Optical ablation by high-power short-pulse lasers

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Laser-induced damage threshold measurements were performed on homogeneous and multilayer dielectrics and gold-coated optics at 1053 and 526 nm for pulse durations $\tau$ ranging from 140 fs to 1 ns. Gold coatings were found, both experimentally and theoretically, to be limited to 0.6 J/cm$^2$ in the subpicosecond range for 1053-nm pulses. In dielectrics, we find qualitative differences in the morphology of damage and a departure from the diffusion-dominated $\tau^{1/2}$ scaling that indicate that damage results from plasma formation and ablation for $\tau \leq 10$ ps and from conventional heating and melting for $\tau > 50$ ps. A theoretical model based on electron production by multiphoton ionization, joule heating, and collisional (avalanche) ionization is in quantitative agreement with both the pulse-width and the wavelength scaling of experimental results. © 1996 Optical Society of America

1. INTRODUCTION

The application of chirped-pulse amplification (CPA) to broadband, high-energy solid-state lasers has permitted the realization of terawatt-class systems producing subpicosecond pulses. Further increase in the peak power available from such systems is now limited by damage to optical surfaces caused by the intense short pulses. The pulse-width dependence of the breakdown threshold is of interest for all optical components in a high-power laser system: mirrors, polarizers, bulk dielectrics, coatings, and diffraction gratings. At this time the gratings used for pulse compression typically exhibit the lowest damage threshold for any component in a CPA system. These high-efficiency diffraction gratings have traditionally been based on metallic gratings. High-efficiency multilayer dielectric diffraction gratings offer a promising alternative that may exhibit a damage threshold many times that of gold gratings, depending on pulse width. In this paper we present measurements and theoretical analysis of laser-induced damage to gold-coated, multilayer dielectric, and pure dielectric optical components.

Investigation of the pulse-width dependence of laser-induced damage to dielectrics has been the subject of numerous studies over many years. For pulses longer than a few tens of picoseconds the generally accepted picture of 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 through conventional heat deposition (heating of the lattice), resulting in melting, boiling, or fracture of the dielectric material. Because the controlling rate is that of thermal conduction through the lattice, this model predicts a $\tau^{1/2}$ dependence of the threshold fluence on pulse duration. This is in reasonably good agreement with numerous experiments in which a $\tau^\alpha$ scaling with nominally $0.3 < \alpha < 0.6$ was observed in a variety of dielectric materials (including samples with defects) from 10 ps to more than 100 ns.

Here we report measurements of damage thresholds for fused silica, calcium fluoride, and multilayer dielectrics for pulses ranging from 140 fs to 1 ns. In each of these large-band-gap materials we observe a change in the mechanism and morphology of laser-induced damage for pulses shorter than 20 ps. A deviation of breakdown threshold from long-pulse scaling in dielectrics was reported by Soileau et al. for pulses in the range 4–10 ps. Du et al. extended the pulse-width range to 150 fs and found that the breakdown threshold fluence increases for pulse widths less than 10 ps. Although we also observe a deviation from the long-pulse scaling, we find no evidence for an increase in damage threshold with decreasing pulse width. Instead, we observe a smoothly 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. We develop a general theoretical model of laser interaction with dielectrics, based on multiphoton ionization, joule heating, and collisional (avalanche) ionization, which is shown to be in good agreement with our data in this short-pulse regime.

2. DAMAGE THRESHOLD MEASUREMENT

Damage testing was performed with laser pulses generated by a 1053-nm Ti:sapphire CPA system. The front end of this system produced 1-ns stretched pulses as much as 60 mJ at 10 Hz in a TEM$_{00}$ Gaussian mode. The pulses were then compressed in a single-grating compressor of variable length. By varying the dispersive path length of the compressor, we obtained pulses of continuously adjustable duration from 300 fs to 1 ns. Pulse durations were measured with a single-shot autocorrelator (0.3–1.5 ps), a streak camera (10–1000 ps), and a fast photodiode (100–1000 ps). The temporal profile in the compressed pulses depends strongly on the spectral and temporal profiles 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 therefore compressed a near-Gaussian spectral profile to ob-
tain temporally smooth Gaussian output pulses. This allowed us easily to relate the time evolution of the pulse intensity to the measured fluence.

