# Ultrafast Electron Dynamics in Femtosecond Optical Breakdown of Dielectrics

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We measured the optical breakdown threshold (OBT) in dielectrics with different band gaps for single and double 25-fs 800-nm transform-limited laser pulses. Our pump-probe double pulse measurements indicate that the plasma energy in dielectrics experiences ultrafast decay which lasts only ~100 fs and does not follow an exponential decay curve. Therefore, a decay term must be included in the electron density rate equation. Our double pulse measurements also demonstrate that the OBT is temperature dependent. The OBT in dielectrics was determined using a novel technique, which eliminates the ambiguity in its definition and also allows real-time data acquisition. [S0031-9007(99)08660-3]

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Many studies have been conducted on the ultrafast breakdown of dielectrics in order to understand the different processes involved [1–5]. Such investigations are important to such diverse fields as micromachining, medical physics, and solid state physics. Generally, laser-induced breakdown in dielectrics is described in terms of three major processes: (i) multiphoton ionization (MPI) and/or tunneling causing the excitation of electrons to the conduction band, (ii) electron-electron collisional ionization (avalanche process) due to Joule heating, and (iii) plasma energy transfer to the lattice [3,4,6]. While the first two processes deposit energy in the plasma, the third process releases the deposited energy to the lattice, thereby inducing the actual damage. This transfer of energy to the lattice is expected to occur after the laser pulse [3]. Until recently, the above processes were studied by measuring the pulse duration dependence of optical breakdown threshold (OBT). Although single pulse OBT measurements have been extended to the 5-fs range, it is very difficult to extract information regarding electron dynamics from such measurements, especially the time scale for the plasma energy decay.

We have developed a new pump-probe double pulse experiment, which is more sensitive to dynamic behavior. Although another pump-probe technique by Mazur *et al.* [7], with a resolution of ~100 fs, was proposed for the study of semiconductors, this is the first time that the time scale for the plasma energy decay in dielectrics has been directly measured with ultrahigh temporal resolution (25 fs). Our results from the double pulse OBT measurements indicate that this energy decay is an ultrafast process (~100 fs). Therefore, for a pulse longer than 100 fs, the plasma energy decays within the pulse duration, which indicates a need to redefine the breakdown process. Although we have modified the existing electron density model to include the decay behavior, our simplified model does not fit the data very well. In order to determine a definite time scale and to investigate the impact of including such decay behavior on interpreting other single pulse experiments, a more sophisticated model is needed.

Every experiment on OBT requires an experimental criterion for damage. Typically, OBT is determined using either visual acquisition [3] or by ablation depth measurement [4]. However, the former does not give a well-defined criterion for damage, while the latter is not a real-time technique. Our technique, which determines the OBT by monitoring the plasma radiation due to damage, gives a well-defined criterion in real time. We consider strong plasma radiation to be the signature of optical breakdown and we have verified this by two different methods.

The Ti:sapphire laser system used for the experiments is described in Ref. [8]. The standard output of the system is 25-fs 0.5-mJ transformation-limited pulses at a 1 kHz repetition rate. However, we usually run the system at 10 Hz to prevent the samples from being exposed to too many pulses above the OBT. The beam first goes through a half-wave plate and a polarizer so that we are able to vary the laser energy. The linearly polarized beam is then sent to a beam-splitting device, where each single pulse is split into two temporally identical pulses with variable delay from 0.067 to 130 ps set with a precision stepper motor (Newport). For a 25-fs Gaussian beam, the intensity drops to 0.68% of the maximum intensity after 33.5 fs from the peak. Therefore, even at the smallest delay (67 fs), the temporal overlap between two pulses is negligible. The zero time delay is carefully set by monitoring the temporal interference signal between the two pulses. The collinearity of beams is achieved by adjusting their interference pattern at zero time delay. Longer delays, up to 3 ns, are made possible by moving the base of a retroreflector along a track. Also, the pulses used are temporally smooth as shown by our FROG traces (~25 fs) [8]. The collinear beams are focused onto the front surface of the sample at near normal incidence by a gold-coated off-axis parabolic mirror (Janos, 2-in. focal length) to a diameter of $\approx 35\ \mu$m (Fig. 1). With a long-working-distance microscope and a CCD (charge-coupled device) camera, we are able to zoom in on the target spot in order to limit our field of view to only the neighborhood

