Dynamic response of thin films on substrates subjected to femtosecond laser pulses

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Abstract

Femtosecond laser pulses can be used to precisely debond the interface between transparent films and substrates, thus providing a critical tool for interface characterization. This paper presents a model for the dynamic response of the film in the debonded region due to excitation from the laser pulse. The model illustrates that the time-scale for transfer of energy to the film is typically much smaller than the inertial timescale associated with film bending; in such cases, the film deflection is governed entirely by the impulse delivered to the film and is insensitive to the details of the pressure generated by the laser pulse. Results for the maximum stress in the film and the dynamic energy release rate for debonding are used to generate regime maps that indicate failure modes (e.g. film fracture or interface debonding) as a function of pulse characteristics and film properties. The model is shown to exhibit good agreement with experiments, and hence, provides a simple framework to design laser experiments to generate precisely defined interface flaws while avoiding film failures.

keywords: dynamic mechanical analysis, thin film, debonding, fracture, femtosecond laser ablation

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1. Introduction

A persistent challenge in measuring and/or predicting delamination of thin films on substrates is the ability to create well defined interface flaws, which can be subjected to various loading states to extract the interface toughness controlling flaw stability [1–7]. Femtosecond laser pulses are a highly effective technique for introducing precisely defined interface flaws beneath transparent films [8–10], thus creating exciting new opportunities for interface characterization [11, 12]. A quantitative understanding of the relationship between pulse characteristics and the induced film deformation is critically needed to develop this technique, such that one can identify pulse characteristics that avoid film cracking or promote delamination (beyond the boundaries of the exposed region). This is the principle motivation of this work, which develops a model that relates pulse characteristics, film properties, and mechanical response of the film. The model can be then be used to develop effective pulse protocols to produce well defined interface flaws while avoiding film fracture.

For bare substrates, it is well established that femtosecond laser pulses effectively eject material from the surface [13, 14] with ablation depths and lateral dimensions that can be precisely controlled [10, 15, 16]. For a transparent film on a substrate, the laser pulse passes through the film and excites material in the ablation region, releasing material at the film/substrate interface and generating pressures that lead to upward motion of the film over the region that has been exposed. This process is shown schematically in Figure 1. The pressures generated in the cavity formed between the film and substrate are difficult to predict owing to the (largely unknown but undoubtedly) complex equation of state associated with phase changes of the nanoscopic layer of substrate material excited by the laser [10, 17, 18].

A seemingly overlooked aspect of the problem is the typical discrepancy in time-scales relating to the excitation of ablated material and the dynamic motion of the film. Measurements and simulations of excited material reveal that the ablated material returns to a low energy state in a period on the order of tens of nanoseconds [10, 18, 19]. Conversely, the timescale of film motion is governed by the inertia and stiffness of the film, which often produces natural frequencies of the film with a much longer period (e.g. hundreds of nanoseconds, as will be elucidated). Here, we consider scenarios wherein the motion of film (induced by the laser) is controlled by the impulse transmitted to the film from the ablated material and is therefore insensitive to the details of the pressure-volume-time relationship of the excited material. That is, in the limit that the generated
pressure pulse duration is much shorter than the inertial timescale of the film, the film response is
dictated solely by the velocity imparted to the film through momentum transfer from the material
ejected from the substrate. The model presented here is shown to produce predictions that are in
quantitative agreement with previous experimental observations of the velocity of the mass ejected
from the substrate in bare substrate experiments [20], the velocity of an oxide film [9], and film
fracture studies conducted for this work.

It should be noted that the present model requires that several key assumptions hold. The
foremost assumption is that the film is largely transparent to the laser pulse and is therefore
unaected by irradiation. Further, this means that the fraction of pulse energy imparted to the
substrate is known such that previous studies of ablation velocities and energy absorption [17, 21–
23] provide accurate estimates of the response of the substrate material just beneath the film. The
conceptual picture underlying the model is that energy absorbed by the substrate under the film
is translated to kinetic energy in the ablated material, which then impacts the film, transferring
energy and momentum over a time-scale that is much smaller than that needed to change the
position of the film. The result is that the film is subjected to an initial velocity distribution
that entirely defines the subsequent motion of the film. A second assumption is that the Gaussian
distribution of laser energy leads to dynamic response in the film that is dominated by its first
vibration mode; this is motivated by the quantitative similarity between the Gaussian profile of the
laser energy, the first mode shape of the film, and the quasi-static deflection profile arising from
uniform pressure.\(^2\) (See Figure 1C.)

Finally, the model presented in this work assumes that residual stresses in the film are negligible,
and that there is no coupling between in-plane and out-of-plane deformations (i.e. displacements
and rotations are “small”). For many systems, these effects can be important (e.g. [11]): however,
this paper identifies several key dimensionless parameters and relationships that are unaected
by these effects, as will be elucidated. A companion paper extends the analyses presented here
to include residual stress and large deflection [24], with essentially no conceptual changes to the
considerations detailed here.

