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Laser-induced Damage in Dielectrics with Nanosecond to Subpicosecond Pulses
I. Experimental

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ABSTRACT

We report extensive laser-induced damage threshold measurements on pure and multilayer dielectrics at 1053 and 526 nm for pulse durations, τ, ranging from 140 fs to 1 ns. Qualitative differences in the morphology of damage and a departure from the diffusion-dominated $\tau^{1/2}$ scaling indicate that damage results from plasma formation and ablation for $\tau \leq 10$ ps and from conventional melting and boiling for $\tau > 50$ ps. A theoretical model based on electron production via multiphoton ionization, Joule heating, and collisional (avalanche) ionization is in good agreement with both the pulsewidth and wavelength scaling of experimental results.

Keywords: short-pulse, laser-induced damage, dielectric breakdown, multiphoton ionization

1. INTRODUCTION

Pulsewidth-dependent scaling and morphology of laser-induced damage have been the subject of numerous studies[1-28]. For pulses longer than a few tens of picoseconds, the generally accepted picture of bulk damage to defect-free dielectrics involves the heating of conduction band electrons by the incident radiation and transfer of this energy to the lattice. Damage occurs via conventional heat deposition resulting in melting and boiling of the dielectric material. Because the controlling rate is that of thermal conduction through the lattice, this model predicts[6] a $\tau^{1/2}$ dependence of the threshold fluence upon pulse duration τ. This is in reasonably good agreement with numerous experiments[8-28] which have observed a τ^{α} scaling with nominally $0.3 < \alpha < 0.6$ in a variety of dielectric materials (including samples with defects) from 20 ps to over 100 ns. Due to the difficulty in changing pulse duration, most of these scaling measurements were done at relatively few pulselengths.

Recently, the application of chirped-pulse amplification[29,30] (CPA) to solid-state lasers has enabled terawatt class systems producing subpicosecond pulses. Further increase in the peak power available from such systems[31] is now limited by damage to optical surfaces due to the intense short pulses. This duration is significantly shorter than the time scale for electron energy transfer to the lattice. As a result, damage caused by subpicosecond pulses is characterized by ablation, with essentially no collateral damage. Many applications, ranging from materials processing to biomedical technologies, could potentially benefit from the more localized energy deposition of short-pulse lasers as compared to long-pulse lasers.

Here, we report extensive measurements of laser-induced damage thresholds for fused silica and calcium fluoride for pulses ranging from 140 fs to 1 ns. In each of these large-bandgap materials we observe a change in the damage mechanism and morphology for pulses shorter than

20 ps. Although we observe a strong deviation from the $\tau^{1/2}$ scaling, we find no evidence for an increase in damage threshold with decreasing pulsewidth as reported by Du *et al* [27]. Instead, we observe a decreasing threshold associated with a gradual transition from the long-pulse, thermally-dominated regime to an ablative regime dominated by collisional and multiphoton ionization, and plasma formation. A general theoretical model[28] of laser interaction with dielectrics, based on multiphoton ionization, Joule heating, and collisional (avalanche) ionization, is shown to be in good agreement with the data in this short-pulse regime.

2. EXPERIMENTAL

For damage testing, we used laser pulses generated by a 1053-nm Ti:sapphire CPA system[32] as diagrammed in Figure 1. Seed pulses of 100 fs from a Kerr-lens mode-locked, Ti:sapphire oscillator were stretched to 1 ns in a four-pass, single-grating (1740 line/mm) pulse stretcher. Amplification by nearly 10^9 to the 6-mJ range was achieved in the TEM_{00} stable cavity mode of a linear regenerative amplifier. The low gain (≈1.2/pass) of Ti:sapphire at 1053 nm necessitated over 120 passes to produce the desired gain. This required a short cavity (1.2-m) to minimize the decay of upper state population due to spontaneous emission, amplified spontaneous emission near 800 nm, and temperature effects, which combine to give an effective lifetime of approximately 2 µs. The short linear cavity and corresponding small mode size (1.4-mm diameter) limited the energy extraction from this stage due to increasing B-integral and self-phase modulation of the stretched, chirped pulse[32,33]. Further amplification to the 60-mJ level was achieved in a Ti:sapphire ring regenerative amplifier which, due to its 3-m length, supported a larger TEM_{00} mode (2.3-mm diameter) and reduced nonlinear effects. This system operated at 10 Hz.