Frequency-doubled pulses at 526 nm were generated from the 1053-nm compressed pulses in a thin (4-mm) potassium dideuterium phosphate crystal. The conversion efficiency to the second harmonic was kept below 25% to avoid temporal distortion. Pulse lengths were measured with a single-shot autocorrelator, with the shortest frequency-doubled pulses measuring 200 fs. Data were also taken at 825 nm and 140 fs with a Cr:LiSrAlF6 CPA system.25

We adjusted the energy delivered to the damage sample with a half-wave plate before compression, relying on the grating as the polarizer. The energy of each pulse was recorded from calibrated leakage through a mirror. The energy stability of the pulses from the ring regenerative amplifier was typically 1.5% rms. The laser pulses were focused onto the damage sample by a 1-m focal-length lens, with a variable distance to the sample. We adjusted the laser spot size on the sample from 0.3- to 1.0-mm diameter (e2 intensity). With the shortest pulses the intensity (up to 4 \times 10^{12} \text{W/cm}^2 on the 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 made at or just below damage threshold. A 4:1 image of the beam was taken from a 4% reflection at the position of the damage sample and measured on a CCD camera. The laser spatial mode at the sample had a 98% or better fit to a Gaussian, so we combined the effective diameter 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, we used Nomarski microscopy to inspect the sample for damage. Our definition of damage includes any visible permanent modification to the surface observable with the Nomarski microscope. The smallest damage spots that we could resolve were approximately 0.5 \mu m in diameter, a factor of 10^6 smaller in area than the laser spot size and nearly impossible to observe by methods requiring degradation of transmission or scattered light. We considered only front-surface damage to avoid the complications that self-focusing, group-velocity dispersion, and self-phase modulation contribute to the determination of the temporal and spatial structure of laser pulses. We also investigated samples with electron microscopy to determine the morphology of the damage at higher resolution.

Initial optical damage can have many sources: ablation of a very small amount of material (a few atomic layers); formation of a color center, shallow traps, or structural lattice defects; or melting or boiling of a very small volume. These weak effects can be difficult to detect on a single laser pulse. To amplify this damage to a more easily observable size, and to minimize statistical uncertainty, all our threshold damage determinations were made with multiple pulses of a given fluence on each site. This is in contrast to the single-shot threshold measurements of Du et al.,23 which required detection of plasma emission or a decrease in transmission caused by the single pulse. We typically used 600 shots at 10 Hz. We examined many fluence levels (15–30) above and below the damage threshold for a given pulse width to establish the threshold value.

3. DAMAGE TO GOLD-COATED OPTICS

Optical damage to metallic surfaces has been measured as a function of pulse width down to 1 ps. A \tau^{1/2} dependence of the damage threshold on pulse width \tau is observed down to 1 ns, below which the threshold becomes nearly independent of pulse width. We have measured the damage thresholds of gold films and diffraction gratings down to 300 fs and have investigated the threshold dependence on gold film thickness.

Scanning electron microscopy of the damage morphology of gold films reveals strong clues to the mechanism responsible for damage. Damage spots created by very short pulses (0.6 ps) exhibit negligible melting or collateral damage, indicative of extremely rapid ablation and vaporization [Fig. 1(a)]. In contrast, long pulses (900 ps) create damage regions whose appearance is consistent with melting, flow, and subsequent resolidification [Fig. 1(b)]. These observations form the basis of a simple model describing the thickness dependence of laser damage to gold-coated optics.