\(^2\)This is simply for convenience: one could work out the response of the film using modal decomposition without
invoking this assumption.
2. Model

2.1. Dynamic response of the film subject to pressure pulse of short duration

This section describes the response of a circular film, clamped at its edges and subjected to a pressure pulse \( p(r, t) \). For small deflections and no residual stress, the dynamic equation describing the transverse deflection of the film is given by:

\[
\frac{\bar{E}h^3}{12} \left( \frac{w''(r, t)}{r} + 2 \frac{w''(r, t)}{r^2} + \frac{w'(r, t)}{r^3} \right) + \rho h \ddot{w}(r, t) = p(r, t)
\]

where \( \bar{E} = E/(1-\nu^2) \) is the plane strain modulus of the film (with \( \nu \) being the Poisson’s ratio of the film and \( E \) being the Young’s modulus), \( h \) is the film thickness, \( w \) is the transverse (out-of-plane) displacement, \( \rho \) is the film density, \( p(r, t) \) is the pressure distribution applied to the film, and primes denote differentiation with respect to the radial coordinate. For a circular film with radius \( a \), the imposed boundary conditions are \( w(a, t) = w'(a, t) = 0 \) and \( w'(0, t) = 0 \), along with the condition that \( w(0, t) \) is finite.

For any pressure loading, the response can be written in terms of the modal superposition given by:

\[
w(r, t) = \sum_{n} z_n(t) W_n(r)
\]

where \( z_n(t) \) represents the time-dependent contribution of mode \( n \) and \( W_n(r) \) defines the mode shape for the \( n^{th} \) mode. The mode shapes for the present boundary conditions are given by:

\[
W_n(\beta_n, \bar{r}) = \frac{I_1(\beta_n)J_0(\beta_n \bar{r}) + J_1(\beta_n)I_0(\beta_n \bar{r})}{I_1(\beta_n) + J_1(\beta_n)},
\]

where \( \bar{r} = r/a, \ J_{0,1} \) are the Bessel functions of the first kind and \( I_{0,1} \) are the modified Bessel functions of the first kind. The constants \( \beta_n \) are defined by the roots of the characteristic equation:

\[
I_1(\beta)J_0(\beta) + J_1(\beta)I_0(\beta) = 0,
\]

and in turn, they define the natural frequencies according to:

\[
\omega_n = \beta_n^2 \sqrt{\frac{\bar{E}h^2}{12 \rho a^4}}
\]
The amplitudes $z_n(t)$ are determined by the solutions to:

$$
\ddot{z}_n(t) + \omega_n^2 z_n(t) = f_n(t); \quad f_n(t) = \left( \frac{1}{\rho h} \right) \int_0^1 \bar{r} W_n(\bar{r}) p(\bar{r}, t) d\bar{r} \int_0^1 \bar{r}^2 W_n^2(\bar{r}) d\bar{r} \tag{7}
$$

where $f_n(t)$ is a generalized force that has units of acceleration.

In order to elucidate the fundamental scaling controlling the dynamic response of the film, we adopt a single term approximation using only the first mode. The approach is exact in the limit that the spatial distribution of pressure is identical to the first mode shape, and it can be expected to be highly accurate for pressure distributions of similar shape. For the first mode, $\beta_1 = 3.19622$ and $W_1(\bar{r}) = 0.053 J_o(\beta_1 \bar{r}) + 0.947 J_o(\beta_1 \bar{r})$. In the single term approximation, the film deflects with the shape $W_1(\bar{r})$, with the peak deflection (at $\bar{r} = 0$) determined by the time-dependent pre-factor $z_1(t)$. Here, we use $w_o(t)$ to denote the peak center point deflection, i.e. $w_o(t) = z_1(t) = w(0, t)$.

Further, to demonstrate that the time-dependence of the pressure is immaterial for pulses of short duration, consider the case where the pressure distribution is assumed to be $p(\bar{r}, t) = p_o (\sin \pi t/t_o) W_1(\bar{r})$ for $t \leq t_o$ and $p(\bar{r}, t) = 0$ for $t > t_o$. That is, we assume the pressure rises to a peak magnitude of $p_o$ over $t < t_o/2$ and falls to zero pressure at time $t = t_o$. In this case, the dynamic equation of motion and solution for $0 < t \leq t_o$ are given by:

$$
\ddot{w}_o + \omega_1^2 w_o = \frac{p_o}{\rho h} \sin (\pi t/t_o); \quad w_o(t) = \left( \frac{p_o t_o}{\rho h} \right) \frac{\omega_1 t_o \sin (\pi t/t_o) - \pi \sin (\omega_1 t)}{\omega_1^4 t_o^2 - \pi^2 \omega_1^2} \tag{8}
$$

