguments made to motivate the analytic model, if the differential work of indenting the coated substrate, relative to the half-coated substrate, is negligible, then our analytic model should be accurate. In other words, if the work done by the indenter in creating the indent mark and associated plastic flow and in initiating the cracks is the same for both the coated and uncoated substrate, then the analytic model is valid. To apply the model requires two indentation measurements: one on an uncoated substrate, to obtain the crack length $b$, and another on the coated substrate, to obtain the crack length $a$.

Indentation crack lengths on a bare Si wafer and on the same substrate coated with SiO$_2$ and with Cr, with coating thicknesses between 0.052 and 0.11 μm, has been reported by Zhang et al. [31]. They measured the crack length at different indentation loads as well as the residual stress in the coatings using curvature technique. Zhou et al. [32] performed similar experiments on the PZT/Si system. In these works, the crack length data and residual stresses are reported, the Young modulus and Possion ratio for the coatings ($E_{SiO_2} = 70$ GPa, $\nu_{SiO_2} = 0.17$ [33], $E_{Cr} = 200$ GPa, $\nu_{Cr} = 0.21$ [34], $E_{PZT} = 85$ GPa, $\nu_{PZT} = 0.28$ [32]) and substrate ($E_{Si} = 185.6$ GPa, $\nu_{Si} = 0.223$ [35]) have been established, and the substrate toughness of $K_{S\text{Si}} = 0.8$ MPa m$^{1/2}$ is easily calculated from the reported crack lengths $b$ on the uncoated substrates. With this information, we have simply plotted the crack length data $a$ vs. $b$, obtained the relevant slope parameter $\phi$, and calculated the coating fracture toughnesses using Eq. (9). Our results for the SiO$_2$ and PZT coatings are shown in Table 4; the predicted fracture toughness varies in a small range with changes in the coating thickness. The mean toughness values are 0.58 MPa m$^{1/2}$ for the SiO$_2$ and 0.88 MPa m$^{1/2}$ for the PZT, which are very close to the measurements of 0.58 MPa m$^{1/2}$ for fused silica [35] and 0.7–1.5 MPa m$^{1/2}$ for PZT [36]. For the Cr film, $E_{Cr}$ and $\nu_{Cr}$ are, respectively, 458 GPa and 0.30. For the Cr film, Eq. (7) generates a negative value of the fracture toughness, making Eq. (9) invalid; this demonstrates that the model is not strictly applicable to the fully coated substrate geometry in all cases. For the Cr film, it is likely the work to indent the Cr is significantly different that that required to indent the Si, leading to inapplicability of the model, rather than failure of the model (which was designed and tested for the partially coated sample geometry only). The successful predictions for the SiO$_2$ and PZT coatings indicates that our analytical model (Eq. (9)) is robust and can be used, under some circumstances, for the direct indentation on coated substrates. We strongly advocate, however, the use of our proposed partially coated sample geometry, for which the numerical and analytic models are appropriate.

5. Conclusions

A new methodology based on indentation and finite element analysis has been developed to measure the fracture toughness of thin films. Severe substrate/film delamination is avoided by fabricating a specimen in which the coating is deposited only on a portion of the substrate and then indenting the substrate near the coating edge. Comparison of the induced crack lengths in the coating and the substrate, and correlation with...
full 3D FE results, is then used to determine the film toughness. An analytic equation (Eq. (9)) for calculating the fracture toughness of thin film has been introduced that is based on an energy balance argument, and has been validated by comparison to extensive finite element results, including the effects of residual stress. The overall method has been applied to a CVD diamond coating on a silicon substrate, and a fracture toughness of 8.4 MPa m^{1/2} is obtained for a film with residual bi-axial tensile stress of 1 GPa. The present methodology, coupling a new but easily made sample geometry with a numerical and/or analytical analysis, should prove useful for accurately determining coating toughnesses in a wide range of substrate/coating systems of technological interest.

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