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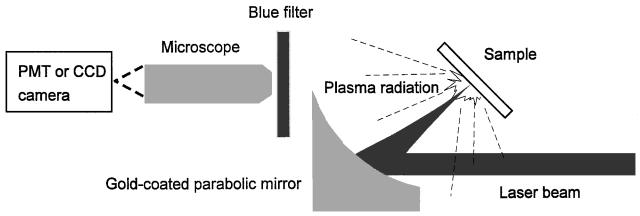


FIG. 1. Experimental setup.

of the spot on the surface. With proper calibration, we can also measure the spot size with the same setup. Experiments are then performed on samples of fused silica (FS, Corning 7940, 0.5 mm thickness) and barium aluminum borosilicate (BBS, Corning 7059, 0.4 mm thickness) with band gaps of about 9 and 4 eV, respectively. The surface is polished to optical quality and a new spot is used for each run. Since the samples we use are transparent, the amplified spontaneous emission (ASE) from the laser system does not influence our results. During data acquisition, the CCD camera is replaced with a photomultiplier tube (PMT) (Products for Research, Inc.) to measure the plasma radiation from the breakdown region of the samples. In order to minimize the pickup of the scattered laser light, a blue filter (Oriel, 51690) is placed in front of the PMT. Since the plasma radiation has a broad bandwidth, a significant amount of plasma radiation is transmitted through the blue filter. Only thin samples are used in the experiments to ensure weak generation of self-phase modulation (SPM) from the bulk, without reducing the plasma radiation generation due to surface damage. The output of the PMT is integrated by a boxcar (Stanford Research System) and then sent to a data-acquisition computer. This computer also collects integrated boxcar signals from a reference photodiode, which monitors the laser energy. The synchronized PMT and reference photodiode signals are displayed on the computer monitor in real time.

During the experiment, we start with zero laser energy and slowly increase it. The PMT output strength increases accordingly. There are two possibilities, as shown in Fig. 2(a). In case 1, the signal always follows line $AB$. There is no breakdown in this case. The increase of signal strength is due mainly to the residual SPM, which passes through our blue filter. In case 2, the signal follows line $AB$, and then it follows line $BC$. There is breakdown in this case. The portion from $A$ to $B$ is still mainly from SPM, while the remainder ($B$ to $C$ and beyond) is dominated by the plasma radiation. Since strong plasma radiation is considered to be the signature of breakdown, we define the interception point to be the OBT.

One independent test of the validity of our method is whether the output signal exhibits hysteresis. This was done by initially increasing the laser energy from zero to its maximum value and then decreasing it slowly to see if the PMT signal retraces its original path. The hysteresis loop is seen whenever there is a rapid change in slope.

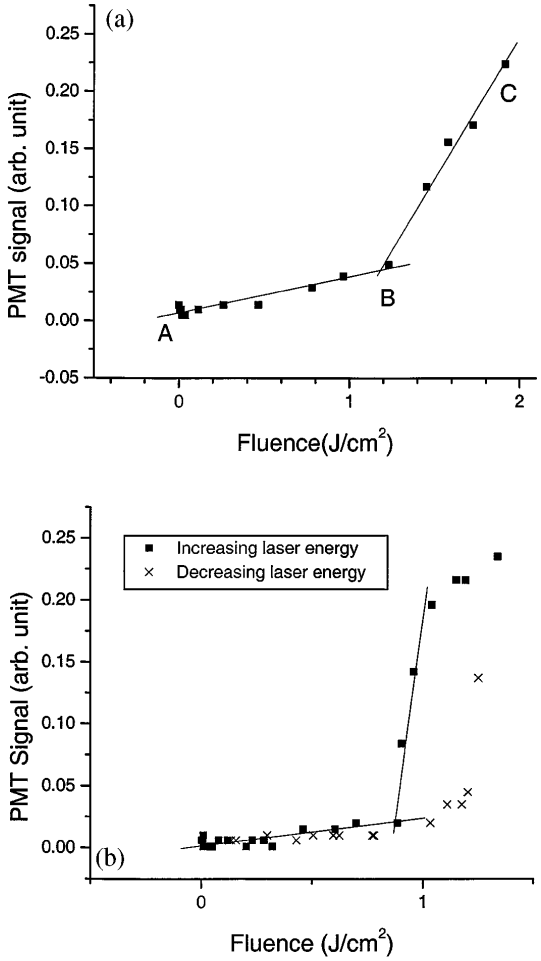


FIG. 2. (a) OBT as indicated by the rapid change in slope of the PMT signal. (b) PMT signal exhibits hysteresis after breakdown occurs.