We model the metallic thin film or grating as a one-dimensional structure subjected to a constant heat flux at the surface and permit negligible heat transfer at the interface between the metal and the dielectric substrate. Laser energy is absorbed in a skin depth, which for gold at a laser wavelength of 1 \mu m is only 3 nm, much smaller than the film thicknesses considered here. We account for the dependence on grating parameters such as groove spacing, depth, and shape by adjusting the absorption coefficient. The temperature T(x, t) throughout the film is controlled by the one-dimensional heat-conduction equation

\[
\frac{\partial T(x,t)}{\partial t} = \alpha \frac{\partial^2 T(x,t)}{\partial x^2}.
\]

![Fig. 1. Damage to gold film with 1053-nm pulses: (a) long pulse, 900 ps; (b) short pulse, 0.6 ps.](image)
where \( \alpha = \kappa_T/r_c p \) is the thermal diffusivity, \( \kappa_T \) is the thermal conductivity of the metal, \( p \) is the density, and \( c_p \) is the heat capacity. We assume that damage occurs when the temperature reaches the melting point of the metal for long pulses or the boiling point for short pulses.

Starting with a gold film at a uniform temperature, Fig. 2 shows the temperature distribution in a 100-nm-thick gold coating immediately following irradiation by 500-mJ/cm² 1053-nm laser pulses of various durations. As a result of the finite thermal diffusivity of gold, the thermal wave cannot penetrate more than approximately 10 nm (\( \alpha t/L^2 \approx 1 \), with \( \alpha = 143 \) nm²/ps) for picosecond pulses. At this fluence the surface is heated beyond the boiling point and is damaged, regardless of the thickness of the coating. For longer pulses the thermal diffusivity is sufficiently high that metals can conduct heat away from the surface during the pulse. For pulses that deposit their energy during a few tens of picoseconds the surface temperature is depressed by conduction throughout the bulk of the film. For pulses longer than 100 ps thermal equilibration across the film is nearly complete. In this case the bulk temperature, and hence the damage threshold, is determined by the thickness of the coating.

We use this simple one-dimensional heat-conduction model to predict the dependence of damage threshold on coating thickness and laser pulse width. In Fig. 3 we show our calculated and measured damage thresholds for 1053-nm laser pulses incident upon gold films of various thicknesses. For long (800-ps) pulses there is an approximately linear dependence on film thickness up to 200 nm, followed by an asymptotic approach to the damage threshold of the bulk material. For short (600-fs) pulses our model predicts a linear dependence on film thickness near the penetration depth. Beyond this, the damage threshold becomes independent of film thickness. The difference between the model and the measured data for coatings thinner than 100 nm was due to pinholes and defects in our films. As the coating thickness increased above 100 nm, the coatings became more uniform and increasingly defect free. The calculations shown in Fig. 3 are also applicable to high-efficiency gratings, provided that any change in the absorption of the incident radiation owing to the presence of the grating structure is considered. Although it agrees well with our measurements, our simple model neglects the two-temperature (electron, ion) description of the thermal response of metals to laser excitation and the temperature dependence of the surface absorption.

The pulse-width dependences of the damage threshold for a gold mirror and a high-efficiency gold diffraction grating are shown in Fig. 4. The 1480-line/mm grating was produced with a gold thickness of 200 nm and exhibited 94% diffraction efficiency in first-order Littrow configuration. It was damage tested at its 52° angle of use. The gold thickness of the mirror was 500 nm, and it was tested at 0°. As shown, the damage threshold of each optic decreases as \( \tau^{1/2} \) until approximately 200 ps, below which the threshold becomes independent of pulse width. This is the same qualitative dependence observed by Corkum et al., for solid copper and molybdenum mirrors at 9.3 μm. Following their analysis, our measured critical time of \( \tau_c = 200 \) ps corresponds to an electron–phonon coupling factor of \( g = 3.5 \times 10^{11} \) J/(s K cm³).

### 4. DAMAGE TO FUSED SILICA

Because gold-coated optics cannot withstand fluences much greater than 0.6 J/cm² in the short-pulse (subpicosecond) regime, we developed multilayer dielectrics to provide the high-damage-threshold gratings necessary for pulse compression of high-energy (kilojoule) laser pulses. We began with the desire to understand the physical damage mechanisms in dielectrics and chose...
fused silica as our first sample because of its availability in high-purity samples and its well-known materials properties.

