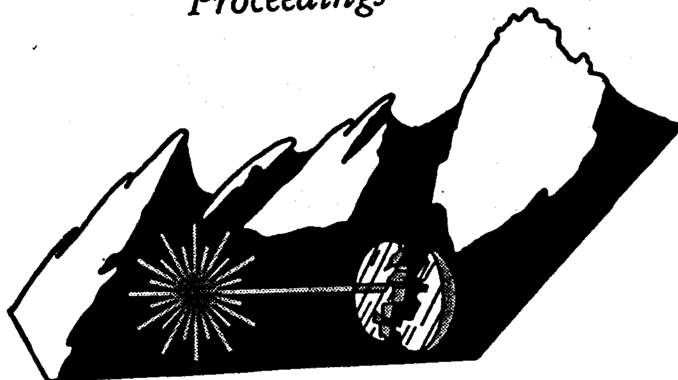
26th ANNUAL BOULDER DAMAGE SYMPOSIUM
Proceedings



LASER-INDUCED DAMAGE IN OPTICAL MATERIALS: 1994

24–26 OCTOBER 1994
BOULDER, COLORADO

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Behavior of laser induced emission intensity versus laser power density during breakdown of optical materials

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ABSTRACT

Breakdown in SiO_2 is studied versus fluence using an intensified CCD spectrometer. Broad-band photoluminescence spectra were measured versus number of laser pulses. Before the breakdown of fused silica, the intensity of this photoluminescence increases. After breakdown, a plasma is formed and ablated Si emission lines are measured. The plasma is characterized by its emission spectra and excitation temperature temporal profiles. The temperature profiles of the plasma are calculated by the Bolzmann method. These data are studied to provide fundamental information on breakdown mechanisms in optical materials.

Keywords: Laser ablation, optical materials, fused silica, damage threshold, excitation temperature, photoluminescence, emission spectra

1. INTRODUCTION

At high laser fluence, lower than the breakdown of a material, strong optical absorption occurs in otherwise non-aborbing media, leading to physical damage, ejection of material into a vapor plume, and creation of an ionized plasma. The actual absorption mechanisms under high power, short pulse irradiation are not completely defined. Absorption of the laser energy in wide band-gap materials is often dominated by lattice defects[1-6]. Laser damage in these materials can be strongly enhanced by defects produced by the laser itself (such E' center in SiO^2)[7,8]. When such optical materials are exposed to a series of laser pulses, the surface will damage eventually, even when the laser power density is lower than the single shot damage threshold. The laser induces microscopic structural changes in the material ultimately lead to surface damage.

In this report, photoluminescence of fused silica (SiO_2) and silicon emission line were measured versus number of laser pulses, below, at, and above the optical damage threshold. The peak intensity of photoluminescence increases even before physical damage can be observed, supporting an accumulation of defects type mechanism. When the laser power density is higher than the breakdown threshold, emission characteristics of the ejected atoms may provide fundamental information on the laser material interaction mechanisms. We measured the intensity of silicon emission versus time and calculated the excitation temperature development in the plume.

2. EXPERIMENTAL

A diagram of the experimental setup is shown in figure 1. A pulsed KrF excimer laser (Questek model 2680) is used as the optical source. The 248 nm UV beam is directed into the sample using two

mirrors, and focused onto the sample surface using a plano-convex UV grade quartz lens with a 20 cm focal length. This lens is mounted on a micrometer translation stage so that focusing (fluence) can be finely adjusted. The pulse energy is monitored by a power meter. The pulse duration is 30 ns and the repetition rate is 5 Hz. An aperture of 6 mm diameter is placed between the excimer laser and the focusing lens to vary the laser beam spot size and energy at target surface, and to utilize the more homogeneous portion of the excimer spatial power profile.