This implies that the sample has undergone irreversible surface modification, which in turn implies permanent damage [Fig. 2(b)]. The actual shape of the hysteresis loop depends on the maximum laser fluence reached during each run. However, no such effect is seen in case 1 of Fig. 2(a), where there is no significant change in slope. A second test of validity is visual acquisition. For this purpose, the possible damage site is illuminated with amplified spontaneous emission and the scattered light is observed with the long-working-distance microscope and CCD camera combination. Since the samples have very high optical quality, little scattered light is seen unless damage occurs. Our results confirm that the scattered light is very bright whenever the hysteresis loop is exhibited.

In the experimental setup described above, with either beam blocked, we were able to measure the single pulse breakdown threshold. As mentioned before, the laser energy was continuously varied until a rapid change of slope occurred, which was then taken to be the OBT. For a 25-fs pulse, the threshold was measured to be 1.50 J/cm$^2$ for FS and 0.83 J/cm$^2$ for BBS with an error

of approximately 7.5%. This agrees with the results in Ref. [4] to approximately a factor of 1.5, which were obtained with a completely different technique.

In the double pulse measurements, both beams were always used below the single pulse breakdown threshold. The energy ratio (1:1.3) between the two beams was kept constant, with the delayed beam (probe beam) being the stronger one. Their laser energies were simultaneously varied to get the rapid change in slope, indicating damage. This was carried out with various delays between the two pulses, and the results obtained are shown in Fig. 3. As shown in the graph, the OBT for the second pulse (probe pulse) is always below the single pulse OBT up to $\sim$12 ps. Also there is a sharp increase in OBT for the second pulse in the delay range between 67 and 200 fs, whereas beyond 200 fs, the OBT is almost a constant.

A simple rate equation to determine the evolution of the free electron density $n(t)$ in a dielectric medium exposed to laser pulses below 10 ps was derived by Stuart *et al.* [3], as given below:

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

where $I(t)$ is the intensity of the laser pulse, $\alpha$ is the avalanche coefficient, and $\sigma_k$ is the $k$-photon absorption cross section with the smallest $k$ satisfying $k\hbar\omega \geq \Delta$, where $\omega$ and $\Delta$ are the laser frequency and the band gap, respectively. Optical breakdown threshold is associated with a threshold electron density $n_{\text{th}}$ and can be predicted by solving the rate equation. In accordance with previous studies [3,4], $n_{\text{th}}$ is chosen to be $10^{21}$ cm$^{-3}$, which is near the plasma critical density for the laser wavelength, $\alpha_{\text{FS}} = (4 \pm 0.6)$ cm$^2$/J and $\sigma_6 = 6 \times 10^{8\pm0.9}$ cm$^{-3}$ ps$^{-1}$ (cm$^2$/TW)$^6$ for FS, $\alpha_{\text{BBS}} = (1.2 \pm 0.4)$ cm$^2$/J and $\sigma_3 = 7 \times 10^{17\pm0.5}$ cm$^{-3}$ ps$^{-1}$ (cm$^2$/TW)$^3$ for BBS [4]. In fact, the plasma energy decay is considered by the theoretical model in Ref. [3]. However, it is believed to last for picoseconds. Thus, for a subpicosecond laser pulse, the decay happens after the pulse and, as such, it is decoupled from the energy deposition to the plasma. As long as enough energy is deposited to the plasma, the breakdown will eventually take place. Hence, Eq. (1) does not include a decay term since this equation is limited to subpicosecond laser pulses only [3]. Consequently, Eq. (1) predicts a constant OBT for the double pulse experiment, which cannot explain our results.