A. Measured Damage Thresholds
The results presented here were obtained with 1-cm-thick superpolished fused-silica samples (Corning 7940) exhibiting less than 1-nm rms surface roughness. We measured the same damage thresholds with a 200-μm-thick fused-silica étalon, which we tested to examine any possible differences between thick and thin samples. Some samples were cleaned with acetone or methanol, but 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 (0.3-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 with short pulses, which would ionize and ablate any low-lying states or surface contamination with lower threshold, gave the same threshold as our measurements with constant fluence. We believe that our measurements correspond to a uniform, defect-free surface and can be compared with calculations based on the intrinsic properties of fused silica.

Our measured threshold damage fluence for fused silica at 1053 nm is shown in Fig. 5 as a function of laser pulse length (FWHM). The data at 140 fs and 825 nm were taken to confirm the decreasing trend in damage fluence with pulse width. In the long-pulse regime (τ > 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 occurs over the entire area irradiated, as shown in the electron micrograph of Fig. 6(a). The damage is thermal in nature and characterized by melting and boiling of the surface. This is more easily seen in Fig. 7(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 [Fig. 6(b)]. Damage occurs only over an area with sufficient intensity to produce ionization. With insufficient time for energy coupling to the lattice, there is no collateral damage. As a result the damaged area can be many orders of magnitude smaller with short (τ < 10-ps) pulses than with long pulses. For the case of fused silica the damaged area produced by the 0.5-mm diameter, 500-fs pulse was 2 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 [Fig. 7(b)].

The damage threshold in the short-pulse limit occurs within a very narrow range of fluence (2%), in contrast to that for long pulses. Fused silica irradiated with 10,000 shots at 2% below our determined threshold showed no evidence of damage with 0.4-ps pulses. For long pulses...
that in gases, leads to an irreversible change in the conduction band. The resulting avalanche, similar to ionization promotes another valence electron into the conduction band by scattering from phonons. If an electron can achieve oscillating in response to the laser field, transfer energy electron avalanche in which conduction-band electrons, in wide-band-gap dielectrics for which the energy and momentum scattering rates and the multiphoton ionization rates are known.

We take the damage threshold to be indicated by the occurrence of a sufficiently high electron density. A reasonable lower limit would be of the order of $10^{21}$ cm$^{-3}$, roughly the density at which the energy density of conduction electrons equals the binding energy of the lattice. A more realistic choice is the critical electron density $n_{cr}$ at which the plasma becomes reflective ($10^{21}$ cm$^{-3}$ for 1053 nm), as it is just below this density that the laser is strongly absorbed. Our calculations indicate the threshold is only logarithmically dependent on this choice.

For insulators or other materials with a large band-gap energy, $U_1 \gg \hbar \omega$, the number density $f(\epsilon, t)\, d\epsilon$ of electrons with kinetic energy between $\epsilon$ and $\epsilon + d\epsilon$ at time $t$ can be described by a Fokker–Planck equation:

$$\frac{\partial f(\epsilon, t)}{\partial t} + \frac{\partial}{\partial \epsilon} V(\epsilon)f(\epsilon, t) - D(\epsilon) \frac{\partial f(\epsilon, t)}{\partial \epsilon} = \frac{\partial f(\epsilon, t)}{\partial t} + \frac{\partial J(\epsilon, t)}{\partial \epsilon} = S(\epsilon, t),$$

where

$$V(\epsilon) = \frac{\sigma(\epsilon)E^2(t)}{3} - U_{\text{phon}}\gamma(\epsilon)$$

accounts for electron energy transfer to the lattice at rate $\gamma(\epsilon)$ and joule heating of the electrons with conductivity per electron $\sigma(\epsilon)$:

$$\sigma(\epsilon) = \frac{e^2}{m^*\left(1 + \omega^2\tau_m^2\right)}.$$

The transport (momentum) scattering rate $1/\tau_m(\epsilon)$ and the energy scattering rate $\gamma(\epsilon)$ are both energy dependent, varying in fused silica by 2 orders of magnitude for energies in the conduction band. The electron energy diffusion $D(\epsilon)$ is proportional to both the electron conductivity and the laser intensity:

$$D(\epsilon) = \frac{2\sigma(\epsilon)E^2\epsilon}{3}.$$

In Eqs. 3–5, $E$ is the laser electric field amplitude oscillating at frequency $\omega$ and $U_{\text{phon}}$ is the characteristic phonon energy (0.033 eV). The final term in Eq. 2 includes sources and sinks of electrons provided by impact and multiphoton ionization:

$$S(\epsilon, t) = R_{\text{mp}}(\epsilon, t) + R_{\text{pl}}(\epsilon, t).$$

**Figure 8.** Damage threshold fluence of fused silica with 0.4-ps pulses of varying diameter gives consistent values.

**Figure 9.** Single-shot damage spot size (●) approaches zero as the laser fluence is reduced to our multiple-shot threshold value (○).
Impact ionization was included assuming that the excess kinetic energy is equally divided between the two resultant electrons:

\[ R_{\text{imp}}(\epsilon, t) = -\nu_i(\epsilon)f(\epsilon) + 4\nu_i(2\epsilon + U_i)f(2\epsilon + U_i). \]  

(7)

We used the Keldysh impact ionization rate, \( \nu_i(\epsilon) = 1.5(\epsilon/U_i - 1)^2 \text{fs}^{-1} \). Integration of Eq. (7) over energy shows that the net rate of electron production is simply \( \int d\epsilon \nu_i(\epsilon)f(\epsilon) \). The source term \( S(\epsilon, t) \) also includes multiphoton ionization at rate \( R_{\text{m}}(\epsilon, t) \). For 1053-nm light, eight-photon absorption cross-section values were not available, so we used the strong-field Keldysh formula for the multiphoton ionization rate \( P(I) \). Evaluation of the Keldysh expression leads to a result that is fitted very well by the eight-photon absorption form. \( \nu_i(\epsilon) \) is the intensity in terawatts per square centimeter. This expression is valid up to intensities of the order of \( 10^9 \text{TW/cm}^2 \). For extremely short intense pulses, tunneling through the binding barrier takes place during a time shorter than the laser period. In this case \( P(I) \) is given by the ADK tunneling ionization expression. With 526-nm light, four-photon absorption is the relevant process and \( P(I) = \sigma_4(I/\hbar\omega)^4N_s \). We used the cross section \( \sigma_4 = 2 \times 10^{-144} \text{cm}^8 \text{s}^4 \). This was measured for NaCl, but other insulators have nearly the same value. The quantity \( N_s \) is the solid atom density. In any case, our results are not highly sensitive to the exact numerical value of these photoionization rates. The boundary conditions for Eq. (2) require the vanishing of the distribution function \( f(\epsilon, t) \) at \( \epsilon = -\infty \) and the current \( J(\epsilon, t) \) at \( \epsilon = 0 \).

The electron number density \( n \) and the average kinetic energy per electron \( \langle \epsilon \rangle \) are defined by the moments of the electron distribution:

\[ n = \int_0^\infty f(\epsilon)d\epsilon, \]  

(8)

\[ n(\epsilon) = \int_0^\infty \epsilon f(\epsilon)d\epsilon. \]  

(9)

Electron scattering from various types of phonon determines the transport scattering and loss rates needed in solving Eq. (2). Currently it is impossible to construct a first-principles theoretical model of all these interactions. We used the semiempirical model of Arnold et al. which gives a good account of electron scattering in fused silica.

A solution of the full kinetic equation in Fig. 10 illustrates the evolution of electron density produced by a 100-fs, 12-TW/cm² laser pulse. The pulse intensity and the electron density produced by photoionization alone are included for reference. Because photoionization is extremely intensity dependent, the electron production takes place principally at the peak of the pulse. After these seed electrons are produced, a small electron avalanche reaches the plasma critical density. This dense plasma is not produced until late in the pulse, and only the trailing edge of the laser pulse experiences strong absorption or reflection.

