Mechanisms Governing Laser-Induced Damage in Absorbing Glasses Under Exposure to Nanosecond Pulses

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Significant progress has been made during the past 15 years in understanding the mechanisms of laser-induced damage in transparent optical materials and mirrors. Above a threshold laser fluence, defect structures initiate plasma formation that leads to exposure of the material to high localized pressures followed by an explosive boiling process that involves ejection of superheated material, the launching of a shock wave, and stresses that result in mechanical damage of the surrounding "cold" material.[1] The dynamics and ensuing relaxation pathway are typically manifested as a microscopic crater on the surface or a microscopic void formed in the bulk of the material, depending on the location of damage initiation.

In contrast, laser-induced damage in absorbing dielectric materials under nanosecond laser pulses has received far less attention. Such absorbing materials are typically used as optical filters to attenuate the laser light for various applications including laser safety, sensor protection, and attenuation of stray beams in high-power laser systems such as at the National Ignition Facility. In addition, nominally transparent optical materials (such as glasses) operating in a high-radiation environment can develop color centers, thereby becoming absorbing at the operational wavelength.

Early work suggested that nonlinear absorption plays an important part in laser damage to absorbing optical glasses[2] and that damage occurred at either the surface or internally.[3] Past damage-morphology studies of such glasses under relevant excitation conditions indicated that material modifications are manifested with a typical "melted-surface" morphology[4] but such observations were often observed at fluences where the calculated surface temperature reached during the laser pulse was well below the melting point of the glass. Other reports suggested that the damage morphology changes as a function of the laser beam size and repetition rate.[5] These previous results indicate that key issues regarding laser-induced damage in absorbing glasses remain unclear.

The current study was designed to investigate the dominant mechanisms of laser-induced damage in absorbing optical materials under irradiation with 355-nm pulses having a temporal duration of the order of a nanosecond. A wide array of diagnostics tools was employed to enable one to monitor beam propagation inside the material, quantifying changes in the optical properties of the material, and capturing time-resolved transient material modifications with adequate spatial and temporal resolution. The results suggest that, in addition to linear absorption, excited-state absorption is a key mechanism contributing to enhanced energy deposition. In addition, there are two competing damage-initiation mechanisms: the first through self-focusing activated by a transient, fluence-dependent change of the refractive index; the second through melting of the material as a result of the increase of its temperature.

Figure 1(a) demonstrates a typical example of the first observed damage mechanism, showing the beam intensity profiles along the center of the beam at the exit surface of a 4-mm-thick, Ce^{3+}-doped silica glass sample for four different input laser fluences (0.8 J/cm^2, 6.4 J/cm^2, 8.3 J/cm^2, and 11 J/cm^2, respectively). At a lower peak fluence (<2 J/cm^2), the beam profile remains mostly unchanged. A significant narrowing of the beam is observed, however, at the sample's exit surface at higher fluences. These results clearly demonstrate a self-focusing behavior of the propagating beam into the material that strongly depends on the laser fluence.