After amplification, we compressed the pulses in a four-pass, single-grating (1740 line/mm) compressor of variable length (Figure 2). By varying the dispersive path length of the compressor, we obtained pulses of continuously adjustable duration from 0.4 to 1 ns. Pulse durations were measured with a single-shot autocorrelator[34] (0.4-1.5 ps), streak camera (10-1000

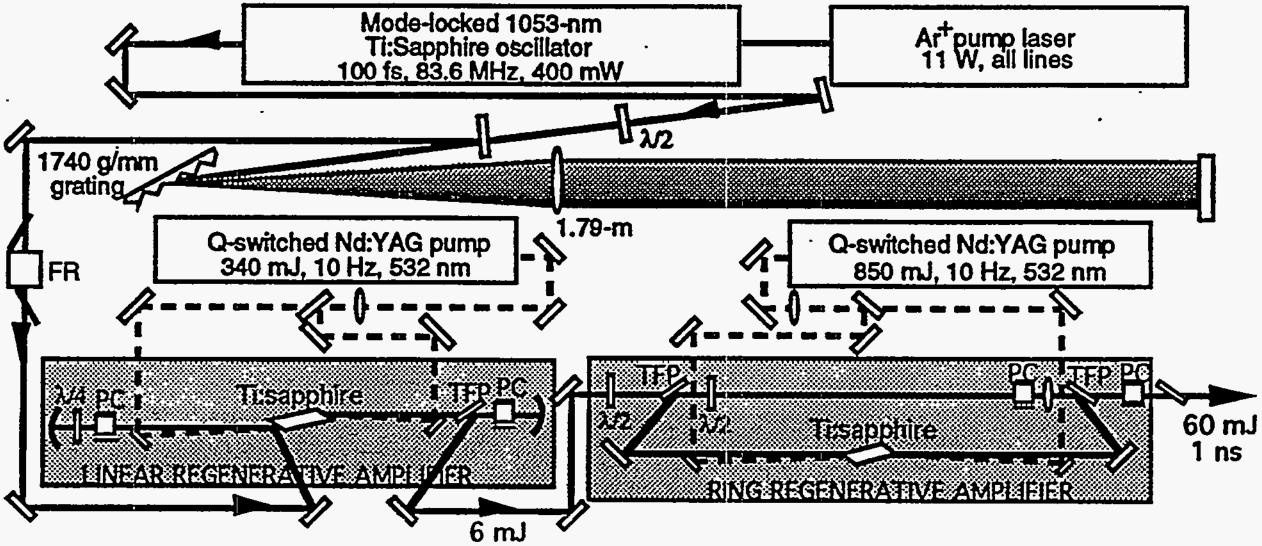

Figure 1. Schematic diagram of 1053-nm Ti:sapphire laser system which produces 60-mJ, 1-ns stretched pulses at 10 Hz with better than 3% rms stability. PC-Pockels cell, TFP-thin film polarizer, FR-Faraday rotator.

ps), and fast photodiode (100-1000 ps), and calibrated against the linear position of the fold mirrors. The temporal profile of the compressed pulses depends strongly on the spectral and temporal profile of the stretched pulse. Pulse compression with spectral clipping is analogous to diffraction from a hard-edge aperture in the spatial domain, and results in a modulated temporal profile in the "intermediate" range of compression. For these damage measurements, we compressed a near-Gaussian spectral profile to obtain temporally smooth output pulses. This allowed us to easily relate the time evolution of the pulse intensity to the measured fluence.

We also measured damage thresholds with 526-nm light generated by frequency-doubling the 1053-nm compressed pulses in a thin (4-mm) potassium dideuterium phosphate (KD*P) crystal. The conversion efficiency was kept below 25% to avoid any temporal distortion of the second-harmonic pulse. We measured our shortest 526-nm pulses with a single-shot autocorrelator to be 275 fs. This was in good agreement with the expected $2^{1/2}$ scaling from the 1053-nm pulsewidth, so this scaling was used for the other 526-nm pulsewidths. A single data point at 825 nm and 140 fs was taken with a Cr:LISAF CPA system[35] to confirm the decreasing trend in damage fluence with pulsewidth in fused silica.

The energy of each pulse was monitored with the leakage through a 92% reflectivity mirror. We adjusted the energy delivered to the damage sample with a half-waveplate before compression, using the strong dependence of grating efficiency upon input polarization. The rms energy stability was typically less than 3%, and we report the average value here. Due to saturated amplification in the regenerative amplifiers, the maximum energy never exceeded the average by more than 6%. We estimate the absolute uncertainty in our determination of the energy to be 8%.

We performed damage measurements with laser spot sizes adjustable from 0.3 to 1.0-mm diameter (e^{-2} intensity). The typical diameter used was 0.5 mm, with 0.3 mm used to reach the

