

Femtosecond Optical Breakdown in Dielectrics

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We report measurements of the optical breakdown threshold and ablation depth in dielectrics with different band gaps for laser pulse durations ranging from 5 ps to 5 fs at a carrier wavelength of 780 nm. For $\tau < 100$ fs, the dominant channel for free electron generation is found to be either impact or multiphoton ionization (MPI) depending on the size of the band gap. The observed MPI rates are substantially lower than those predicted by the Keldysh theory. We demonstrate that sub-10-fs laser pulses open up the way to reversible nonperturbative nonlinear optics (at intensities greater than 10^{14} W/cm² slightly below damage threshold) and to nanometer-precision laser ablation (slightly above threshold) in dielectric materials. [S0031-9007(98)05969-9]

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Laser-induced breakdown resulting in damage to dielectrics has been the subject of extensive experimental and theoretical investigations since powerful lasers became available [1–3]. It has been described in terms of three major processes: (i) the excitation of electrons in the conduction band by impact and multiphoton ionization (MPI), (ii) heating of the conduction-band (henceforth free) electrons by the radiation, and (iii) transfer of the plasma energy to the lattice. For pulses of a few picoseconds or shorter, heat diffusion is “frozen” during the interaction [4] and the shocklike energy deposition leads to ablation. This new regime of laser-matter interactions holds promise for a number of intriguing applications in science and technology.

Although breakdown experiments were recently extended to the subpicosecond regime [5–9], both the nature of the avalanche and the role of multiphoton ionization have remained controversial up to now. Du *et al.* [5] were the first to observe a deviation from the $\sqrt{\tau}$ scaling of breakdown threshold fluence F_{th} and an increasingly deterministic character of breakdown for $\tau < 10$ ps as opposed to longer pulses. These observations were explained in terms of an avalanche scaling with the square root of the laser intensity, and MPI was found to serve only for the production of seed electrons for the avalanche. By contrast, Stuart *et al.*’s model [6] yields an avalanche that scales *linearly* with the laser intensity. Combining this model with the Keldysh MPI rate [10], these investigators found that MPI is likely to become the dominant channel for free electron generation for $\tau < 100$ fs, which was predicted to result in F_{th} as low as < 0.1 J/cm² for $\tau \approx 10$ fs in fused silica. In this Letter, we report $F_{th} \approx 1.5$ J/cm² in fused silica for $\tau \leq 10$ fs. Our investigations confirm a linear scaling of the avalanche with intensity [6] and yield MPI rates which are orders of magnitude lower than predicted by Keldysh’s theory [11].

This work significantly extends previous studies of ultrashort-pulse-induced breakdown in several respects.

The pulses for femtosecond breakdown experiments are delivered by a spatially filtered beam for the first time. The high-quality beam and the absence of heat diffusion and melting result in a regular morphology of the damaged sites (see Fig. 1). Thus, a precise quantification of parameters such as the ablated volume and the ablation depth becomes feasible. These “above-threshold” measurements permit a determination of F_{th} that, in contrast with previous investigations, is not affected by instrument resolution and sensitivity. Extensive measurements of the threshold fluence $F_{th}(\tau)$ supplemented with those of the ablation depth $d_a(F, \tau)$ give valuable new insight into the nature of the relevant carrier generation processes and provide a selective test of existing models of femtosecond optical breakdown. Experiments of this kind have been extended to $\tau \ll 100$ fs and performed on dielectrics with distinctly different band gaps for the first time.