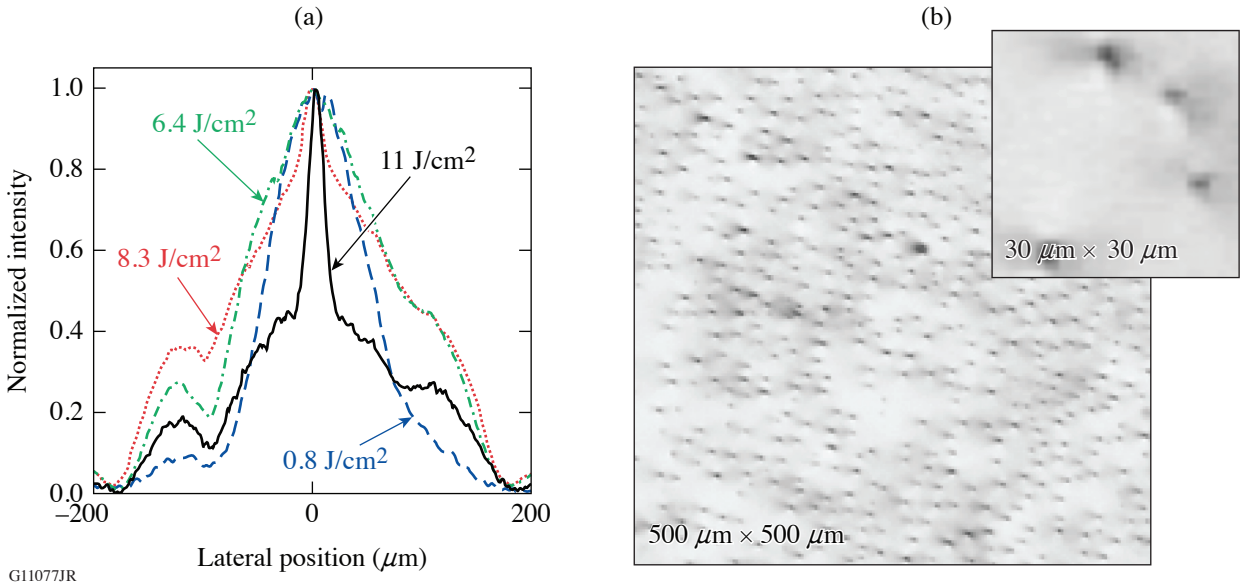

Figure 1
(a) The intensity profile along the middle of the laser beam at the output surface of a 4-mm-thick, Ce^{3+}-doped silica glass sample for different laser (peak) fluences. (b) Images of filamentation-induced damage in Ce^{3+}-doped silica 1.25 mm below the input surface under exposure to ≈20-J/cm^2, 351-nm, 5-ns flat-in-time pulses.

The same material was exposed to single pulses having a beam diameter of the order of 1 cm at various average laser fluences. The entire volume of the sample exposed to the laser beam was subsequently imaged at different depths, starting from the input surface and into the bulk in increments of 250 μm. Figure 1(b) represents a typical image at a depth of 1.25 mm below the surface from a section of the sample that was exposed to a fluence of ≈21 J/cm^2. This image demonstrates the presence of numerous filament damage sites with a diameter of ~2 to 3 μm. This is better demonstrated in the high-magnification inset. The filaments at this fluence are observed to start from a depth of ~500 μm and extend to a depth of ~1750 μm. The highest density of filament damage sites is observed ~1 mm below the surface for all fluences used in these experiments. An examination of individual filaments reveals that their length is ~750 μm.

A detailed investigation of the dynamics of this self-focusing behavior reveals that it arguably originates from a change of the index of refraction following the transition of electrons to a higher excited state. This modulation of the refractive index arises from the difference in the polarizability between the ground and excited states of the impurity ions and even transient defects formed during the excitation process. The relaxation time of this change of the refractive index is the same as the lifetime of the excited state; therefore, it can be much longer than the laser pulse (of the order of 100 ns for Ce^{3+}-doped silica glass). For large beams, the modulation of the refractive index leads to beam breakup and the formation of filaments as shown in Fig. 1(b). The filaments are located over a narrow depth zone (~2 mm) since the self-focusing mechanism is counteracted by the attenuation of the laser beam as it propagates inside the material.

The second damage-initiation mechanism in absorbing glasses is associated with heating of the material near the surface, resulting from the nonradiative relaxation following laser-energy deposition. This can support above-melting temperatures, which introduces nonreversible material modifications (damage). The dominant damage-initiating mechanism for a specific material and irradiation conditions depends on the material's electronic and thermophysical properties.

To understand this dual behavior of laser damage in absorbing glasses, we need to consider that there is a laser-induced damage threshold (LIDT) associated with the material surface reaching above melting temperature ($LIDT_{melt}$), as well as an analogous laser fluence for damage initiation via self-focusing ($LIDT_{focus}$). The experimentally observed damage behavior of a specific material and excitation condition is governed by the mechanism with the lowest threshold. The change in the damage mechanism

with beam size can arise from the thermomechanical properties of the material and the fact that the peak surface temperature can be a function of the laser spot size. For small beams, the flow of energy (such as via heat diffusion and/or electron transport) reduces the peak temperature, thereby increasing the effective $LIDT_{melt}$ value. On the other hand, for large beams, there is no (or not significant) energy flow, which leads to the generation of higher local temperatures for the same laser fluence. Consequently, a larger beam size promotes a higher localized temperature in the material for the same laser fluence. In materials with similar $LIDT_{melt}$ and $LIDT_{focus}$, experimentation with different laser beam spot sizes can lead to different observables because of the change in the governing damage-initiation mechanisms. For similar reasons, the local temperature can be a function of the pulse repetition rate.

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