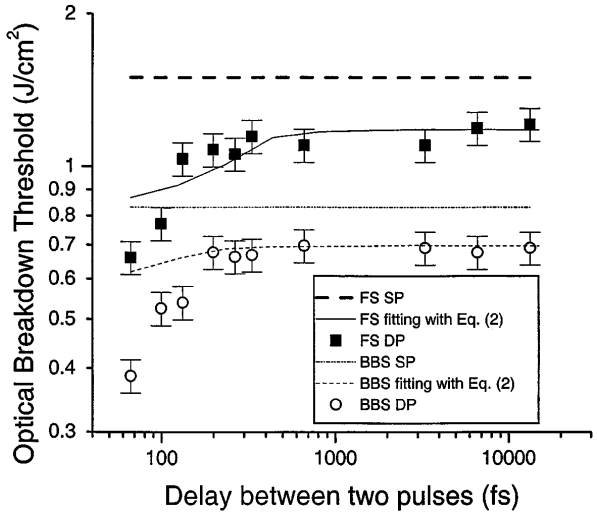


FIG. 3. Experimental OBT vs delay for single pulse (SP) and the probe pulse in the double pulse configuration (DP) along with their rescaled theoretical fits based on the modified rate equation [Eq. (2)] with $\alpha_{\text{FS}} = (4 \pm 0.6)$ cm$^2$/J and $\sigma_6 = 6 \times 10^{8\pm0.9}$ cm$^{-3}$ ps$^{-1}$ (cm$^2$/TW)$^6$ for FS, $\alpha_{\text{BBS}} = (1.2 \pm 0.4)$ cm$^2$/J and $\sigma_3 = 7 \times 10^{17\pm0.5}$ cm$^{-3}$ ps$^{-1}$ (cm$^2$/TW)$^3$ for BBS, and $\tau = 60$ fs.

Our results in Fig. 3 show that in the region between 67–200 fs there is a fast monotonic increase in OBT, whereas from 200 fs onwards the OBT becomes almost a constant. This leads us to believe that the first pulse generates some plasma in the sample, below $n_{\text{th}}$, whose energy or electron density decays with time. The second pulse then interacts with the residual plasma, if there is any, thereby enhancing the avalanche process so that is takes less laser energy for the second pulse to damage the sample. Since the OBT becomes almost a constant after 200 fs, we conclude that the relaxation of the plasma energy is ultrafast and, after 200 fs, the decay is complete. For a 100 fs or shorter pulse, the decay process is actually coupled with MPI and avalanche. Motivated by this observation, we decided to add an exponential decay term to Eq. (1) to fit our experimental data, as shown in Fig. 3,

$$\frac{dn(t)}{dt} = \alpha I(t) n(t) + \sigma_k I(t)^k - \frac{n(t)}{\tau} . \qquad (2)$$

However, our data do not fit an exponential decay curve exactly. The decay time constant $\tau$ is found to be approximately 60 fs from the best fit. Thus, the decay process is ultrafast and cannot be neglected for the 100 fs regime. Although the new rate equation does not fit the experimental data very well and is not a rigorous model, we can still explore the impact of including such a decay term on interpreting other single pulse experiments. Further theoretical modeling of such a decay process needs to be done in the future.

For the same values of $\alpha$ and $\sigma_k$ as in Eq. (1), the single pulse OBT's obtained from Eq. (2) do not agree with the experimental results of Lenzner *et al.* [4]. The modified $\alpha$ and $\sigma_k$ values, which better fit the data of Ref. [4], are $\alpha_{\text{FS}} = 9.0$ cm$^2$/J and $\sigma_6 = 3.0 \times 10^4$ cm$^{-3}$ ps$^{-1}$ (cm$^2$/TW)$^6$ for FS, $\alpha_{\text{BBS}} = 6$ cm$^2$/J and $\sigma_3 = 3 \times 10^{16}$ cm$^{-3}$ ps$^{-1}$ (cm$^2$/TW)$^3$ for BBS (Fig. 4). All of these values are more than a factor of 2 different from the previously determined values [4].