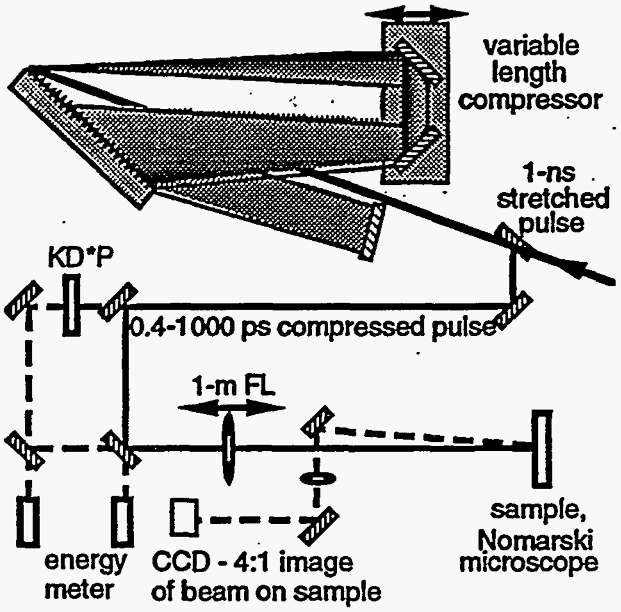

Figure 2. The 1-ns stretched pulses are compressed to 0.4-1000 ps by a variable length compressor. A 1-m lens focuses the pulse on the sample and the spot size is measured with a 4:1 image.

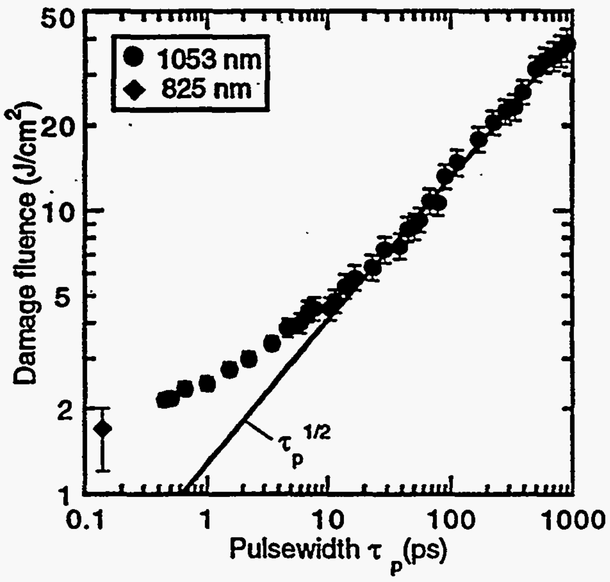

Figure 3. Pulsewidth dependence of threshold damage fluence for fused silica

higher threshold fluences required by the longer pulselengths. Laser pulses were focused onto the damage sample by a 1-m focal length lens, with a variable distance to the sample. The spot size was measured on a CCD camera. With the shortest pulses we used, the intensity (up to 4×10^{12} W/cm^2 on sample) became high enough to cause significant (10% effect) whole-beam self-focusing in the focusing lens and the air path leading to the sample. All beam size measurements were therefore performed with a 4:1 image of the beam taken from a 4% reflection at the position of the damage sample, and at or just below damage threshold. The laser mode at the sample had a 98% or better fit to a Gaussian, so the effective diameter as measured on the camera system was combined with the measured energy to give the pulse energy fluence. Our estimated absolute uncertainty in fluence was 15%, but relative values should be within 5%.

After irradiation, Nomarski microscopy was used to inspect the sample for possible damage. We define damage to be any visible permanent modification to the surface observable with the Nomarski microscope. The smallest damage spots we could observe were approximately 0.5 μm in diameter, a factor of 10^6 smaller in area than the laser spot size and nearly impossible to observe by other methods (e.g., degradation of transmission, scattered light, etc.). To avoid the complications of spatial and temporal distortion caused by self-focusing, group velocity dispersion, and self-phase modulation when propagating laser pulses through optical materials, we considered only front-surface damage. Depending on the focusing geometry and pulse duration, the rear surface or bulk of the transparent samples would often damage before the front surface, so we were careful not to let this damage propagate to the front surface.

Initial damage, at threshold, may have many forms: ablation of a very small amount of material (a few atomic layers); formation of a color center, shallow traps, or lattice defects; or melting of a very small volume. These weak effects are very difficult to detect. In order to "amplify" this damage to an easily observable size, and to minimize statistical uncertainty, we conducted our damage testing with multiple pulses of a given fluence on each site. This is in contrast to the single-shot measurements of Du *et al*[27], where their diagnostics of pulse transmission and plasma emission require a macroscopic damage site on a single pulse. This occurs with long-pulse breakdown, but the localized nature of short-pulse damage near threshold will lead to reduced sensitivity for single-shot measurements. We typically used six hundred shots at 10 Hz, unless damage was obvious sooner. Many fluence levels (15-30) were examined above and below the damage threshold for a given pulsewidth in order to establish the threshold value.

An important practical issue is the presence of surface cracks, nodules or voids, since such features are known to increase the local field intensity by a factor of η^4, where η is the refractive index[2,36]. This increase takes place over a region comparable to the feature scale length, and is offset by convection of electrons away from the region. A simple estimate of convection shows that for picosecond pulses the local enhancement is insignificant for feature scale lengths less than 40 nm.