C. Rate-Equation Approximation

To gain additional physical insight, we investigated the validity of a rate-equation description of the conduction electron number density and average energy. For the rate-equation approach to be valid the transient behavior must be minimal, the structure of the electron distribution function should remain relatively constant after the transient, and the addition of source terms must not significantly distort the distribution function.

A numerical solution of the kinetic equation at constant laser intensity and excluding multiphoton ionization shows that an avalanche is established within a few femtoseconds for an intensity of 1 TW/cm² (see Fig. 11). During the avalanche the electron distribution grows in magnitude without changing shape, \( f(\epsilon, t) = g(\epsilon)\exp(\beta t) \). The distribution \( g(\epsilon) \) is stationary, but non-Maxwellian. The length of the initial transient is weakly dependent on initial conditions and decreases monotonically with intensity growth. The ionization rate \( \beta \) is proportional to the laser intensity because all the energy absorbed from the pulse is used to produce electrons.

The shortness of the transient solution for constant intensity suggests that, for a time-varying pulse shape \( I(t) \), we may expect a solution for the electron distribution function of the form

\[ f(\epsilon, t) = g(\epsilon)\exp\left(\int_{-\infty}^t \beta dt'\right). \]  

(10)

The supposition that \( \beta \) is proportional to \( I \), \( \beta = \alpha I(t) \), is
tested in Fig. 12, in which we plot, from a solution of the full kinetic equation, the electron density as a function of the instantaneous fluence for the case of a Gaussian pulse. It is evident that the linearity between $\beta$ and $I$ holds throughout nearly the entire pulse. Using this exponential growth and the proportionality between $I$ and $b$, pulse. It is evident that the linearity between transient the electron growth rate is proportional to the laser intensity. From this slope we find the value of $a$.

Note that $1/\alpha$ has units of energy per area; it provides a scale fluence for damage. The avalanche rate $\alpha$ is calculated with the full kinetic equation and is physically determined by the scattering rates and the band gap in the dielectric.

We now reintroduce multiphoton ionization $R_m$ in the source term $S(\epsilon, t)$. This term takes the form $P(I)F(\epsilon)$, where $P(I)$ is the multiphoton ionization rate and $F(\epsilon)$ is the distribution function of the photoelectrons normalized so that $\int F(\epsilon)d\epsilon = 1$. The presence of photoionization may perturb the electron distribution function $f(\epsilon, t)$, bringing into question the validity of Eq. (10). With a peaked temporal intensity profile (such as our Gaussian), photoionization rates and thus the initial electron production are very strongly peaked at the center of the pulse. Avalanche ionization occurs predominantly during the second half of the pulse, after the creation of the initial electrons. This can be seen from Fig. 11. The rate equation

$$\frac{dn}{dt} = \beta n = \alpha I(t)n. \quad (11)$$

The scale fluence $2/\alpha$ is 0.2 J/cm$^2$ for fused silica. The breakdown threshold fluence is only logarithmically sensitive to the multiphoton cross section and the plasma critical density.

From the form of Eq. (13) one can see that if the number of seed electrons were independent of intensity, e.g., as the result of defects, $\phi_{cr}$ would be independent of pulse duration. In the present case, $n_0$ increases rapidly with intensity, so $\phi_{cr}$ decreases with pulse duration as the avalanche becomes less important. It is clear from Eq. (13) that avalanche ionization is not significant if the threshold fluence is such that $\alpha \phi_{cr}/2$ is small, i.e., $\phi_{cr}$ of the order of 0.2 J/cm$^2$. Below this fluence, multiphoton ionization is almost completely responsible for dielectric breakdown. Indeed, because photoionization produces an ever-increasing share of electrons for shorter pulse lengths, the short-pulse damage threshold must asymptotically approach the fluence at which photoionization alone produces the electron critical density $n_{cr}$. The threshold fluence for such short pulses scales as

$$\phi_{cr} \propto \tau^{m-1/m} n_{cr}^{1/m}.$$

when $m$-photon ionization is the dominant process.