At the end of the pressure pulse $t = t_o$, the displacement and velocity of the center point of the film are:

$$
w_o(t_o) = \frac{\pi p_o t_o}{\rho h (\pi^2 - \omega_1^2)} \sin \omega_1 t_o = \frac{p_o t_o^2}{\rho h} \left( 1 + O \left( (\omega_1 t_o)^2 \right) \right) \tag{9}
$$

$$
\dot{w}_o(t_o) = \frac{\pi p_o t_o}{\rho h (\pi^2 - \omega_1^2)} \left( 1 + \cos \omega_1 t_o \right) = \frac{2 p_o t_o}{\rho h} \left( 1 + \frac{\omega_1 t_o}{\pi^2} + O \left( (\omega_1 t_o)^2 \right) \right) \tag{10}
$$

Thus, we can see that when $\omega_1 t_o \ll 1$, the film experiences negligible motion during the pressure pulse, and acquires an initial velocity dictated by the peak pressure and the duration of the pulse.

\textsuperscript{3}Note that this decoupled set of single degree of freedom dynamic equations is a consequence of the fact that the modes are orthogonal over the domain of interest, such that:

$$
\int_0^1 W_n(\bar{r}) W_m(\bar{r}) d\bar{r} = 0 \text{ for } m \neq n \tag{6}
$$
However, conservation of momentum implies that the momentum of the film at the end of the pressure pulse \((t = t_o)\) is equal to the impulse delivered by the pressure pulse. That is, if we compute the momentum at \(t = t_o\) using velocity distribution given by the first mode shape,

\[
M = \int_0^a 2\pi r p h \dot{w}(r, t_o) dr = 0.979 a^2 p h \dot{w}_o(t_o)
\]

and equate this result to the impulse of the pressure pulse,

\[
Im = \int_0^{t_o} \left( \int_0^a 2\pi r p(r, t) dr \right) dt = 0.623 a^2 p_o t_o,
\]

we obtain the same velocity at \(\dot{w}_o(t_o)\) as we did using the pressure pulse (noting that \(2/\pi = 0.623/0.979\)).

Thus, by re-defining zero time as the end of the pressure pulse, we can compute the initial velocity from the momentum acquired during the pressure pulse and ignore the details of the pressure-time excitation of the film. This is justifiable when the timescale of the pressure pulse is small in comparison to the natural period (inverse frequency) of the film. For oxide films with micron-scale thickness and spot sizes \((a)\) in the tens of microns, one computes \(1/\omega_1\) is on the order of hundreds of nanoseconds. However, estimates of shock pressures based on pump-probe shadowgraphic microscopy reveal that the pressures in the shock front created by ablated material decrease from several GPa to several tens of MPa in the first ten nanoseconds for fluences < 3 J/cm\(^2\) [20]. Therefore, in the remainder of the paper, we compute \(\dot{w}_o\) from energy/momentum considerations of the laser pulse, and use this as the initial condition that dictates the dynamic response of the film.

2.2. Estimate of impulse delivered during laser pulse

Here, we presume that the laser pulse ejects a small mass from the substrate that impacts the film. The details of this process \((i.e.\) the time/space details of the colliding masses) are unimportant, provided the collisions of all ejected particles with the film occur over a time scale that is small compared to the inertial time scale of the film \((i.e.\) in the above, that \(\omega_1 t_o \ll 1\)). Let us assume that the laser pulse imparts a total energy of \(\pi a^2 \phi_{avg}\) to the substrate, and that the total energy of the ejected particles is \(f \pi a^2 \phi_{avg}\), such that \(f\) represents the fraction of laser energy that is converted to kinetic energy in the ejected mass and \(\phi_{avg}\) is the average fluence of the laser pulse.
Let $d(r)$ be the spatial distribution of the depth of ejected particles, and $v^{(1)}(r)$ be the spatial distribution of the velocity of the ejected particles prior to any collision. The momentum of the system prior to impact is simply that of the particles, and is given by:

$$M^{(1)} = \int_{0}^{a} 2\pi \rho_s d(r) \left[ v^{(1)}(r) \right] r dr$$  \hspace{1cm} (13)

where $\rho_s$ is the density of the substrate. After impact, the momentum of the system involves contributions from both the reflected particles and the film motion:

$$M^{(2)} = \int_{0}^{a} 2\pi \rho_s d(r) \left[ v^{(2)}(r) \right] r dr + \int_{0}^{a} 2\pi \rho h \dot{w}(r) r dr$$  \hspace{1cm} (14)

where $v^{(2)}(r)$ is the velocity distribution of the ejected particles after impact, and $\dot{w}(r)$ is the velocity of the film at the end of the impact. For purely elastic impact, the kinetic energy before and after impact is conserved, such that:

$$\int_{0}^{a} \pi \rho_s d(r) \left[ v^{(1)}(r) \right]^2 r dr = \int_{0}^{a} \pi \rho_s d(r) \left[ v^{(2)}(r) \right]^2 r dr + \int_{0}^{a} \pi \rho h \left[ \dot{w}(r) \right]^2 r dr$$  \hspace{1cm} (15)

Note that there is no potential (strain) energy in the film after impact, because the assumption of a short impact duration implies the film does not deform during the impact.