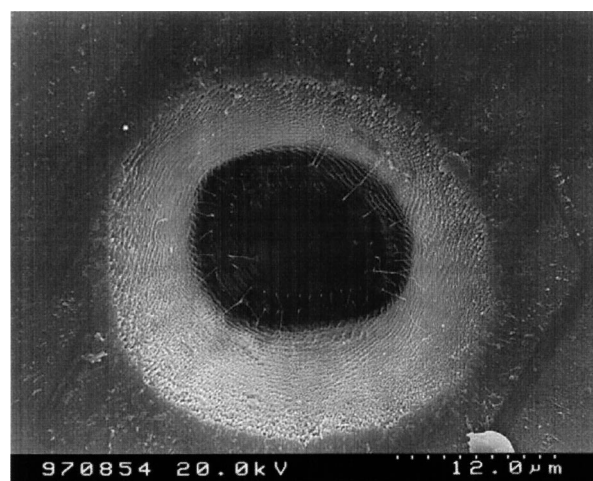


FIG. 1. Scanning electron micrograph of front-surface damage on fused silica produced by 5-fs pulses at a fluence of $6 \times F_{th}$. The regular morphology provides conclusive evidence of an excellent laser beam quality free from hot spots in the intensity profile.

The Ti:sapphire laser system used for the experiments is described in detail elsewhere [12]. The output pulses emerge from a hollow-fiber-chirped-mirror compressor seeded with 20-fs 1-mJ pulses at a 1-kHz repetition rate. Bandwidth-limited pulses with durations between 5 and 20 fs can be generated by varying the argon gas pressure in the hollow fiber. Pulse durations of $\tau \geq 20$ fs are realized by evacuating the fiber and temporally stretching the pulses. For all pulse durations the hollow fiber acts as an efficient spatial filter, resulting in a nearly diffraction-limited output beam ($M_{x,y}^2 < 1.1$) [12]. The linearly polarized laser pulses were focused on the front surface of the sample to a $1/e^2$ diameter of $\approx 30 \mu\text{m}$ [13]. Imaging the focused spot onto a CCD camera with calibrated magnification allowed accurate characterization of the intensity distribution on target. Experiments were performed on two isotropic dielectrics, fused silica (FS, Corning 7940), and barium aluminum borosilicate (BBS, Corning 7059). The surfaces of the samples were formed by a direct drawing process from the melt, resulting in a residual surface roughness of < 10 nm and < 13 nm for FS and BBS, respectively. The band-gap energies of the two materials are $\Delta_{fs} \approx 9$ eV and $\Delta_{bbs} \approx 4$ eV.

In situ monitoring of breakdown was accomplished by a He-Ne laser focused onto the central fraction of the interaction region. For a quantitative evaluation of ablation, the samples were investigated by light and scanning electron microscopy. In order to make V_a and d_a accurately measurable, each site was exposed to 50 pulses at a given fluence. As an example, Fig. 2 depicts the ablated-volume/pulse V_a versus on-axis fluence F for two different pulse durations in FS. The linearity of $V_a(F)$ is striking and found to be a general feature for the entire pulse width

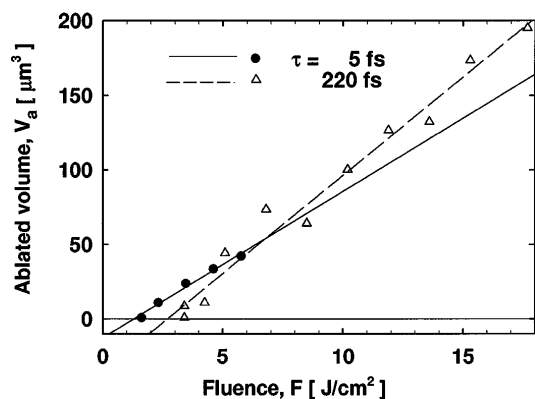


FIG. 2. Volume V_a of fused silica ablated by one laser pulse versus energy fluence F for two different pulse durations. Each site was irradiated by 50 laser pulses. The straight lines are obtained from linear regression. The intersection of the regression line with the horizontal axis ($V_a = 0$) yields the threshold fluence F_{th} for optical damage. For pulse durations $\tau \geq 20$ fs, typically 10 measured data points in the range between F_{th} and $\approx 10 \times F_{th}$ have been taken. For $\tau < 20$ fs, only data obtained at $F \leq 6 \text{ J/cm}^2$ were considered for regression to avoid the influence of plasma formation in air.

regime studied. This characteristic can be utilized for determining the damage threshold fluence F_{th} by extrapolating the regression line of V_a on F to $V_a = 0$ [14]. It is inherent in this procedure that even relatively small residuals yielding a robust gradient tend to give rise to comparatively large confidence intervals for F_{th} . Yet, we opted for this way of evaluating the threshold fluence, because it permits a *fully objective* determination of F_{th} , allowing reproduction of the reported data or extension of the presented studies to other materials under exactly the same conditions. Figure 3 shows F_{th} for pulse durations between 5 ps and 5 fs in FS and BBS. The error bars depict relative (random) errors, the absolute (systematic) error of the measurements is less than $\pm 15\%$.