3. DAMAGE RESULTS

3.1 Fused silica

The results presented here for fused silica were obtained with 1-cm thick "super-polished" samples (Corning 7940) exhibiting less than 1-nm rms surface roughness. We measured the same damage thresholds with a 200-μm thick fused-silica etalon, which was tested to examine any possible differences between thick and thin samples. Some samples were cleaned initially with acetone or methanol, and all were cleaned when damage debris accumulated on the surface. No difference in threshold was found between samples or areas on a given sample that were or were not cleaned. Defects visible through the microscope were avoided. With short (0.4 ps) pulses, damage always occurred at the location corresponding to the peak of the Gaussian intensity profile, indicating that defect sites did not contribute to our measured thresholds. Ramping the fluence (R:1) with short pulses, which would ionize and ablate any low-lying states or surface contamination with lower threshold, gave the same threshold as our S:1 measurements. We thus believe that our measurements correspond to a uniform, defect-free surface and can be compared to calculations based on the intrinsic properties of fused silica.

Our measured threshold damage fluence for fused silica at 1053 nm as a function of laser pulse length (FWHM) and a single point at 825 nm are shown in Figure 3. In the long-pulse regime ($\tau > 20$ ps), the data fit well to a $\tau^{1/2}$ dependence (actual fit: $\tau^{0.504}$), characteristic of transfer of electron kinetic energy to the lattice and diffusion during the laser pulse. The damage

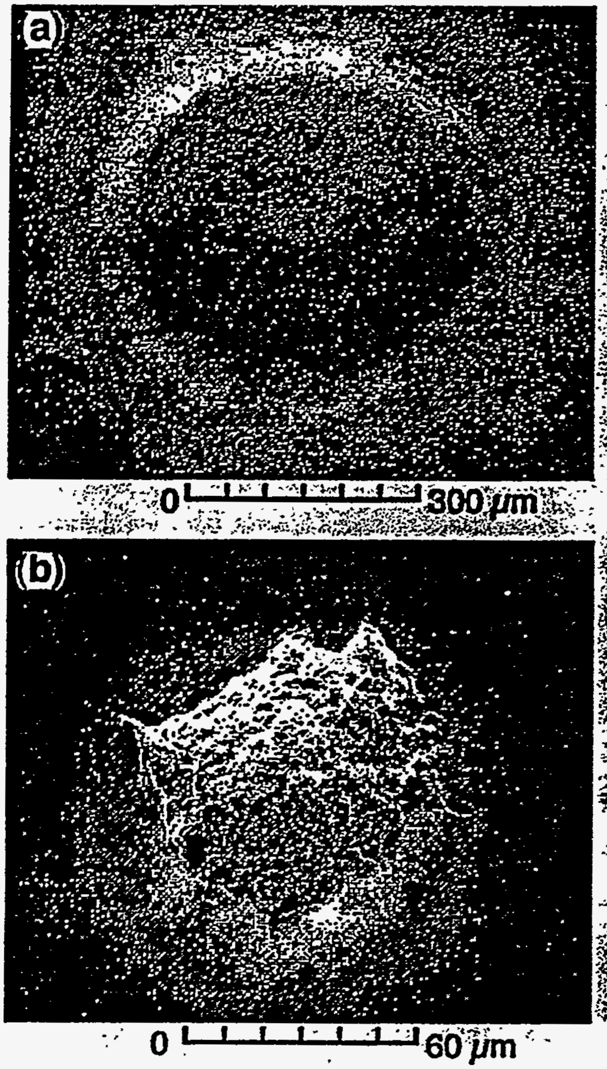

Figure 4. Laser damage spots on fused silica created by: (a) long-pulse, 900-ps, 300 μm diameter, (b) short-pulse, 0.4-ps, 500-μm diameter.

