Conf-931054--7

UCRL -JC-113493
PREPRINT

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This paper was prepared for submittal to
The 1993 Boulder Damage Symposium
Boulder, Colorado
October 27-29, 1993

December 21, 1993

Lawrence Livermore National Laboratory

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ABSTRACT

Laser induced damage to optical coatings is generally a localized phenomenon associated with coating defects. The most common of the defect types are the well-known nodule defect. This paper reviews the use of experiments and modeling to understand the formation of these defects and their interaction with laser light. Of particular interest are efforts to identify which defects are most susceptible to laser damage. Also discussed are possible methods for stabilizing these defects (laser conditioning) or preventing their initiation (source stabilization, spatter particle trapping).

1. INTRODUCTION

It has been known for over 20 years that defects act as initiation sites for laser-induced damage process in optical coatings [1]. Here we use the term "defects" to mean the μm-scale inhomogeneities that can be seen optically in almost all coatings. Experiments using laser beams with small spot sizes have shown that defects damage at fluences significantly lower than that of the "defect-free" material, often by factors of greater than 2 or 3 [2]. The general form of the μm-scale defect is the well-known nodule defect. While these nodules are likely responsible for the localized nature of optical coatings damage, other defects, such as structural defects (grain boundaries, columnar growth) and point defects, also play an important role in determining the mechanical stability and absorption levels in the coatings. We focus here only on the nodule-type defects. While significant progress has been made in improving the damage thresholds of optical coatings over the last two decades, several questions concerning nodule growth and interaction with the laser continue to be asked. These include:

Which defects are most susceptible to laser damage and why?
What are the sources of the seeds that initiate the nodule growth?
How can we nondestructively evaluate the defects in coatings?
How can we minimize the impact of the nodules on the coating performance?

The following paragraphs will discuss the current status of each of these questions.

2. NODULAR DEFECTS

2.1 Nodule geometries

Insight into the questions posed above can be gained by examining the internal structure of the nodular defects. Figure 1 shows cross-sections of two defects in a HfO_2/SiO_2

multilayer mirror coating. The cross-sections shows a seed that initiated the defect and a cone-shaped structure resulted from the deposition of material on the seed. The shape of the nodule and the characteristic (or nature) of the nodule-coating interface is influenced by the seed size and shape. As indicated in Fig. 1, as the seed size becomes large the nodule structure, particularly at the nodule/coating interface, becomes less ideal, i.e. the layered structure is not continuous and voids may be present [3]. For very large seeds the defect may not resemble a nodule at all, as shown schematically in Fig. 1c. The latter type of defect is sometimes called a spatter defect [4].

The cross-sections shown in Figs. 1a and 1b were obtained using the focused ion-beam (FIB) milling technique. This technique, used extensively in the semiconductor chip industry, allows the cross-sectioning of individual defects without the need for sectioning the whole sample. Earlier studies of nodule structures using scanning electron microscope (SEM) cross-sectioning methods made it difficult or impossible to target particular defects.

2.2 Seed sources

The most significant improvements in eliminating defects in coatings in the last 20 years has been due to the elimination of seed sources. Figure 2 shows schematically many of the sources of seeds particles from the chamber and the substrate [5]. Defect cross-sections indicate that most defects start within the coating rather than on the substrate, suggesting that good polishing and cleaning procedures were followed. If state-of-the-art vacuum system procedures are followed, seed particles resulting from venting, pumping, or the motion of mechanical part in the chamber should also not be significant. Seeds caused by flaking of coating material from the chamber walls can be eliminated by frequent re-foiling of the chamber. Insight into the sources of defect seeds can be gained by examining the shape and chemical composition of the seeds in cross-section.

Using the cleanest coating procedures, the main source of seed material is then the coating material evaporation source itself. Figure 3 shows that the heating of the source material can produce seeds by several mechanisms including: explosions caused by the heating of gas inclusions or μ-arcing; splashing of molten material; electrostatic repulsion of charged particles; thermal-induced cracking; and temperature-induced solid-state phase transitions. The last mechanisms is likely a problem in HfO_2 and ZrO_2 where phase changes cause 3% and 7% volume changes, respectively, at temperatures near 1700^oC [6]. Recent experiments have shown that in HfO_2/SiO_2 mirror coatings that the seeds are in fact HfO_2 [3]. The smooth, near spherical nature of the seeds in Figs. 1a and b suggests that the material ejected from the source was molten rather than solid.