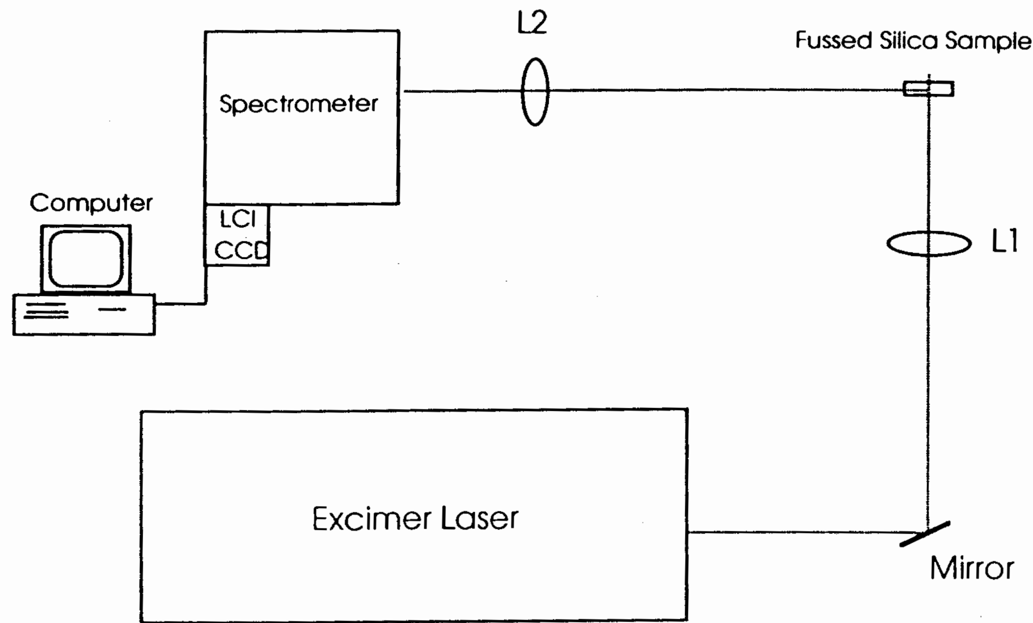

Figure 1. Diagram of experiment setup for time resolved emission studies in optical materials.

The SiO_2 samples are mounted on a hanger such that the transmitted laser beam does not strike any underlying surfaces. The samples are always placed before the effective focus of the lens. The fused silica (SiO_2) samples are 1 mm in thickness.

Photoluminescence and radiation from the plasma are observed at a right angle to the laser beam with a quartz lens (F= 120 mm), in order to get an image onto the vertical entrance slit of a Cezrny-Turner spectrometer (Spex Industries Model 270M with a 1200 groves/mm grating). The spectrometer slit height is set at 10 mm. The slit width is 25 mm. The spectrometer is equipped with a thermoelectrically cooled, charge coupled device (CCD) (EG & G Princeton Applied Research Model OMA VISION) with a lens coupled intensifier (LCI) system. The system was controlled by OMA SPEC 4000 software (EG & G Princeton Applied Research). This CCD spectrometer system is able to view a 35 nm wavelength region or 150 nm wavelength region by changing the grating in the spectrometer. A mercury lamp is used for wavelength calibration. With the LCI, the optical emission can be time resolved. Two types of experiments were conducted. The first involves using a fixed delay time and gate width, to observe the spectral changes versus number of laser pulses. The second uses a fixed gate width, and increase the delay time to observe the development of laser induced emission.

The fluence of the laser beam on the SiO_2 is changed by varying the distance of the focusing lens, with a fixed laser energy. Fluence at the sample surface is estimated from the energy of the laser beam,

and the calculated spot area. The spot area is calculated using simple geometric optical principles and compared to burn patterns.

3.RESULTS AND DISCUSSION

3.1 Photoluminescence versus number of laser pulses

Pure fused silica exhibits fluorescence or photoluminescence when exposed to UV radiation. The photoluminescence spectra measured after exposure to a different number of laser pulses is shown in Figure 2. The spectra are recorded starting at the laser pulse, and for a duration of 120 ns (gate time). The fluence is 12 J/cm^2. Before physical damage (<200 pulses), there is only one broad emission peak, centered at 288 nm. The intensity of this photoluminescence increases with number of pluses, until about 340 pulses, at which time another peak at ≈ 387 nm appears. Both peaks further increase with additional laser pulses. Figure 3 shows how the intensity of two peaks (288 nm and 387 nm) changes with number of laser pulses. Before 250 pulses, the intensity of the photoluminescence peak at 288 nm remains essentially constant and then increases. After ≈ 325 pulses a large increase in emission and damage are observed. The intensity of the 387 nm line only increases slightly with successive pulses, until ≈ 340 pulses. The 288 nm intensity decreases and the 387 nm intensity dramatically increases. Severe physical damage is observed when the 387 nm line emission exist.