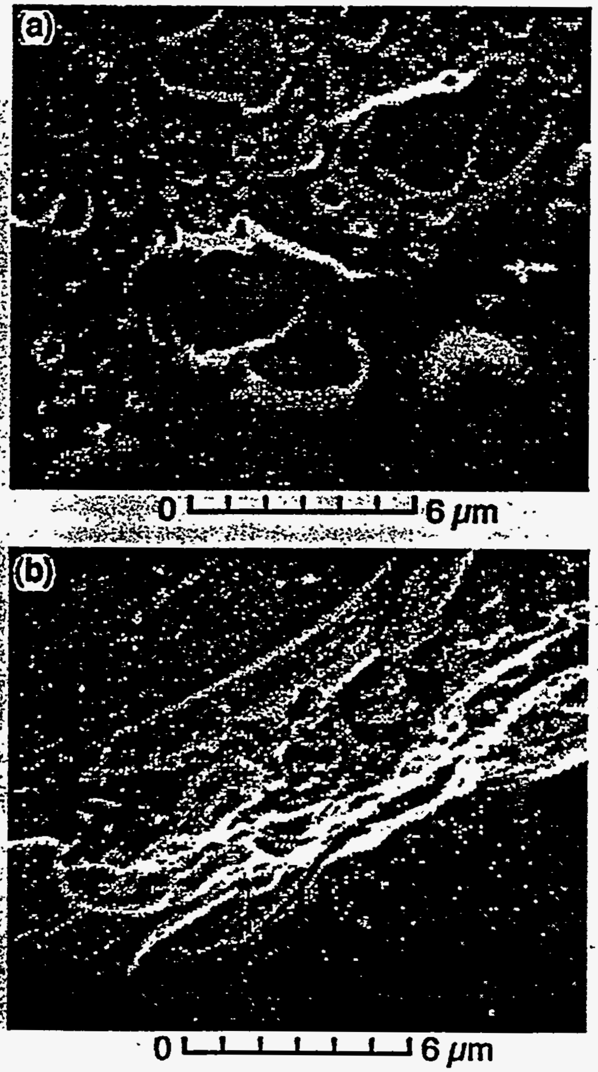

Figure 5. Edges of laser damage spots of Fig. 4: (a) long pulse, 900 ps, (b) short-pulse, 0.4 ps.

occurs over the entire area irradiated as shown in the electron micrograph of Figure 4(a) (All damage micrographs shown are the result of multiple pulses). The damage is thermal in nature and characterized by melting and boiling of the surface. This is more easily seen in Figure 5(a), which shows the edge of the long-pulse damage spot. For pulses shorter than 20 ps, the damage fluence no longer follows the $\tau^{1/2}$ dependence and exhibits a morphology dramatically different from that observed with long pulses. Short-pulse damage is confined to a small region at the peak of the Gaussian irradiance distribution (Figure 4(b)). Damage occurs only over an area with sufficient intensity to produce ionization. With insufficient time for lattice coupling, there is no collateral damage. As a result, the damaged area can be many orders of magnitude smaller with short ($\tau<10$ ps) pulses than with long pulses. For the case of fused silica shown in Figure 4, the damaged area produced by the 0.5-mm diameter, 500-fs pulse was two orders of magnitude smaller than that produced by the 0.3-mm diameter, 900-ps pulse. Short-pulse damage appears as a shallow fractured and pitted crater characteristic of a thin layer of material removed by ablation (Figure 5(b)). We found damage in the short-pulse limit to be deterministic, with only a couple percent fluence range between damage and no damage. Fused silica irradiated with 10000 shots at 2% below our determined threshold showed no evidence of damage with 0.4-ps pulses. For long pulses we found a roughly 10% range in fluence where damage would or would not occur.

In Figure 6, we concentrate on the short-pulse region and include our measured damage thresholds at 526 nm. The solid curves are the results of our theoretical modeling of laser-induced damage in the short-pulse limit, and are in very good agreement with both the *pulsewidth* and *wavelength* scaling of the measured data. This model is based on initial electron production by multiphoton ionization, Joule heating, and collisional (avalanche) ionization, and is described in detail elsewhere in these proceedings[28]. We chose the plasma critical density (i.e. strongly absorptive regime) as the theoretical indicator of macroscopic damage. The calculated threshold is not sensitive to this choice, decreasing by ≈20% for an electron density of 10^{19} cm^{-3} at which the

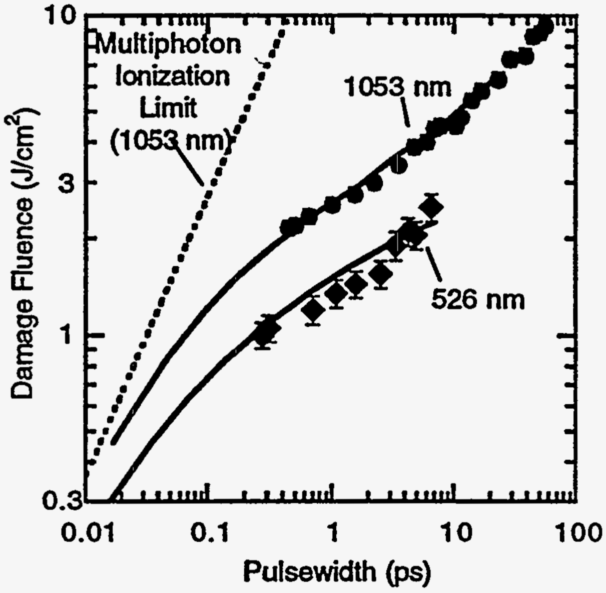

Figure 6. Measured and calculated (solid lines) damage fluence for fused silica at 1053 and 526 nm. Dashed line indicates calculated damage limit due to multiphoton ionization alone.