We must now make assumptions regarding the spatial distribution of particles ejected from the substrate, the velocity distribution before impact, and the velocity distribution after impact. These distributions are ultimately controlled by the distribution of the fluence in the laser pulse, e.g. a Gaussian distribution. For simplicity, we assume both distributions (the depth of ejected material and the resulting velocity) have the same spatial distribution, and further, that it is given by the first mode shape of the film $W_1(\bar{r})$. That is, the assumed spatial distribution of ejected mass and ejection velocity is maximum in the center and zero at the edges where it has zero slope, as shown in Figure 1C. This approximation is reasonable given the qualitative similarity between a Guassian distribution and the first mode shape (see Figure 1C).

Thus, the distributions of the depth of ejected mass, the velocity of the ejected mass, and the film deflection are given by:

$$d(r) = d_o W_1(r); \quad v(r) = v_o W_1(r); \quad w(r) = w_o(t) W_1(r)$$  \hspace{1cm} (16)

where $d_o$, $v_o$, and $w_o$ represent the maximum ejected depth, maximum ejected velocity, and maximum film deflection. This implies that the momentum equations given above become:

$$M^{(1)} = 0.57 \rho_s a^2 d_o v^{(1)}_o = M^{(2)} = 0.57 \rho_s a^2 d_o v^{(2)}_o + 0.98 \rho h a^2 \dot{w}_o$$  \hspace{1cm} (17)
For elastic impacts, the energy is conserved such that the ejected particles will rebound with finite kinetic energy. The energy balance expressions are given by:

\[
\frac{f \pi \rho \phi_{avg} a^2}{2} = 0.2 \rho_s d_o a^2 \left[ v_{o}^{(1)} \right]^2 \]

\[
0.2 \rho_s d_o \left[ v_{o}^{(1)} \right]^2 = 0.2 \rho_s d_o \left[ v_{o}^{(2)} \right]^2 + 0.29 \rho h \left[ \dot{w}_o \right]^2 \]

where Eq. 18 represents the translation of laser energy to kinetic energy, and the Eq. 19 represents conservation of energy before and after impact. (Note again that the film acquires no potential energy during the impact event, as it does not move over this small timescale.) Equations 17-19 involve three unknowns: the initial velocity of the ejected mass prior to impact, and the velocities of the ejected mass and film after impact. The solution yields:

\[
v_{o}^{(1)} = 2.24 \sqrt{\frac{f \pi \phi_{avg}}{\rho_s d_o}}; \quad \dot{w}_o = 5.30 \sqrt{\frac{f \pi \phi_{avg}}{\rho_s d_o}} \left( \frac{1}{1 + 2.04 \frac{\rho h}{\rho_s d_o}} \right) \]

For purely plastic impacts, the energy of the system is not conserved and we simply have the momentum balance before and after impact. Plastic impact implies the velocity of the ejected material after impact is equal to that of the film after impact, such that Eq. 19 is replaced by:

\[
0.57 \rho_s d_o a^2 v_{o}^{(1)} = (0.57 \rho_s d_o a^2 + 0.98 \rho h a^2) \dot{w}_o \]

Solving for the initial ejected velocity \( v_{o}^{(1)} \) and the initial film velocity \( \dot{w}_o \) yields:

\[
v_{o}^{(1)} = 2.24 \sqrt{\frac{f \pi \phi_{avg}}{\rho_s d_o}}; \quad \dot{w}_o = 2.24 \sqrt{\frac{f \pi \phi_{avg}}{\rho_s d_o}} \left( \frac{1}{1 + 0.72 \frac{\rho h}{\rho_s d_o}} \right) \]

One observes that the initial film velocity is approximately one-half the result for an elastic impact. Comparisons between these predictions and direct observations from experiments are presented in Section 3.

2.3. Film failure and interface debonding

The results detailed above can be used to evaluate two different failure scenarios: (i) the film cracks at the outer edge of the laser pulse, leading to spalling of the film above the exposed area, and (ii) the film debonds at the outer edge of the exposed area. Evidence of these two failure scenarios are shown in Figure 2 for a silicon dioxide film grown on a silicon substrate; in Figure 2A, the final deformed shape of the film as measured via interferometry is shown for a laser pulse...
of relatively low fluence ($\phi_{avg} \approx 230 \text{ mJ/cm}^2$). At low fluences, the film remains intact and comes to rest in a slightly deformed state due to ejected material attached to the backside of the film, residual stresses in the oxide, or both. (A micrograph of the surface is essentially featureless due to the small displaced amplitude of the buckle.) At moderate fluences ($400 < \phi_{avg} < 450 \text{ mJ/cm}^2$), one observes the formation of cracks around the outer edge of the exposed region, as shown in Figure 2B. At high fluence ($\phi_{avg} > 550 \text{ mJ/cm}^2$), the entire film cracks along the periphery of the exposed region, spalling a circular section of film (Figure 2C).