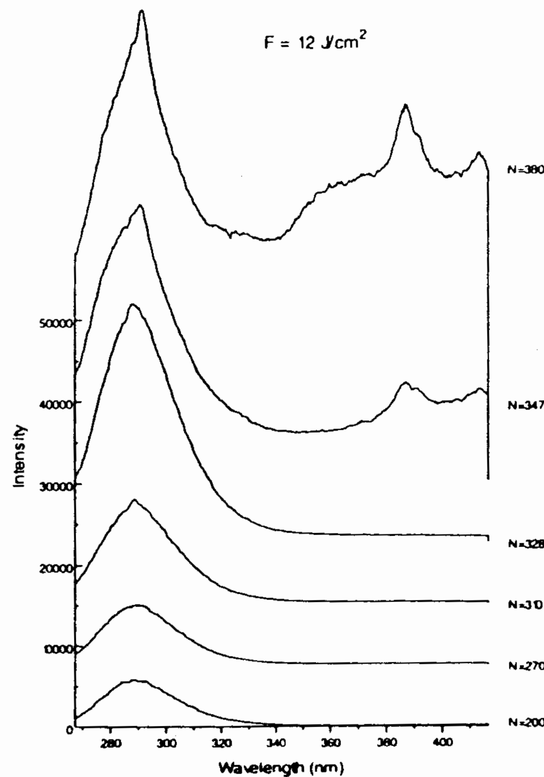

Figure 2. Photoluminescence spectra versus number of laser pulses. The delay time is 0 ns, and gate width is 120 ns.

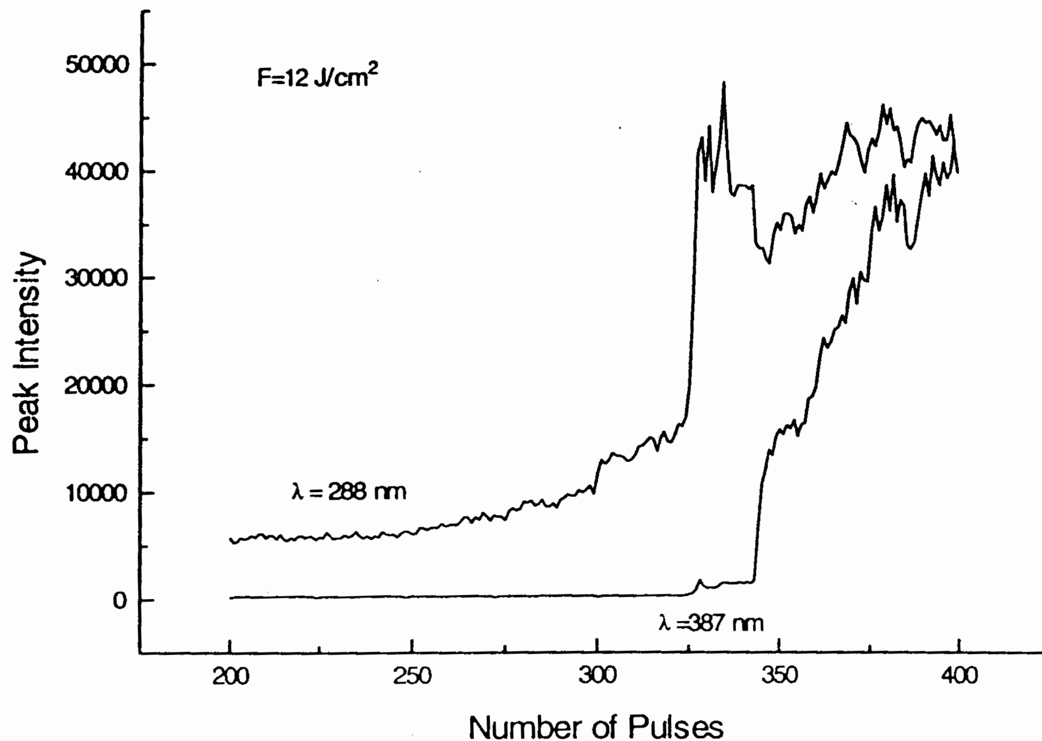

Figure 3. The peak emission intensity at 288 nm and 387 nm versus number of laser pulses. The delay time is 0 ns, and gate width is 120 ns.