If the delay between the two pulses is large enough, the second pulse OBT should converge to the single pulse OBT. However, Fig. 3 shows that, even at picosecond delays, the second pulse OBT is still below the single pulse OBT. Our explanation is that the OBT is a function

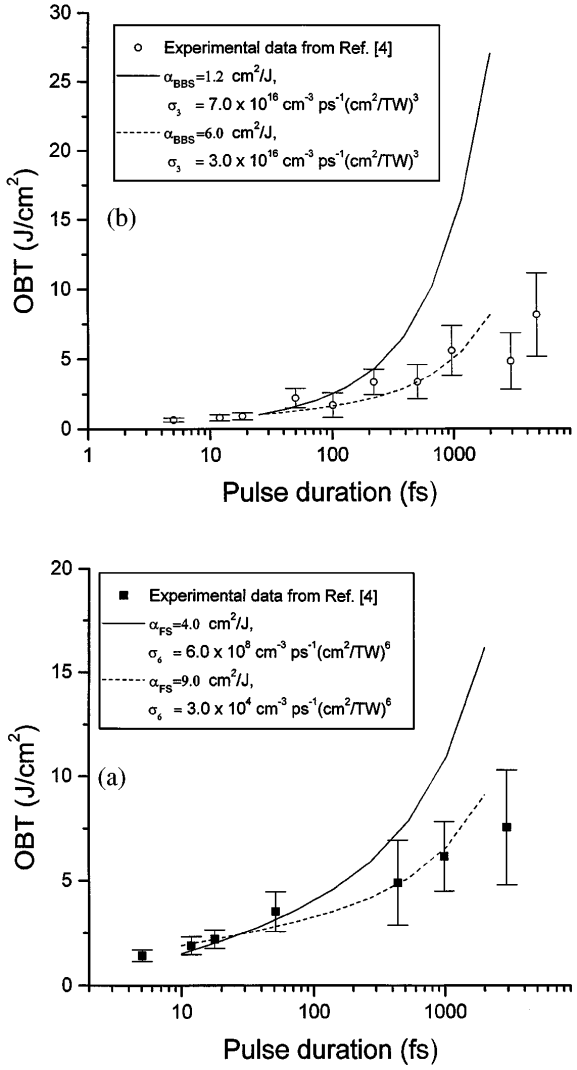


FIG. 4. Fits to previous pulse-duration data [4] with the new model, which includes decay [Eq. (2)]. (a) FS and (b) BBS.

of temperature. After the appearance of the first pulse, the plasma temperature increases and the OBT thus decreases. In order to verify this argument, an experiment was carried out to extend the delay into the nanosecond range, when the thermal equilibrium should be reached. The results indicate that the second pulse OBT does indeed approach the single pulse OBT, as shown in Fig. 5. Since our ASE has a pulse duration ~300 ns, our large time delay data are not affected by it. Hence, we conclude that the OBT is a function of temperature and is lower at higher temperatures.

We have developed a well-defined criterion to determine the OBT in real time. Our results prove that the double pulse technique is indeed very effective in probing the temporal behavior of OBT. We have found that the

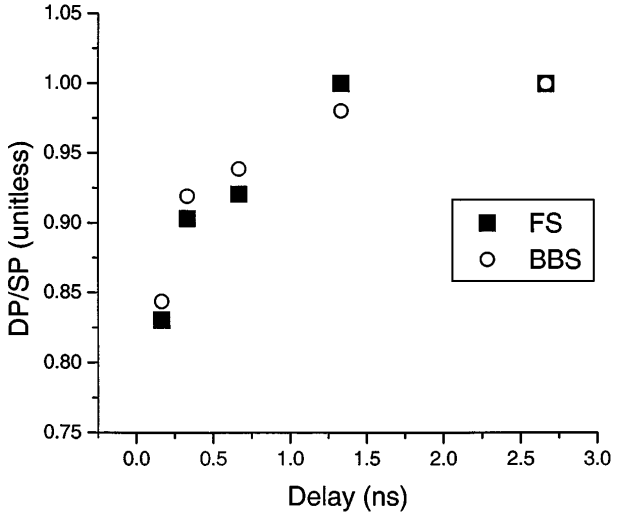


FIG. 5. The OBT for the probe pulse in the double pulse configuration (DP) approaches that of the single pulse (SP) in the nanosecond delay range. The ratio between DP and SP reaches 100% for 2.5 ns delay.

decay of energy takes place within the pulse duration, and this changes the whole picture of the breakdown process. We also notice that there is no signature of decay in the picosecond range, which indicates that the decay process is not due to electron-phonon coupling. However, the observation of decay in the 100 fs range suggests that this could be an electron-electron collision-induced recombination. Even though we have remodeled the existing formula for electron density decay, more theoretical work is needed in this area. We have also noticed that the decay process does not follow an exponential curve. In addition, inclusion of the decay term significantly changes the multiphoton and avalanche rate coefficients.

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