For $\tau > 100$ fs the observed qualitative dependence of F_{th} on τ agrees well with those reported in previous studies for various dielectrics [6,7,9]. Quantitatively, F_{th} for fused silica obtained in our experiments agrees (within the error bars) with those presented in Ref. [7] for $\tau > 190$ fs but exceed by approximately a factor of 2 those reported in Ref. [6]. This same discrepancy appears also with respect to our previous results obtained with the same samples [14,15] for $20 \text{ fs} \leq \tau \leq 3 \text{ ps}$ using the same laser *without* spatial filtering. We surmise that extrinsic effects related to possible hot spots in the intensity profile of the laser beam might have lowered the threshold in our previous experiments. Starting from the same level at $\tau \geq 1$ ps, F_{th} in BBS becomes by a factor as large as ≈ 3 smaller as compared to FS for $\tau \leq 20$ fs. In order to gain insight into the processes responsible for this behavior, the ablation-depth/pulse d_a has also been evaluated for various pulse durations for $F > F_{th}$.

Figures 4 and 5 show the accumulated ablation depth as a function of the number of laser shots for FS and BBS, respectively, revealing important differences (A) and similarities (B) in the behavior of the two materials. (A) The comparable slopes of the regression lines in Fig. 4

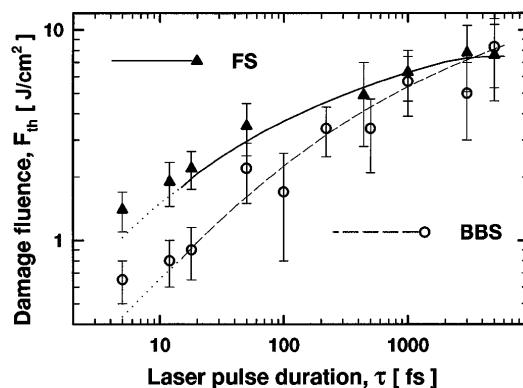


FIG. 3. Threshold fluence in FS and BBS versus τ (full width at half maximum) at $\lambda = 780$ nm. Each site was irradiated by 50 laser pulses. The error bars represent standard deviations and the lines depict theoretical fits. Further details are given in the text.

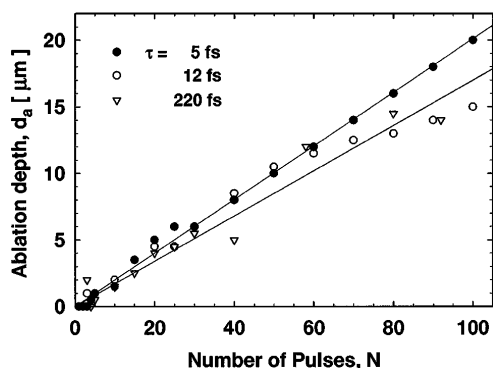


FIG. 4. Ablation depth in FS for increasing number of pulses and different pulse durations, measured at fluence levels of $(5 \pm 1) \text{ J/cm}^2$. The lines represent linear regressions.

yield ablation depths d_a that exhibit hardly any dependence on the pulse duration for FS. In strong contrast, such a τ invariance is limited to the subpicosecond regime in BBS (Fig. 5), whereas d_a rapidly decreases for decreasing pulse durations as τ approaches the 10-fs regime. (B) The reproducibility of ablation is, in both materials, substantially higher in the 10-fs than in the subpicosecond regime. In fact, from the data shown in Figs. 4 and 5, we obtain $d_a(220 \text{ fs}) = (170 \pm 35) \text{ nm}$, $d_a(5 \text{ fs}) = (200 \pm 10) \text{ nm}$ for FS and $d_a(220 \text{ fs}) = (270 \pm 65) \text{ nm}$, $d_a(5 \text{ fs}) = (115 \pm 8) \text{ nm}$ for BBS with the uncertainties representing a 99% confidence interval. The extremely small uncertainty in $d_a(5 \text{ fs})$ for FS is particularly remarkable, considering that it is limited by pulse energy fluctuations (rms $\approx 2\%$).