The lifetime of the photoluminescence was measured to be about 120 ns which is using the gated intensified CCD system. It is interesting to measure the spectra after 120 ns, after the photoluminescence decay. Figure 4 show the spectra taken after 120 ns with a gate width of 40 ns, for a different number of laser pulses. For number of laser pulses (N) =110, there is only one weak broad peak at 328 nm over this 270 nm to 420 nm range. After ≈ 240 pulses, the spectra show a strong narrow Si emission line at 288 nm and several other broad peaks. The high intensity of these lines completely overwhelms the observation of emission at 328 nm region. Figure 5 shows the peak intensity at 288 nm, 328, nm, and 387 nm versus number of pulses. These data represent the induced damage from another spot in this fused silica, using the same laser fluence (12 J/cm^2). The breakdown at ≈ 240 pulses is different than observed in Figure 3; it is not surprising considering that the mechanisms for damage are highly dependent on a number of properties, including the local microstructure of materials. After surface damage, the peak intensity of 288nm and 387 nm is similar to the peak intensity during 0 to 120 ns in Figure 3.

From the above experimental results, it is clear that the photoluminescence intensity of SiO_2 increases after a number of pulses and before physical damage occurs. These data confirm that defects in the fused silica accumulate as the sample is exposed to a series of UV laser pulses. The laser induces microscopic structural changes in the material that ultimately lead to surface damage.

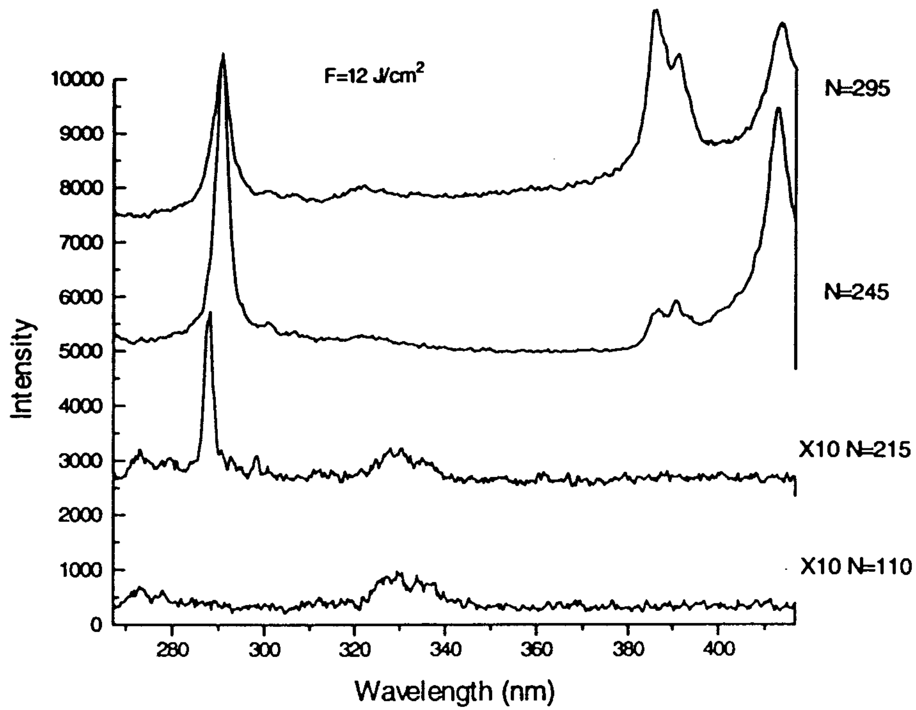

Figure 4. Photoluminescence spectra versus number of laser pulses. The delay time is 120 ns, and gate width is 40 ns.