2.3 Growth models

The simplest geometric model for nodules is based on an omnidirectional coating flux [7]. This model, shown in Fig. 4, agrees qualitatively with experimental observations. The model predicts the parabolic shape of defects observed experimentally but the constant is often incorrect [3]. The correspondence between defect height and seed diameter has been consistently demonstrated experimentally. Once calibrated to a particular coating operation, this model can be used to nondestructively determine seed size and depth by measuring nodule diameter and height with a device such as the AFM.

The influence of deposition angle, rotation and molecular interactions, such as surface migration and capture cross-section, have been studied using hard-disk model computer simulations [8,9]. In general, the diameter of the nodule increases with the incident angle and the capture cross-section. Cross-sections of defects produced under the appropriate conditions have shown some qualitative agreement with the trends predicted by the computer simulations. Quantitative models that can predict less ideal structures such as that shown in Fig. 1b have not been developed however.

3. NODULAR/LASER INTERACTIONS

3.1 Experimental

While damage generally occurs at defects, it is observed that not all defects damage at the same fluence. It is, therefore, of interest to determine which defects are most susceptible to damage. Optical techniques have not been very successful in this quest, as scatter, luminescence and defect diameter have not correlated well with local damage thresholds. The atomic force microscope (AFM), however, has recently provided the capability to provide information on nodule-dome height, nanometer-scale coating morphology, and damage propagation from the defects. [2,10,11]

At fluences near the defect's damage threshold the nodules may be ejected cleanly leaving a smooth crater that does not couple to further laser illumination. At higher fluences nodule ejection may leave a crater with rough edges and cracks that can be sites for further damage [10]. The mechanical stability of the defects, as determined by the nodule/coating boundary, will clearly be important here.

In studies of HfO_2/SiO_2 e-beam deposited mirrors it was determined that nodules with heights greater than ~0.7 μm were highly susceptible to laser damage [11]. No clear correlation of damage susceptibility with nodule diameter was found, however. These observations further emphasize the importance of seed size in understanding both nodule growth and laser damage.

3.2 Mechanisms

In cases where chamber or substrate contaminants provide the source for seeds, high absorption coefficients may explain the increased susceptibility of nodules for laser damage. For cases where the seeds have the same composition as the coating itself, however, other mechanisms must be acting.

It has been suggested that the domed nodules may act as microlenses focusing the laser light and therefore causing local damage [12]. For the close ratio of the nodule diameter to the laser wavelength it is not clear that simple diffractive optics apply. Also, the microlens approach does not take into account the standing-wave-electric-fields (SWEF) in the multilayer stack and the influence of the nodule structure on those fields.

Locally enhanced SWEF likely play a key role in the localized nature of optical coating damage. DeFord and Kozlowski [13] have modeled the electric fields at 3-D nodular defects in multilayer coatings. They found that the E-field is enhanced by up to a factor of four near the nodule seed. The enhancement is a function of the seed size, depth and electronic properties. Enhancement was, in general, highest for large, shallow seeds.

Once the light is coupled into the defect, the mechanical stability of the defect likely plays an important role as well. As shown in Fig. 1, the boundaries of nodule defects can be very discontinuous and voids are often present. In some cases the voids and local stresses cause the nodules to be ejected after the coating is removed from the deposition chamber or, in some cases, even during the coating process. Preliminary data indicate that defects formed by large seeds produce mechanically unstable nodules that show increased damage susceptibility [3,11].

4. METHODS FOR NODULE MINIMIZATION

4.1 Source generation

An ultimate goal in the study high damage threshold coatings is to develop methods to eliminate nodular defects all together. As mentioned above, many sources of seeds have been eliminated using proper vacuum and substrate handling techniques. Elimination of seed materials originating during deposition processes remains a challenge however. There are three main approaches to particulate control to consider:

1. Optimization of deposition parameters: The main objective is to reduce the frequency and severity of thermal, electrostatic, or mechanical instabilities that cause seed particles to be ejected from the source material. Some key deposition parameters are e-gun voltage, e-gun emission current, and beam pattern. A low current (i.e. low deposition rate) allows the source to operate at relatively lower temperatures where effects due to charging, stress relief, and phase transitions are reduced. Setting the e-gun at lower accelerating voltages keeps the e-beam heating nearer to the surface of the melt, thereby minimizing thermal gradients [14]. Also, a broad and rapid beam sweep pattern decreases the power (voltage x current) density into the source. Each deposition chamber and material combination is unique, however, and must be studied individually.