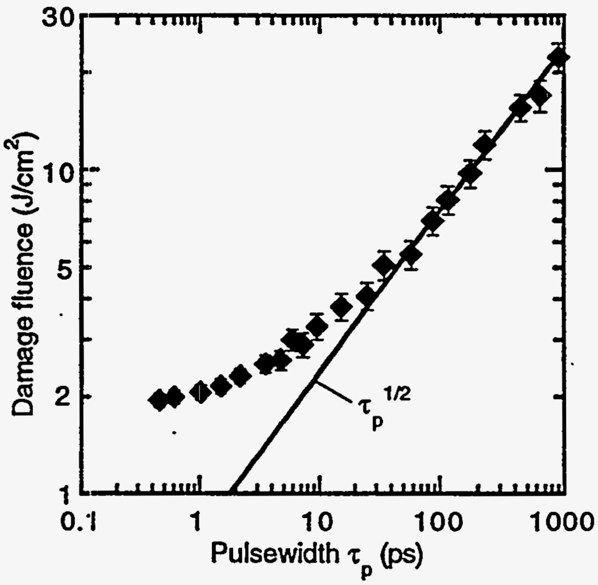

Figure 7. Pulsewidth dependence of threshold damage fluence for calcium fluoride.

energy density of conduction electrons equals the lattice binding energy. As shown, with decreasing pulsewidth the damage threshold will asymptote to the limit where multiphoton ionization alone creates sufficient electron density to cause damage.

3.2 Fluorides

The damage threshold of calcium fluoride exhibits a similar pulsewidth dependence to that of fused silica (Figure 7). In the long-pulse limit, the threshold fluence also scales as $\tau^{1/2}$, and then changes to the short-pulse limit near 20 ps. For long pulses the damage morphology is again consistent with melting. Figure 8(a) shows the melting and recrystalization of the calcium fluoride surface layers, which occurred with no evidence of an avalanche breakdown. This is consistent with the measurements of Jones *et al* [37] on wide-gap alkali halides. Short-pulse damage clearly initiates on scratches left from the polishing process (Figure 8(b)), although as observed by Milam[38] with 125 ps pulses, the damage threshold did not appear to be greatly influenced by the polishing sleeks. The short-pulse (0.4 ps) damage thresholds of BaF_2, CaF_2, MgF_2, and LiF

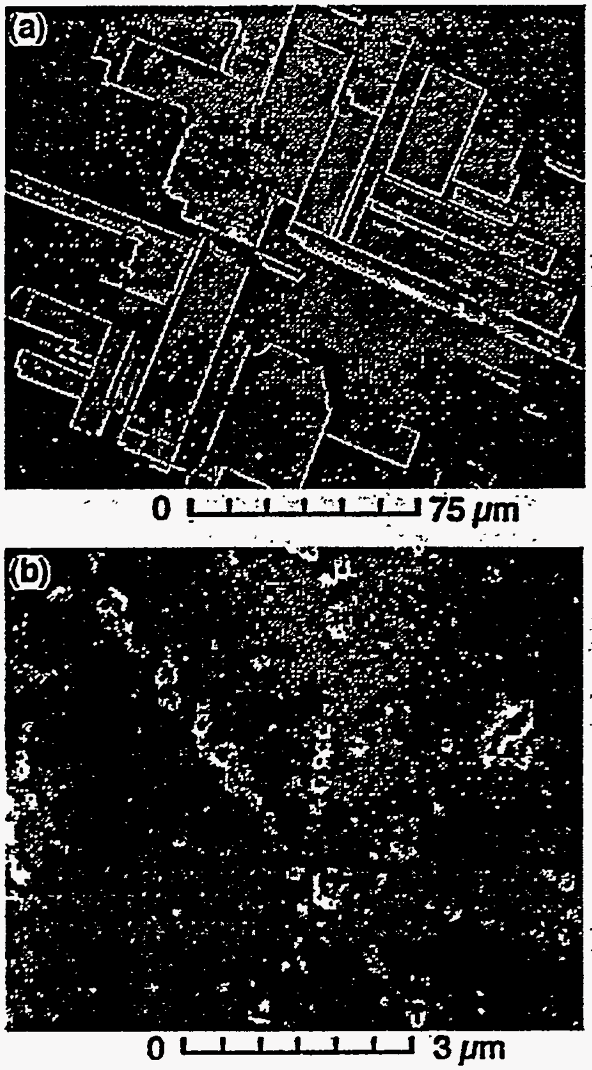

Figure 8. Laser damage morphology of calcium fluoride for (a) 900 ps, and (b) 0.4 ps pulses.