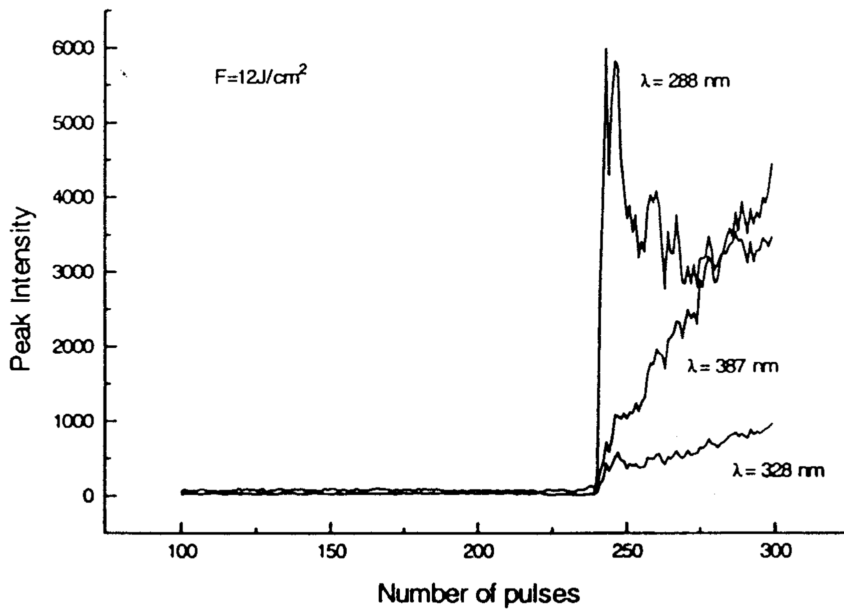

Figure 5. The peak emission intensity at 288 nm, 328 nm, and 387 nm versus number of laser pulses. The delay time is 120 ns, and gate width is 40 ns.

temperature increases during the first 500 ns then decrease and exists out to 3 μs. The maximum excitation temperature reaches 23,000 K.