2. Choice of source material form: Source material geometry and density can have a substantial impact on source stability. In general, the density of the source material should be as close to bulk density as possible in order to minimize explosions resulting from the heating of trapped gasses. When evaporating hafnia, defect formation can be minimized through the use of pellet material rather than large boules [6]. The pellets are believed to minimize the effects of thermal cracking.

3. Filtering of ejected particles: If the source can not be kept from ejecting particles, it may be possible to stop the particles before they hit the substrate. The easiest method is the use of gravity. Long throw distances are therefore attractive, but can become impractical because higher evaporation rates are needed. Other types of particulate filters, shown schematically in Fig. 4, include:

- Electrostatic defection. This technique, using biased electrodes, has been shown to deflect charged particles produced during silicon deposition [15,16]. In cases where ejected particles are not charged, preferential charging can occur in the plume. The particulate, having a larger geometric cross section than the molecular effluent, has a higher probability of colliding with an electron from the beam.

- Rotating vanes. Vanes rotating above the evaporant plume collect the slower moving particulates while the faster moving molecular species traverses the length of the vane without collision. The vane principle has shown limited success in laser ablation processes [17].
- Baffles. Baffles are used to force the evaporant plume through a circuitous path between the source and an opening exposed to the substrates. The baffles are heated such that the evaporant does not condense onto this filter. Particulates, prevented from having a line-of-sight path to the substrate, collide with the baffles and either stick there or deflect back into the source. Baffling has proven successful with low melting point materials such as silicon monoxide.
- Gas jet momentum. A gas jet directs numerous, high velocity molecules into the base of the evaporant plume. As in the electrostatic case, the probability of deflecting particulates is greater than for molecular species because the collision cross-section of the particulates is greater.

For hafnia deposition, the success of filtering techniques on minimizing defect formation during has been limited. In this case the oxide particles are difficult to charge electrostatically [18] and the material has too high of a boiling point (m.p. = 2700°C) for the use of baffles. Rotating vanes would require rotation speeds that are impractical in a deposition chamber.

4.2 Nodule stabilization

If it is not possible to prevent the particles from hitting the substrate, it may be possible to stabilize the defects so that they are not highly susceptible to laser damage. Stabilization of defects appears to be the role of the laser conditioning process. This process of subthreshold illumination often increases the damage threshold of e-beam deposited HfO_2/SiO_2 mirrors and polarizers by factors of 2 to 3. Small area damage tests have shown that laser conditioning increases the damage threshold of the nodules but does not significantly influence the surrounding material [2]. AFM measurements indicate that some smoothing of nm-scale features of the coating surface occurs as a result of laser conditioning. This smoothing may be an indication of fusing of the nodule into the coating, therefore increasing its mechanical stability.

Some stabilization of nodules might also be obtained through the use of energetic deposition techniques such as ion-assisted deposition or ion-beam sputtering. The higher mobility of material deposited by these techniques produces coatings with bulk-like density. The increased mobility might also result in more uniform nodule/coating boundary regions and fewer voids. Unfortunately, such techniques may also have additional sources of defects because of the added complexity of the techniques. In general, coatings deposited using such techniques do not show increased damage thresholds over that obtained from the e-beam technique. This may be because other contributions to laser damage, such as bulk absorption, may play a role. Cross-sectioning studies and small-area laser damage experiments must be done on these films in order to test the defect-stabilization effects of these energetic processes.

5. AUSPICES

This work was performed under the auspices of the U. S. Dept. of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

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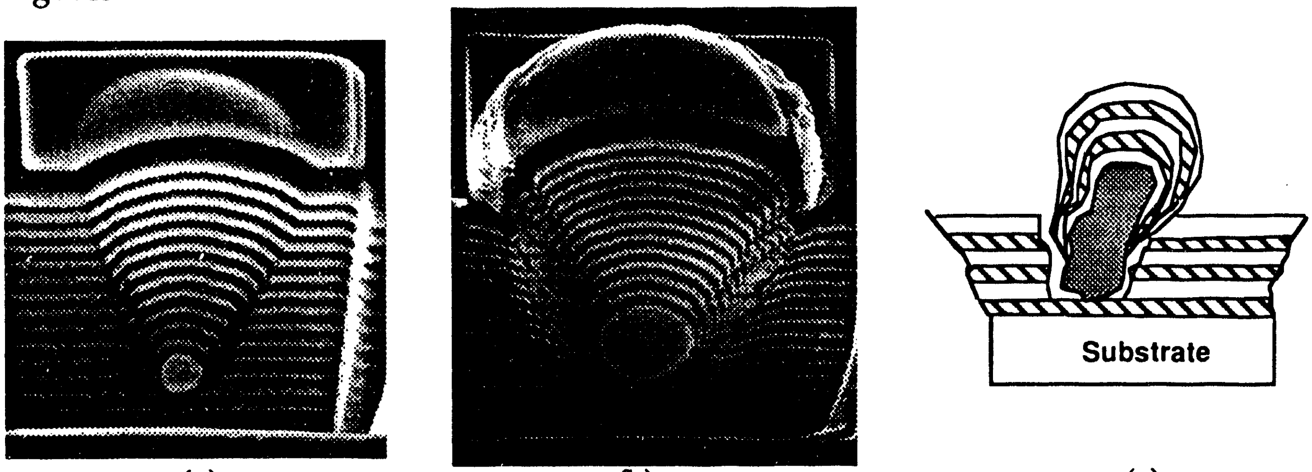
Figures