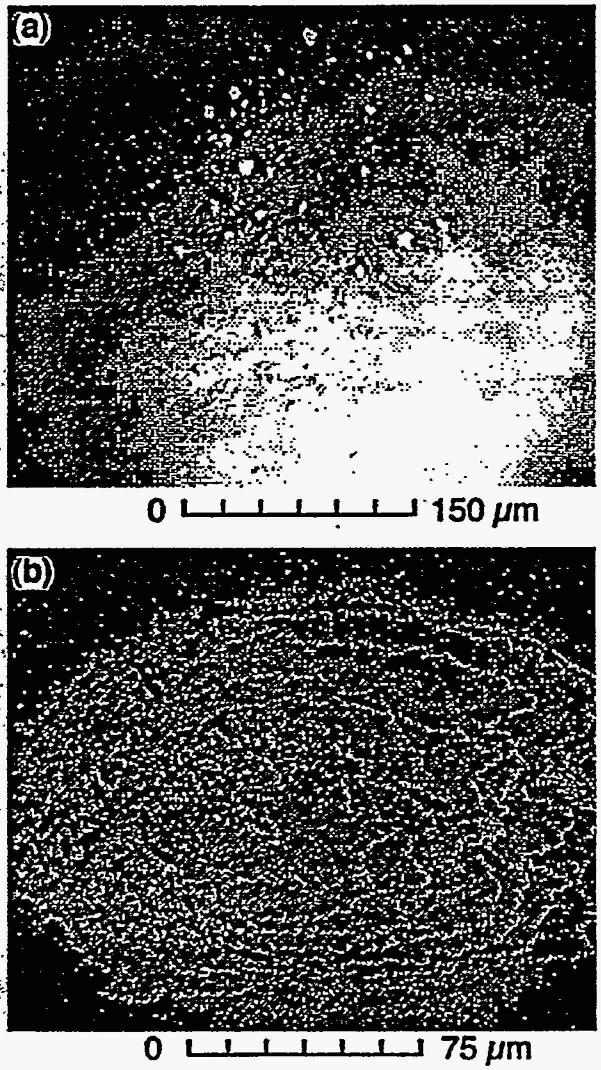

Figure 9. Laser damage morphology of a multilayer dielectric mirror for (a) 900 ps, and (b) 0.4 ps pulses.

(included in Figure 12) scale with bandgap energy, as expected from multiphoton initiated avalanche ionization.

3.3 Multilayer dielectrics

We have tested several different multilayer dielectric mirrors and polarizers. Figure 10 shows the pulsewidth dependence of the damage threshold fluence for a 45° 1053-nm high-reflector. The long-pulse scaling is slightly less than $\tau^{1/2}$, and again there is a deviation near 20 ps with a transition to the short-pulse regime where there is insufficient time for energy coupling to the lattice. The long-pulse damage morphology is characterized by melting and flow (Figure 9(a)), whereas short pulses cause ablation of the individual dielectric layers (Figure 9(b)). As has been thoroughly characterized by Kozlowski, *et al*[39], the initiation of damage with long (ns) pulses is dominated by nodules and defects. We find the same behavior with short pulses, where the presence of defects reduces the damage threshold by as much as 50%. We tried laser-conditioning[40,41] this sample to improve the short-pulse damage threshold. Unfortunately, neither long nor short-pulse conditioning resulted in a higher short-pulse damage threshold.

Figure 11 shows the pulsewidth-dependent damage threshold of the defects on a multilayer dielectric polarizer (1053-nm, 56°S). For these measurements, we placed the largest defects at the center of the Gaussian intensity profile. Damage initiated on and propagated from these sites. A τ^{α} fit through the entire curve gives $\alpha \approx 0.25$.

The short-pulse (0.4 ps) damage thresholds of many of the multilayer dielectrics we tested are included in Figure 12. The cross-hatched area indicates thresholds achieved on "clean" areas. The highest damage threshold we have found on any multilayer dielectric sample with 0.4-ps pulses at 1053 nm is 1.4 J/cm^2.

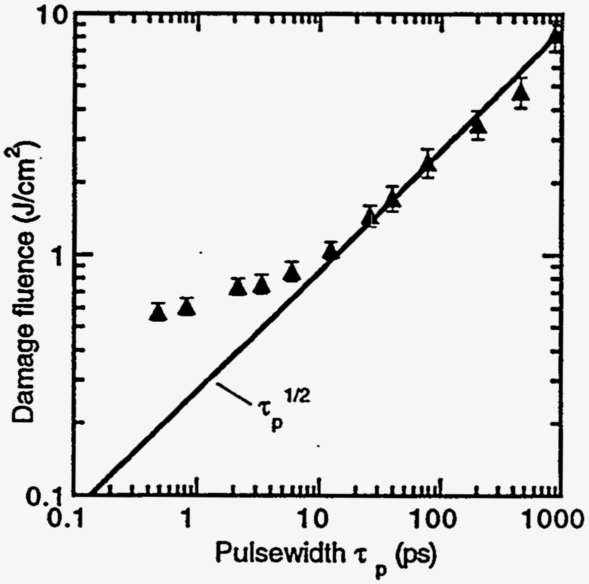

Figure 10. Pulsewidth dependence of threshold damage fluence for a multilayer dielectric mirror on relatively clean areas.