In Fig. 14, we compare our measured and calculated damage thresholds at both 526 and 1053 nm. We calculated the solid curves from the rate equation (easier to solve), using the avalanche rate $\alpha$ determined by the full kinetic equation. These results were verified at a number of discrete points by a complete solution of the full
kinetic equation. The results of the above theoretical modeling of laser-induced damage in the short-pulse limit are in very good agreement with both pulse-width and wavelength scaling of the experimental data. As shown, with decreasing pulse width the damage threshold will asymptote to the limit where multiphoton ionization alone creates sufficient electron density to cause damage.

5. DAMAGE TO FLUORIDES

The damage threshold of calcium fluoride as a function of pulse width exhibits a dependence similar to that of fused silica (Fig. 15). In the long-pulse limit the threshold fluence scales as \( t^{1/2} \) and then breaks away to the short-pulse regime near 20 ps. For long pulses the damage morphology is again consistent with heating and melting. Figure 16(a) shows the melting and recrystallization of the calcium fluoride surface layers, which occurred with no evidence of avalanche breakdown. This is consistent with measurements of wide-gap alkali halides in the long-pulse regime (100 ps). Short-pulse damage clearly initiates on scratches left from the polishing process [Fig. 16(b)], although the damage threshold did not appear to be greatly influence by the polishing sleeks. Our measured short-pulse (0.4-ps) damage thresholds of BaF\(_2\) (1.6 J/cm\(^2\)), CaF\(_2\) (2.0 J/cm\(^2\)), MgF\(_2\) (2.1 J/cm\(^2\)), and LiF (2.5 J/cm\(^2\)) scale with band-gap energy, as expected from multiphoton-initiated avalanche ionization.

6. DAMAGE TO MULTILAYER DIELECTRICS

We have tested several different multilayer dielectric mirrors and polarizers, with each multilayer stack consisting of approximately 20 individual layers of thickness 0.1–0.3 \( \mu \)m. Figure 17 shows the pulse-width dependence of the 1053-nm damage threshold fluence for a 45° high reflector and a 57° polarizer used in reflection. The long-pulse scaling of each is slightly less than \( t^{1/2} \), and again there is a transition to the short-pulse regime near 20 ps. The long-pulse damage morphology of the mirror is again characterized by melting and flow, whereas short pulses cause ablation of the individual dielectric layers (Fig. 18). As has been thoroughly characterized by Kozlowski and Chow, the initiation of damage with long (nanosecond) pulses is dominated by nodules and defects. We find the same behavior with short (0.3-ps) pulses, for which the presence of defects reduces the damage threshold by as much as 50%, depending on the size and type of defect. We tried laser-conditioning the mirror to improve the short-pulse damage threshold. Unfortunately, neither long- nor short-pulse conditioning resulted in higher short-pulse damage threshold.

The short-pulse (0.4-ps) damage thresholds of the multilayer dielectrics that we have tested typically range between 0.7 and 1.35 J/cm\(^2\). These samples consisted of a variety of mirrors and polarizers with either HfO\(_2\)/SiO\(_2\) or Ta\(_2\)O\(_5\)/SiO\(_2\) multilayers deposited by e-beam evaporation or ion-beam sputtering. As with long pulses, we find that the key to achieving increased short-pulse damage thresholds is the reduction in size and number density of the defects. We expect that, with proper design and production, diffraction gratings based on these multilayer structures will exhibit damage thresholds comparable with those of the multilayer itself.
than the laser pulse. The damage threshold continues to decrease with decreasing pulse width but at a rate slower than \( t^{1/2} \) in the range 0.1–20 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. Many applications, ranging from materials processing to biomedical technologies, could potentially benefit from the more localized energy deposition with these short pulses.

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 pulse-width and the wavelength scaling of our measurements. For extremely short pulses (\( t < 30 \) fs), multiphoton ionization alone will provide the critical density of electrons.

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