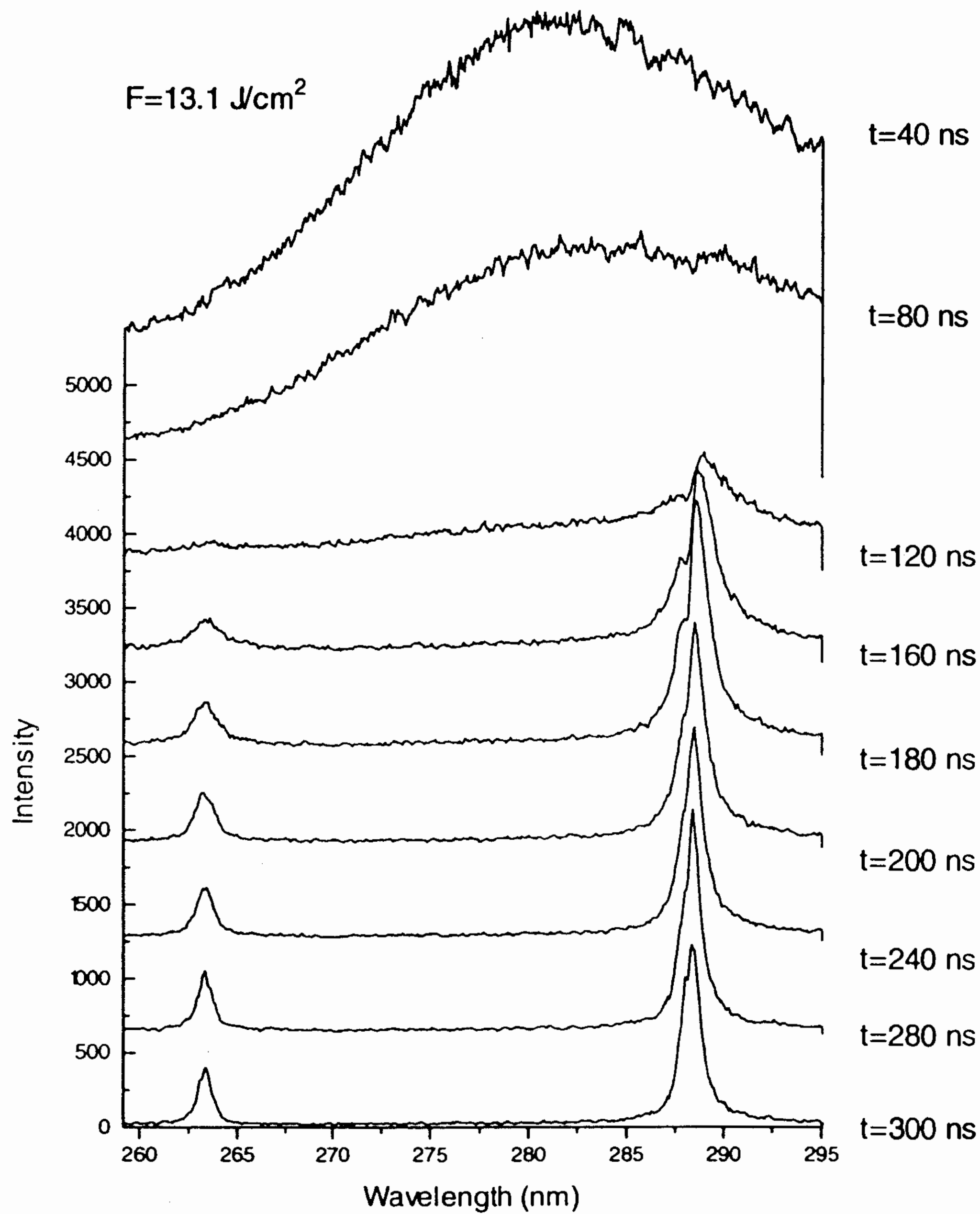

Figure7. Time resolved spectra of silicon emission at $F=13.1$ J/cm^2. Gate width is 40 ns.

3.2 Excitation temperature in plume

Time resolved spectra of silicon emission at 263 nm and 288 nm were measured at different laser power densities (see Figure 6-8). The data in these figure were taken for different laser pulses and at different delay times for each laser pulse. The gate width is 40 ns. The photoluminescence at a fluence of 11.5 J/cm^2 is shown in Figure 6. The photoluminescence exists from 0 ns to about 120 ns. After 120 ns, there is only the normal background noise signal. For fluence of 13.1 J/cm^2, photoluminescence exists during the first 120 ns (see Figure 7); however, after 120 ns, two Si atomic emission lines appear, at 263 nm and 288 nm. At 288 nm, self absorption is observed during the first 300 ns. For fluence ≈ 25 J/cm^2 the emission behavior is similar to the fluence at 13 J/cm^2 (see Figure 8), except the intensity is greater.