Here, we outline the conditions that lead to either type of failure: in the next section, we discuss which failure mode is anticipated. The dynamic response of the film is

$$w(\bar{r}, t) = \left(\frac{\dot{w}_0}{\omega_1}\right) W(\bar{r}) \sin \omega_1 t,$$

which results from an initial velocity distribution given by:

$$\dot{w}(\bar{r}, 0) = \dot{w}_0 W(\bar{r}) \quad \text{(where } t = 0 \text{ references the end of the impact event)}.$$

The bending strains in the film are given by:

$$\epsilon_\theta = \frac{hw'(\bar{r}, t)}{2r}; \quad \epsilon_r = \frac{hw''(\bar{r}, t)}{2};$$

where the primes denote differentiation with respect to $\bar{r}$. Using the first mode shape, the peak strains (with respect to time) in the film are given by:

$$\epsilon_r(0, t) = \epsilon_\theta(0, t) = 2.28 \frac{h\dot{w}_0}{\omega_1 a^2} \sin \omega_1 t = 0.77\dot{w}_0\sqrt{\frac{\rho}{E}} \sin \omega_1 t \quad (23)$$

$$\epsilon_r(a) = 3.09 \frac{h\dot{w}_0}{\omega_1 a^2} \sin \omega_1 t = 1.05\dot{w}_0\sqrt{\frac{\rho}{E}} \sin \omega_1 t \quad (24)$$

with monotonic variations from the center to the outer edge. (Note $\epsilon_\theta(a) = 0$.) Thus, the radial strains are maximum at the outer edge of the film, while the hoop strains are maximum in the center of the film. The film will fail when the induced bending stress at the outer edge ($r = a$, or $\bar{r} = 1$) is larger than the intrinsic strength of the material. It should be noted that while circumferential cracking is the most common failure mode in these experiments, many lower-fluence experiments involve radial cracks emanating from the center. This is not surprising in light of the facts that the hoop stress in the film is only slightly lower than the radial stress and that the failure location is strongly influenced by the spatial distribution of film flaws.

If we assume the presence of small flaws of size $c$ in the film, we can solve for the critical strain that produces film failure according to elastic fracture mechanics. Assuming an edge flaw in the film much smaller than its thickness and loading in pure bending, fracture will be avoided provided
that:
\[ \dot{w}_o^f < \sqrt{\frac{\Gamma_f}{4\rho c}} \]  
(27)
where \( \Gamma_f \) is the toughness of the film.

Interface debonding at the outer edge of the film can be predicted using the dynamic energy release rate [25–27]. For a penny shaped crack of radius \( a \), centered at the origin in an isotropic medium, the full axisymmetric result for the dynamic J-integral is given by:
\[
J = \frac{1}{a} \left[ \int_{\Gamma_o} (Wn_r - T_r u' - T_z w') r d\ell - \int_{A_o} \left( W - T_{\theta r} \frac{u}{r} \right) dA + \int_{A_o} \rho (\ddot{u} \cdot u' + \ddot{w} \cdot w') r dA \right]
\]
where \( \Gamma_o \) defines a contour surrounding the crack tip, and \( A_o \) is the area inside the contour. In the above definition, \( u \) and \( w \) are the radial and vertical displacements as a function of position, respectively. For axisymmetric plates subjected to small deflections, the vertical displacement \( w \) is uniform through the thickness of the plate, while bending implies \( u' = -zw'' = \varepsilon_r \), and \( u = -zw' = r\varepsilon_\theta \). Hence, the J-integral for the axisymmetric plate problem becomes:
\[
J = \frac{1}{a} \left[ \int_{\Gamma_o} (Wn_r - T_r z w'') r d\ell - \int_{A_o} \left( W - T_{\theta r} \frac{z w'}{r} \right) dA + \int_{A_o} \rho \left( z^2 \ddot{w}' \cdot w'' + \ddot{w} \cdot w' \right) dA \right]
\]  
(28)