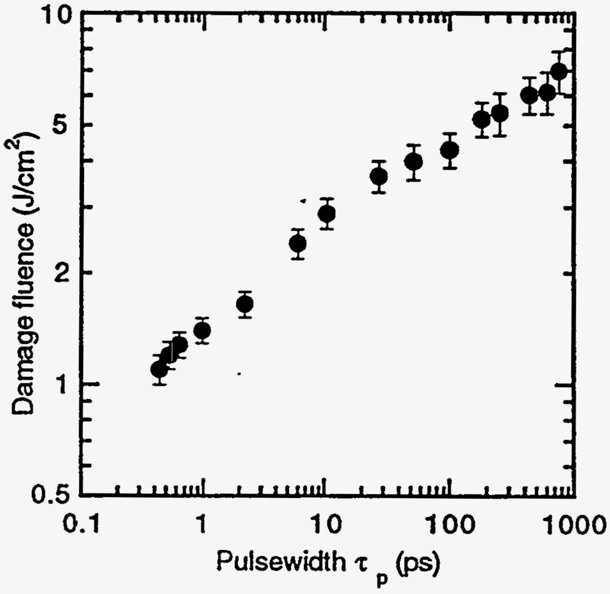

Figure 11. Pulsewidth dependence of threshold damage fluence for the defects on a multilayer dielectric polarizer.

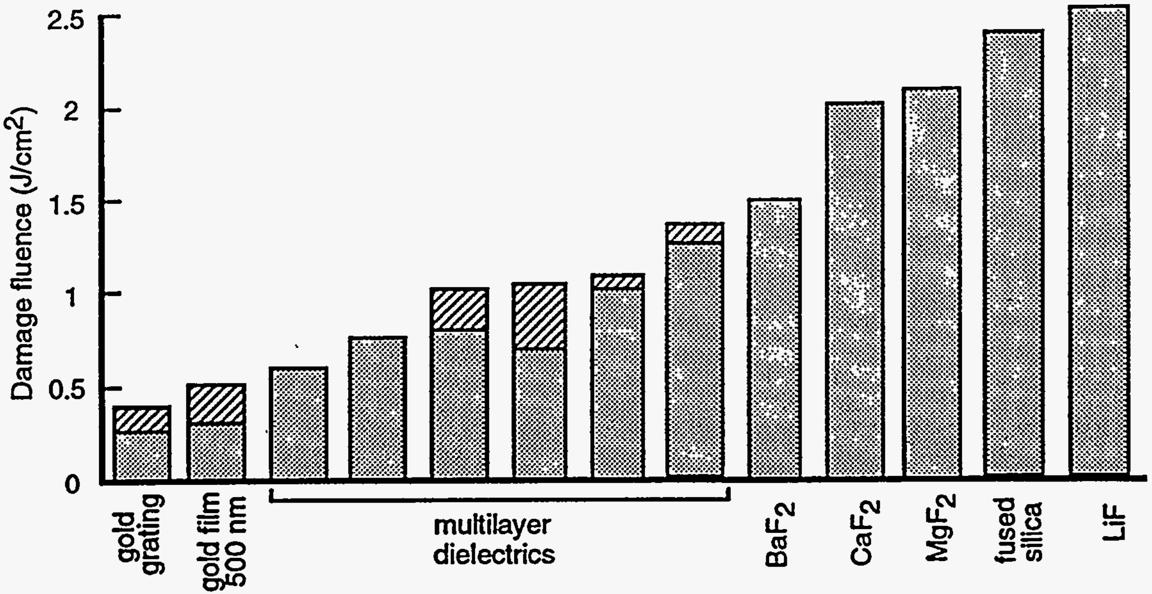

Figure 12. Short-pulse (0.4 ps), front-surface threshold damage fluences for gold film, gold grating, multilayer dielectrics (polarizers and mirrors), and pure dielectrics.

4. DISCUSSION

Using the continuously tunable pulsewidth of our 1053-nm Ti:sapphire CPA system, we have investigated the pulsewidth dependence of laser-induced damage in pure and multilayer dielectrics over the range 0.1-1000 ps. We observe a strong deviation from the long-pulse $\tau^{1/2}$ scaling of laser damage fluence for pulses shorter than 20 ps, below which electrons have insufficient time to couple to the lattice during the laser pulse. The damage threshold continues to decrease with decreasing pulsewidth, but at a rate slower than $\tau^{1/2}$ in the range 0.1 to 20 ps. This departure is accompanied by a qualitative change in the damage morphology indicative of rapid plasma formation and surface ablation. The damage site is limited to only a small region where the laser intensity is sufficient to produce a plasma with essentially no collateral damage. A theoretical model, in which initial electrons provided by multiphoton ionization are further heated resulting in collisional (avalanche) ionization, predicts short-pulse damage thresholds in excellent agreement with both the pulsewidth and wavelength scaling of our measurements. For extremely short pulses (τ<30 fs), multiphoton ionization alone will provide the critical density of electrons.

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