Recently Stuart *et al.* [6] derived a simple rate equation for the evolution of the free electron density $n(t)$ in a dielectric medium exposed to intense laser radiation,

$$\frac{dn}{dt} = \alpha I(t)n(t) + \sigma_k I^k, \quad (1)$$

where $I(t)$ is the intensity of the laser pulse, α is the avalanche coefficient, and σ_k is the k -photon absorption

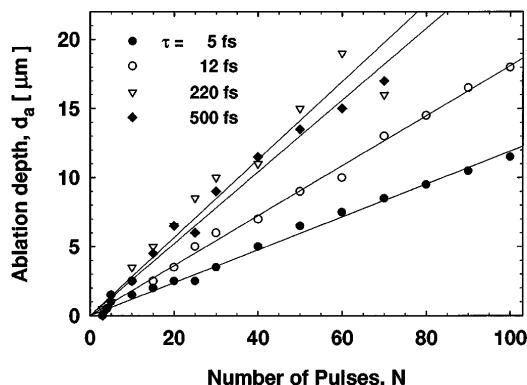


FIG. 5. Ablation depth in BBS for increasing number of pulses and different pulse durations at fluence levels of $(6.2 \pm 0.7) \text{ J/cm}^2$. The lines depict linear regressions.

cross section with the smallest k satisfying $k\hbar\omega \geq \Delta$, where ω is the laser frequency. The energy of the free electrons heated by the laser is subsequently transferred to the lattice. This energy transfer leads to the ablation of the heated zone, which is the major manifestation of femtosecond optical breakdown. In what follows, we show that our experimental observations can be consistently interpreted in terms of Eq. (1).

F_{th} can be predicted by postulating a threshold electron density n_{th} associated with the onset of permanent damage and solving the rate equation. In strong contrast with long-pulse damage, the density of seed electrons for the avalanche does not have to be postulated because it no longer relies on thermal excitation of impurity states, but can be produced by MPI with rapidly increasing efficiency for decreasing pulse duration. The dramatically increased reproducibility of ablation in the 10-fs regime reported above is a direct consequence of the strongly increased deterministic seed electron production for the avalanche. Here we choose, in accordance with previous studies [5,6], $n_{th} = 10^{21} \text{ cm}^{-3}$, which is near the plasma critical density for the laser wavelength. Solving (1) for a range of values of α and σ_k and using χ^2 fitting to the data obtained for $\tau \geq 20 \text{ fs}$ (where MPI applies, as shown below), we obtain the fit parameters $\alpha_{fs} = (4 \pm 0.6) \text{ cm}^2/\text{J}$ and $\sigma_6 = 6 \times 10^{8 \pm 0.9} \text{ cm}^{-3} \text{ ps}^{-1} (\text{cm}^2/\text{TW})^6$ for FS and $\alpha_{bbs} = (1.2 \pm 0.4) \text{ cm}^2/\text{J}$ and $\sigma_3 = 7 \times 10^{17 \pm 0.5} \text{ cm}^{-3} \text{ ps}^{-1} (\text{cm}^2/\text{TW})^3$ for BBS. The errors given are related to a factor of 2 increase in the χ^2 merit function with respect to its minimum value. Whereas α_{fs} is in reasonable agreement with the prediction of the kinetic theory of Stuart *et al.* [6], σ_6 and σ_3 are by some 6 and more than 2 orders of magnitude smaller as compared to the prediction of the Keldysh formula, respectively [10,16].