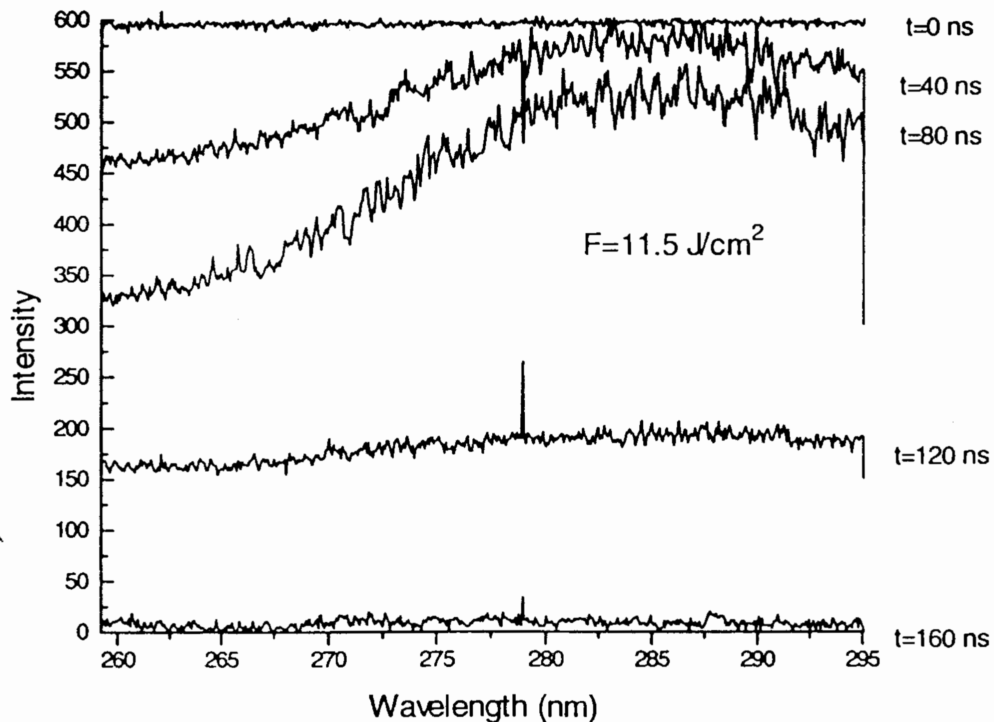

Figure 6. Time resolved spectra of silicon emission at F=11.5 J/cm^2. Gate width is 40 ns.

Temperature of the escaping atoms may be an important property to describe the breakdown damage mechanisms. The excitation temperature of Si atoms in the laser-induced breakdown plasma is calculated using the Bolztmann two-line method, based on the assumption that the Bolztmann distribution is fulfilled during the plasma region. The excitation temperature can be calculated using the 263.13 nm and 288.16 nm atomic Si lines. The silicon 263nm and 288nm line have upper energy of 53387 cm^{-1} and 40992 cm^{-1}, respectively. The degenerancies and Einstein coefficients were taken from the CRC handbook of chemistry and physics[9]. A good signal to noise ratio for the Si line emission intensity is necessary for the excitation temperature calculations. From Figure 8, the background signal is very low after 300 ns delay time. The temperature versus delay time is shown in Figure 9. The