(a) (b) (c)

Figure 1: Cross-sections of nodular defects suggesting a relationship between seed size, nodule shape and nodule mechanical stability. (a) and (b): focused ion-beam cross-sections of a HfO_2/SiO_2 mirror coating [3]; (c) schematic of a "spatter" defect [4].

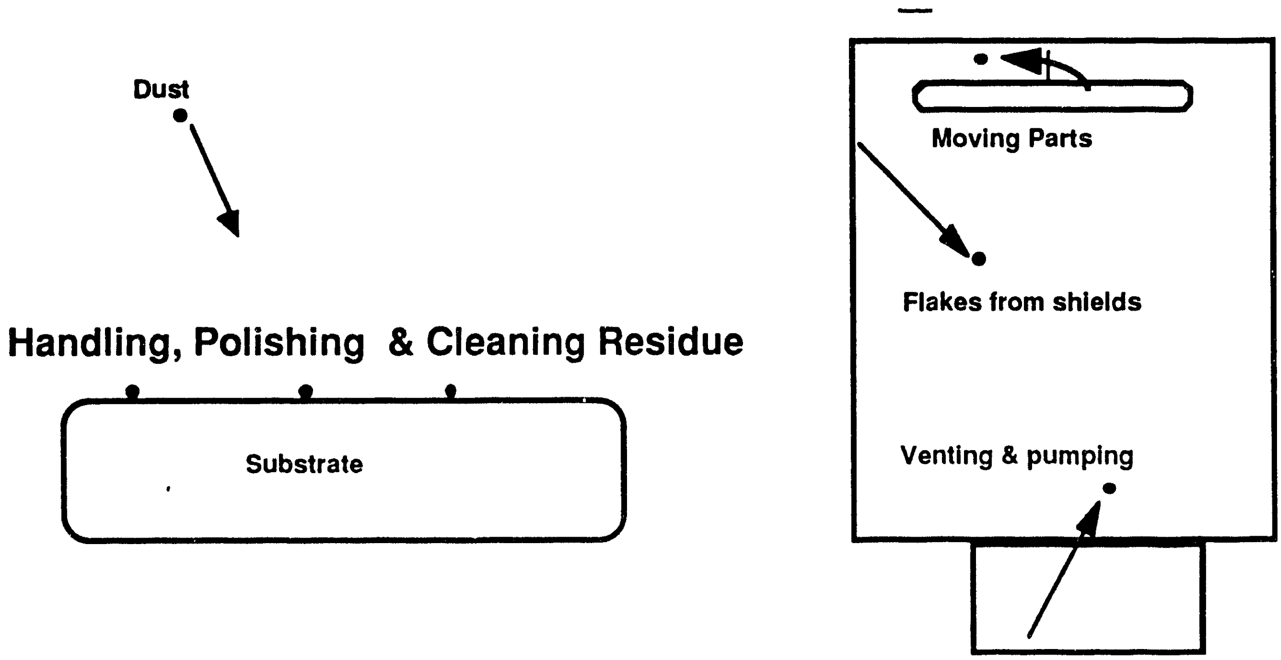


Figure 2: Diagram showing possible sources of particulates that may act as nodule seeds during coating deposition.