The theoretical prediction obtained with the above fit parameters is depicted by solid and dashed line for FS and BBS, respectively, in Fig. 3. The simple perturbative expression for MPI used in (1) tends to fail as the Keldysh parameter $\gamma = \omega\sqrt{2m\Delta}/eE$ (m and e are the mass and charge of the electron, and E is the peak laser field) is becoming comparable to or smaller than unity [10] near threshold for decreasing pulse durations. For this regime the lines representing the predictions of (1) are dotted (to indicate the questionable applicability of the model) in Fig. 3. As γ decreases significantly below 1, ionization in this strong-field, low-frequency limit can be interpreted in terms of the quasistatic tunnel effect [10]. For $\tau = 5 \text{ fs}$ the measured values of F_{th} yield $\gamma \approx 0.5$ for both materials. As a consequence, *photoionization is dominated by tunneling as the breakdown threshold is approached at 5 fs.*

Quantitative prediction of the penetration depth of the incident radiation, and hence that of d_a , would call for solving the coupled rate and wave equations for $n(z, t)$ and $E(z, t)$. Nevertheless, the influence of the pulse

duration on d_a , which is expected to scale inversely proportional to the free electron density, can be qualitatively assessed by inspecting Eq. (1). For a regime in which carrier generation is dominated by impact ionization, (1) predicts an electron density, and hence d_a , that is independent of τ at a fixed fluence. By contrast, in an MPI-dominated regime, d_a is expected to rapidly decrease for decreasing pulse durations (at $F = \text{const}$). The above-determined values of the avalanche and MPI coefficients now allow predicting the qualitative behavior of $d_a(\tau)$ and its comparison with the data in Figs. 4 and 5. The fraction of the critical density produced by photoionization at $F = F_{\text{th}}$ is calculated as $n_p(500 \text{ fs})/n_{\text{th}} \approx 4 \times 10^{-8}$ and $n_p(50 \text{ fs})/n_{\text{th}} \approx 1.5 \times 10^{-4}$ for FS, and $n_p(500 \text{ fs})/n_{\text{th}} \approx 0.09$ and $n_p(50 \text{ fs})/n_{\text{th}} \approx 0.35$ for BBS. These data suggest that *optical breakdown in fused silica is dominated by the avalanche process down to the 10-fs regime, whereas in BBS having a much smaller band gap, multiphoton ionization takes over for pulse durations below 100 fs* [17]. This finding is confirmed conclusively by the data in Figs. 4 and 5. As a matter of fact, d_a is virtually independent of pulse duration for FS in the entire femtosecond regime. In BBS, this applies only to the subpicosecond regime, with d_a becoming subject to a rapid decrease with decreasing pulse duration for τ approaching the 10-fs regime, in accordance with the above considerations [18].

Our results show that, even for a band gap as large as $\approx 9 \text{ eV}$, MPI produces some 10 orders of magnitude higher seed electron density in the 10-fs regime than available in thermal equilibrium. As a result, *sub-10-fs-laser ablation can be accomplished with a precision corresponding to a few tens of atomic layers*. Nevertheless, the MPI rate is orders of magnitude lower than the prediction of the Keldysh theory. This is a dramatic deviation, indicating that *collisions [11] and possible other, not yet identified, mechanisms strongly interfere with multiphoton ionization in dielectrics near breakdown*. The inhibited MPI increases the threshold fluences by more than an order of magnitude for $\tau \leq 10 \text{ fs}$ as compared to those predicted by the same model using the Keldysh ionization rate. The resulting anomalously high *breakdown thresholds in excess of 10^{14} W/cm^2 allow one to explore an entirely new—nonperturbative—regime of reversible nonlinear optics in solids*.

In summary, using a 5-fs spatially filtered laser source, we have performed a comprehensive experimental study of femtosecond optical breakdown in dielectrics. Equation (1) provides a powerful phenomenological description of this phenomenon. Our analysis results in a number of important findings *highlighted* in the last paragraphs.

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 - [17] This conclusion is also supported by a comparison of the relative uncertainties in the evaluated avalanche and MPI coefficients for FS and BBS.
 - [18] Simple inspection yields that our data shown in Figs. 3, 4, and 5 are inconsistent with a \sqrt{I} scaling of the avalanche as proposed in Ref. [5].