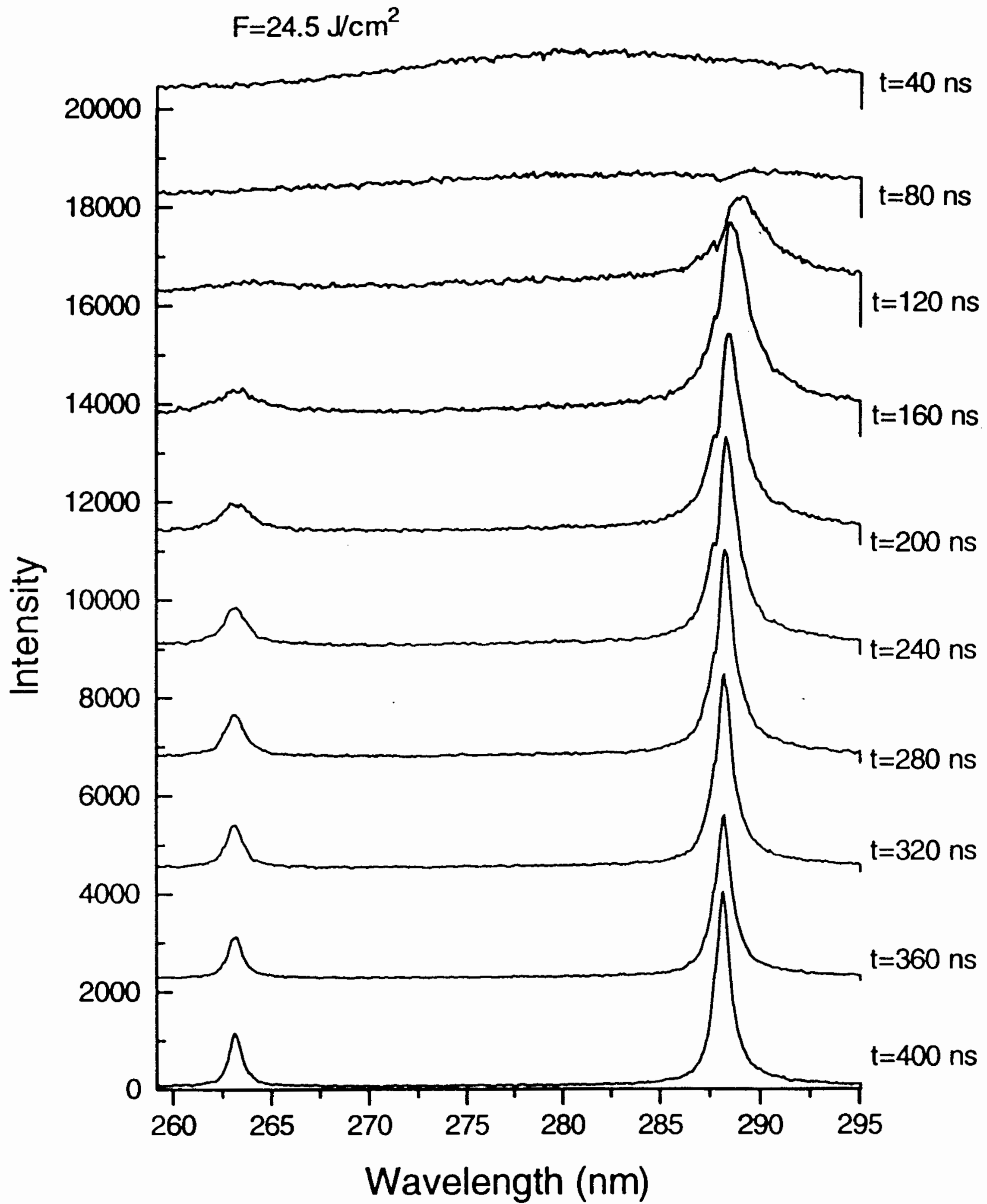

Figure 8. Time resolved spectra of silicon emission at F=24.4 J/cm^2. Gate width is 40 ns.

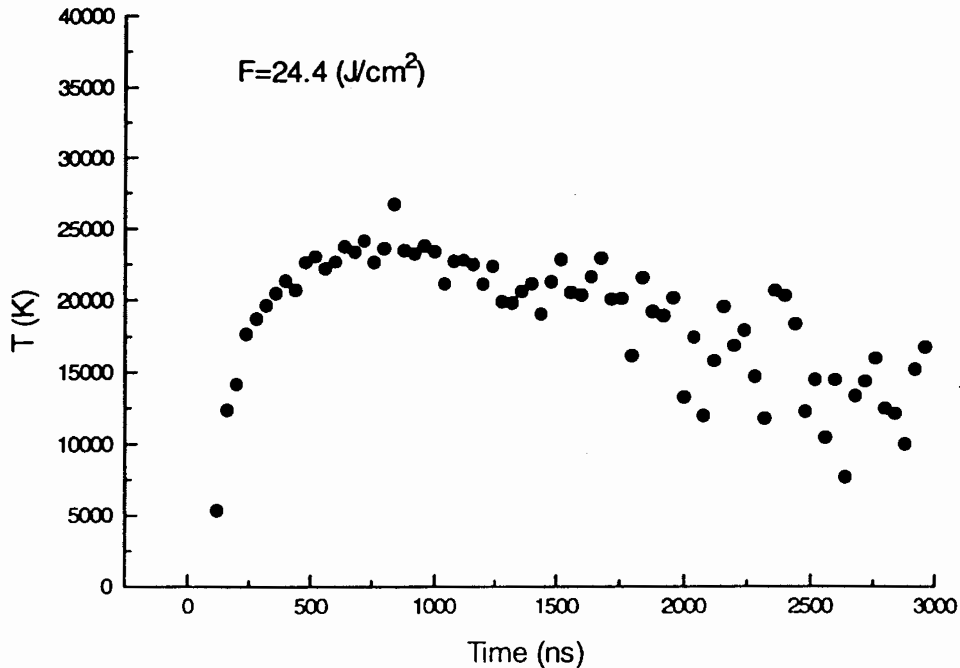

Figure 9. The Si excitation temperature as function of delay time.

4. CONCLUSION

Photoluminescence spectra and silicon emission line intensity were measured during laser irradiation and breakdown of SiO_2. Before breakdown, the intensity of photoluminescence was found to increase. After breakdown, photoluminescence and atomic emission from ejected Si are observed. A time resolved excitation temperature for Si in the plume was measured. The temperature can reach 23,000 K.

5.Acknowledgments

This research was supported by U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences, under contract #DE-AC03-76SF00098.

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