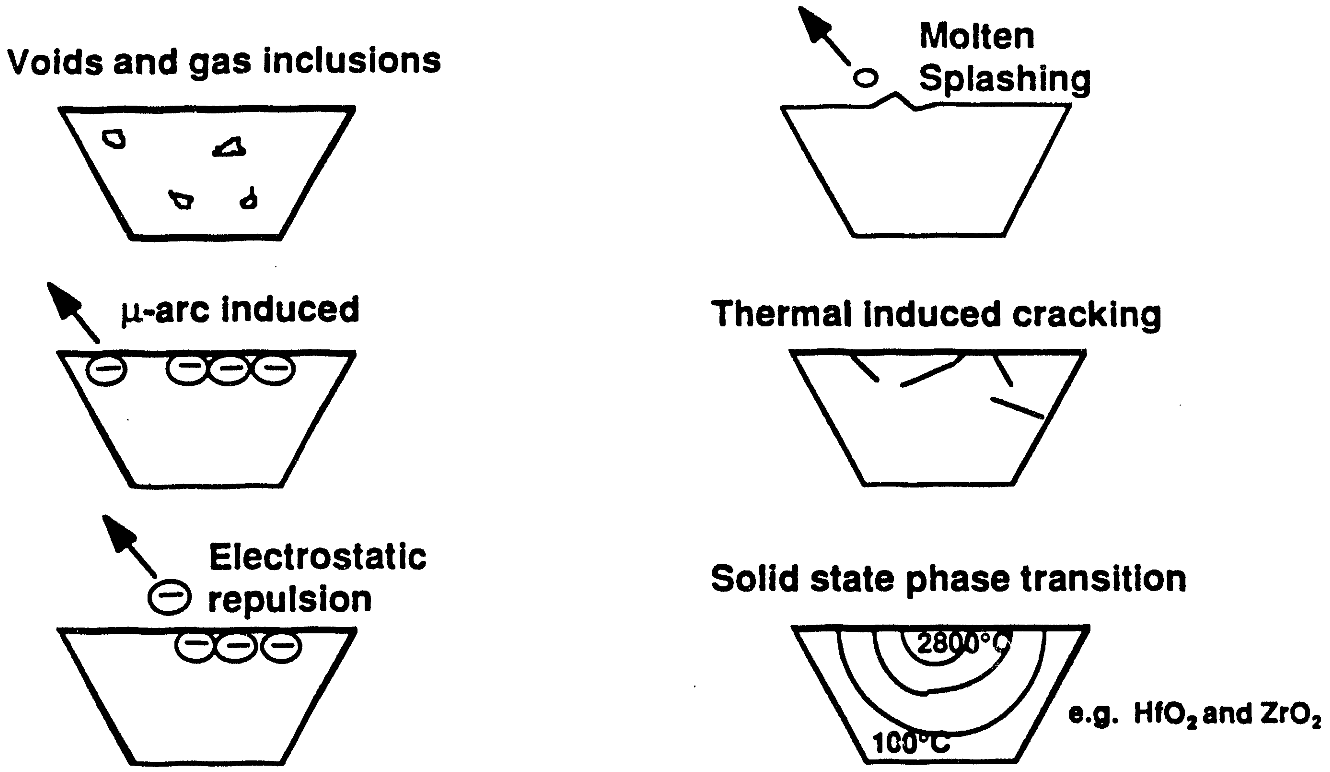
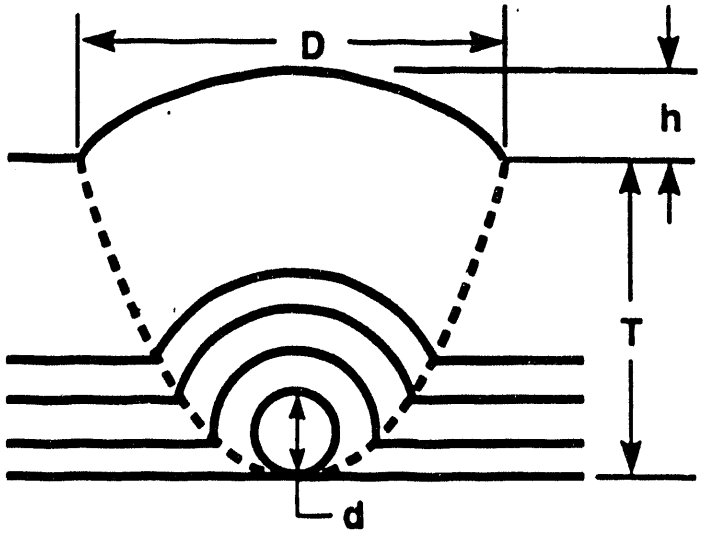


Figure 3: Schematic representation of possible mechanisms for particulate generation from the evaporation source material. [5,6]



$$h = d$$

$$D = (8Td)^{1/2}$$

Figure 4: Simple model for nodular defect geometry assuming omnidirectional flux.

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