Taking a contour that runs through the film along the symmetry axis, along the top surface and back through the film at a radial location far past the crack tip, the first term (i.e. the path integral) is zero, leaving just the remaining two terms (i.e. area integrals). Using the first mode shape, one computes:
\[
J = 1.59 \frac{E h^5}{a^4} [\bar{w}_o(t)]^2
\]  
(29)
Note that the deflection of the center \( \bar{w}_o(t) \) for the impulse problem is given by \( \bar{w}_o(t) = (\dot{w}_o/\omega_1) \sin \omega_1 t \) (where \( \dot{w}_o \) is the initial velocity at the center, as defined earlier). Using the definition of the first natural frequency given earlier, this implies:
\[
J(t) = 0.18 \rho h \dot{w}_o^2 \sin^2 \omega_1 t
\]  
(30)
Interface debonding can be expected when \( J \geq \Gamma_i \), where \( \Gamma_i \) is the interface toughness. Here, we assume an ideally brittle interface with no mode-mixity effects; this is discussed further in the next section. Thus, debonding is avoided provided that:
\[
\dot{w}_o^d < \sqrt{\frac{5.47 \Gamma_i}{\rho h}}
\]  
(31)
3. Discussion

The model prediction for the velocity of the material ejected by the laser pulse (i.e. \(v^{(1)}_o\)) can be compared to direct measurements obtained using pump-probe shadowgraph microscopy on a nickel alloy without an oxide film [20, 28]. These measurements track the location of the shock front as a function of time, which enables calculation of the material velocity leaving the surface. Using Taylor-Sedov models to back-calculate the energy of the shock-wave, one can then compute the kinetic conversion factor \(f\). The ablation depth \(d_o\) and size \(a\) were directly measured using AFM profiles of the ablated surface. Experiments on the nickel-based superalloy CMSX-4 suggest that the initial velocity is approximately \(10 - 12\ km/s\) for a fluence of \(2.7\ J/cm^2\). (This velocity range was estimated based on visual inspection of Figure 3a in [20]; the resolution of the presented data at short times makes it difficult to identify the initial velocity with more precision.)

The measured ablation depth for this fluence was \(d_o \sim 50\ nm\), with an ablated radius of \(a = 9\ \mu m\); the extracted kinetic conversion factor was \(f = 0.123\). Using Eq. 23 above, one computes \(v^{(1)}_o = 10.8\ km/s\) for this case, which is a strong endorsement of the above model. For high fluences \((\phi_{avg} > 60\ J/cm^2)\), the model appears to under-predict the observed initial velocity by a factor of 1.5-2. However, the surface profiles of the ablated surface at high fluence exhibit ‘pile-up’ at the outer edges [16, 20], presumably from more extensive melting, re-deposition of ablated material, or both. This implies that the model is overestimating the mass of material that is accelerated to leave the surface, and hence produces an estimate for velocity that is lower than the measurements. The agreement between predicted and measured velocities at high fluence is much better if the maximum ablation depth is estimated using the volume of ejected material [29] (i.e. by adjusting the value of \(d_o\) to account for material in the pile-up region that has not left the surface).

Similar experimental observations made on silicon substrates coated with thin films of silicon dioxide enable direct comparisons of the film velocity after the pulse, \(\dot{w}_o\). The film velocity after the pulse is shown in Figure 3 for this system with a laser fluence of \(1.3\ J/cm^2\), using the data from McDonald et al [9] and kinetic conversion factors in the range of \(0.01 < f < 0.1\). The observed ablation depth at this fluence is approximately \(80\ nm\) [28]. The agreement between predicted velocity (Eq. 20) and measured velocity is excellent if one assumes plastic impact and \(f = 0.02\). Plastic impacts, where amorphous silicon adheres to the back side of the buckled oxide, have been directly observed experimentally and will be detailed in a future publication.
An important application of the present model is the specification of laser pulses that introduce a well-defined interface flaw while avoiding failure, which can be used for subsequent studies of film delamination. For a given laser pulse, one can determine whether cracking or debonding occurs by comparing the initial film velocity with the critical values associated with each failure mode. There are four possible scenarios:

(i) neither failure mechanism is possible: this happens when the kinetic energy density imparted to the film, $\rho \dot{w}_o^2$, is less than both $5.47 \Gamma_i/h$ and $\Gamma_f/(4c)$.

(ii) film fracture is possible but debonding is not: this happens when $\rho \dot{w}_o^2$ is greater than $\Gamma_f/(4c)$ but less than $5.47 \Gamma_i/h$.

(iii) debonding is possible but film fracture is not: this happens when $\rho \dot{w}_o^2$ is greater than $5.47 \Gamma_i/h$ but less than $\Gamma_f/(4c)$.

(iv) both debonding and film fracture are possible: this happens when $\rho \dot{w}_o^2$ is greater than both $5.47 \Gamma_i/h$ and $\Gamma_f/(4c)$.

These four conditions are shown graphically in Figure 4, which provides a universal map indicating relevant failure modes as a function of system properties. In this figure, data points corresponding to laser pulses applied to a silicon/silicon dioxide system are overlaid: the values associated with these data points correspond to multiple levels of laser fluence, spot size and two values of oxide thickness (1.3 and 3.0 $\mu$m). The relationship between laser fluence and ablation crater depth $d_o$ is taken from the work of Coyne et al [30]. The critical flaw size $c$ is assumed to be 15 and 45 nm, respectively. The film toughness and interface toughness are assumed to be 1 J/m$^2$. The error bars correspond to $\pm 50\%$ changes in toughness and flaw size.

In Figure 4, agreement between the experiment and predictions is ensured by the choice of the defect size in the film. While direct observations of flaw sizes would be ideal (thus removing any fitting parameter), it is difficult to envision how such observations would be made, given the high quality of oxide films grown on silicon. Moreover, one would need an accurate idea of the flaw size distribution and spatial distribution of flaws in the area exposed to the laser pulse. Clearly, the model implies flaw sizes that are reasonable (not negligible but also not a significant fraction of the film thickness). Thus, the model combined with high throughput laser pulses distributed across a surface presents an opportunity to rapidly assess the flaw size distribution in oxides.

To further illustrate the agreement of the model and the Si/SiO$_2$ experiments, Figure 5 plots the
observations of cracking as a function of laser fluence and film thickness. The theoretical prediction for the critical fluence to cause cracking and debonding is also shown, using the same properties described above. For the 1.3 µm thick oxides, experiments were conducted using two optical lenses with different focal lengths, resulting in distinct laser spot sizes: one of them is slightly displaced to 1.35 µm thickness such that one can distinguish between these two different experiments. The agreement is quite good, again due to the chosen flaw size.

In Figures 4 and 5, one observes that some of the experiments were conducted with laser pulses where debonding is predicted. Determining whether or not debonding occurs is quite challenging, as the films remain essentially flat. Large exposed areas can lead to buckling with small deformed amplitudes, but even then it is difficult to identify the extent of debonding. Generally, for the experiments on the Si/SiO₂ system shown in Figures 3-5, obvious evidence of debonding does not occur significantly outside the exposed area. (By contrast, for nickel alloy/Al₂O₃ systems, with much larger residual stresses in the film, there are obvious indications of significant debonding beyond the exposed area; this will be reported in a future publication.) Hence, comparison of experiments in Figure 4 with the debonding axis is meaningless because evidence of debonding was not obtained.

Additional experiments and simulations are required to address the validity of the model with regards to debonding. It is worth emphasizing that the model indicates whether or not dynamic debonding initiates, but does not address the extent of delamination. It seems reasonable to believe that if the pulse excites the film to just above the threshold for debonding, the extent of debonding will be rather small. The peak driving force for debonding is obtained near the peak deflection, such that the driving force decreases a short time after the peak as the film returns to its initial position. Simply put, there may not be a large enough period of time where the energy release rate is higher than the critical value needed to advance the interface crack. Further, the model does not address whether or not the film fractures after dynamic debonding has occurred. Debonding may not dramatically lower the dynamic stresses in the film; cracking after debonding is a real possibility when one considers that the killer flaw in the film may lie just outside the exposed area. (Presumably, if fracture occurs prior to debonding, the driving force for delamination will drop precipitously as cracking alleviates strain energy in the film.)

While the above considerations rule out conclusive statements regarding debonding in the
present experiments, it is interesting to consider the underlying scaling in Figure 4. The “no failure” region extends to the experimental boundary between fractured and intact films if one assumes an interface toughness of $\Gamma_i = 3 \text{ J/m}^2$ (i.e., a value three times larger than used to plot the data points). Such values of interface toughness may be appropriate when one considers that interface debonding is mixed-mode. The point to this observation is that plausible values of interface toughness put the critical condition for cracking and debonding in close proximity, at least for films with relatively small internal defects.

The presented model also suggests which failure mode occurs first in time, which may provide additional insight with regards to interpreting future experiments. The time at which each failure condition is reached can be computed using the above results, which yields the time-to-fracture ($\omega_1 t^f = \sin^{-1} \dot{\bar{w}}_0^f/\dot{\bar{w}}_o$) and the time-to-debonding ($\omega_1 t^d = \sin^{-1} \dot{\bar{w}}_0^d/\dot{\bar{w}}_o$). These results indicate that the failure mode is indicated by the smaller of the values of $\dot{\bar{w}}_0^f/\dot{\bar{w}}_o$ and $\dot{\bar{w}}_0^d/\dot{\bar{w}}_o$. The time to reach both failure modes is equal when the ratio given by:

$$\frac{\dot{\bar{w}}_o^d}{\dot{\bar{w}}_o^f} \approx 22 \frac{\Gamma_i c}{\Gamma_i h}$$

is equal to unity, which defines a straight line in Figure 4 that delineates between “film fracture first” and “dynamic debonding first”. That is, for systems with $\dot{\bar{w}}_0^d/\dot{\bar{w}}_o^f < 1$ debonding will occur first (provided the debond parameter is above the threshold), while for $\dot{\bar{w}}_0^d/\dot{\bar{w}}_o^f > 1$, film fracture will occur first (again, provided the fracture parameter is above the threshold).

Although the placement of the observations in the failure map requires an estimate for the flaw size, the model clearly captures the essential scaling of the problem. The agreement of the model and the observations of cracking suggest that residual stress may induce off-setting effects (at least for low values of compressive stress): compressive stresses will make the film more compliant for large deformations, but also will decrease the total mechanical strain in the film at the edge of the debonding region. Ongoing modeling and simulations are addressing the role of residual stress, which is clearly going to play a key role in other systems, notably oxides grown on nickel-based superalloys.

Finally, it should be noted again that the above treatment does not address the role of mode-mixity in delamination [31], which is known to have a large effect in the static response of ductile systems. Unfortunately, the model neglects a key factor in mode-mixity: the role of axial displacements in the film and their dynamic (temporal) behavior. More sophisticated modeling and
simulations will be needed to address this aspect of the problem; once conducted, an effective mixed-mode phase angle for dynamic debonding might be identified, as has been done for quasi-static systems prone to buckling (i.e. a phase angle of $-70^\circ$ [32]). One would then use the toughness at the ‘equivalent’ phase angle in the above models.

4. Conclusions

Femtosecond laser pulses that ablate material at a film/substrate interface generate a complex time-history of internal pressure, which can ultimately lead to film cracking or debonding. For many films, the pressure burst occurs over timescales that are much shorter than the characteristic timescale of film motion (as determined by the film’s bending stiffness and inertia). This implies that the film motion can be accurately predicted using the impulse delivered by the pressure pulse, obviating the need to resolve its complicated space-time relationships.

The impulse delivered to the film can be computed from conservation of linear momentum (for plastic impacts of the ablated material) and conservation of energy (for elastic impacts), provided one has an estimate of the mass of material that is released from the substrate and its initial velocity. The initial velocity of ablated material is well approximated by equating the laser pulse energy to the kinetic energy of the mass leaving the surface using a conversion factor that can be determined experimentally. Calculations of the impulse delivered to the film using this model and direct observations of system properties lead to accurate predictions of the initial velocity of the film, as verified by direct observations of film velocity.

A conventional dynamic film model for the film motion after the impulse yields the displacement and stress distributions in the film as a function of space and time, which can be used to predict film fracture from pre-existing flaws or film debonding using a dynamic form of the J-integral. A comparison of cracking observations in the Si/SiO$_2$ system and the model indicates that the model will be useful in the design of laser pulse experiments that produce well-defined interface flaws while avoiding film failures. Future work is necessary to quantify the impact of residual stresses in the film, which may alter the dynamic response by introducing coupling between in-plane stresses and out-of-plane motion. Similarly, future work is needed to identify the extent of dynamic debonding once initiated.
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References


Figure 1: (A) Schematic illustration of the laser fluence distribution that passes through the film to excite material at the film/substrate interface, (B) schematic illustration of the film deformation that is triggered by impact of ablated material (assuming plastic impact), and (C) a comparison of the quasi-static deflection profile for uniform pressure, a Gaussian distribution (i.e. the laser energy) and the first vibration mode.
Figure 2: Film responses at (a) low fluence - debonding and no fracture in a 1.3 μm SiO₂ film on Si with a 400 mm focal length lens, (b) moderate fluence - partial fracture in a 1.3 μm SiO₂ film on Si with a f=200 mm lens, (c) high fluence - complete spalling in a 3 μm SiO₂ film on Si with a f=400 mm lens. In this case, it is difficult to confirm the existence of a small amount of interface debonding that may have occurred before film fracture.
Figure 3: A comparison of predicted and measured initial velocities involving silicon dioxide films on silicon substrates [9], using the ablation properties cited in [28].

Data from McDonald, et al., JAP (2007)

Initial oxide velocity, m/s

Oxide film thickness, µm

Converted energy fraction:
\[ f = \frac{E_{\text{kinetic}}}{E_{\text{pulse}}} = 0.1 \]

plastic impact

\[ f = 0.02 \]

\[ f = 0.01 \]
Figure 4: Regime map indicating whether film debonding or film fracture will be triggered by the laser pulse; the initial velocity of the film $\dot{w}_0$ is related to the pulse characteristics as shown above the figure. The line delineating between debonding and fracture regimes is described by equating the film debonding and film fracture parameters. Experimental data are from a series of experiments conducted with 1.3 and 3 $\mu$m thermally grown SiO$_2$ films on Si substrates with 200 and 400 mm focal length lenses.
Figure 5: Failure map indicating the critical average fluence required to trigger debonding and film fracture for the case of silicon dioxide on a silicon substrate. Experimental data are the same as in Figure 4.