Comment on "Ultrafast Electron Dynamics in Femtosecond Optical Breakdown of Dielectrics"

The authors of a recent Letter [1] report new experiments on optical breakdown in which they measure the optical breakdown threshold (OBT) as a function of the intensity of an ultrashort (25 fs) Ti:sapphire laser pulse by observing the breakdown plasma fluorescence. They also measure the OBT for a set of two laser pulses, the second one having an intensity 30% higher than the first one, as a function of the delay between the two pulses. This delay was adjusted between 67 fs and >10 ps. They intended to test, in this way, a widely accepted model [2] in which laser breakdown occurs because of "avalanche" injection of free carriers in the conduction band. Here the first pulse injects a small density of "seeding" free carriers, and the second pulse causes laser heating and thus the avalanche breakdown. The result they obtain, which cannot be accounted for by the model, is that the OBT for the two pulses increases up to a delay of about 200 fs, saturates at a level significantly lower than the single pulse OBT, and stays fixed for more than 10 ps. To interpret their data, they add to the kinetic equations of [2] a population damping term whose origin they do not explain, and assign the saturation of the OBT to an effect of the sample temperature.

We would like to point out that this behavior can be due to self-trapping of electron hole pairs under the form of transient point defects known as self-trapped excitons (STE) [3]. In [4,5] we showed that free carriers in SiO_2 have a very short lifetime: A mean "trapping time" of 150 fs was measured, which is in almost perfect agreement with this new measurement. The kinetic equations used to fit the data in this experiment was identical to the one used by Li *et al.* [1], except that no avalanche term was included, since we worked at intensities well below the OBT. The STE was identified as the trapping center by transient absorption measurements [6]. This trapping mechanism has a very high efficiency: Practically all of the e-h pairs created by the first laser pulse will self-trap. It is remarkable that, in spite of an abundant literature (the case of alkali halides has been thoroughly investigated [7]), such processes have never been considered in the framework of laser breakdown studies.

This self-trapping mechanism is accompanied by a lattice relaxation which stabilizes the e-h pair, to which corresponds a metastable defectlike state in the band gap. Thus the electrons created by the first pulse are still available for the second pulse, and they are more weakly bound than valence band electrons. It is then not surprising that the two pulse experiment yields a smaller OBT since the second pulse will collect all of the electrons excited by the first pulse in addition to those it will inject directly from the valence band. We note that the data suggests that the essential mechanism for laser breakdown is not the avalanche mechanism since, in such a case, the OBT would not depend very much on the number of initially injected carriers (because of the exponential nature of the avalanche process, it is essentially the "heating rate" of the free carriers that matters, that is to say, here the second pulse intensity).

Concerning the "recovery" of the single pulse OBT, we note that the STE is a transient state precursor of the E_1' center, a colored center in SiO_2. In Ref. [6] we showed that only a small fraction on the STE actually turns into E_1' centers: about 10^{-3}, at room temperature, which readily explains the OBT recovery, since most of the STE recombine nonradiatively. At low temperatures (below 150 K) the recombination mechanism is luminescence. In Ref. [8], the luminescence quenching time was shown to decrease rapidly above 150 K from the low-T lifetime of 1 ms to about 10 ns at 250 K. The recovery time measured in this experiment seems quite consistent with such figures.

We thus conclude that the fast decay process invoked in Li *et al.* is well identified. With the help of this self-trapping mechanism, we can explain all of the observations reported in this paper, and moreover we find that the orders of magnitude we anticipate from this explanation are consistent with the measurements presented by Li *et al.* It is thus critical to incorporate such mechanisms in the modeling of optical material under high laser excitation: First, there are mechanisms related to exciton self-trapping, such as significant transient volume increases (which could launch shock waves in the material given the high matter accelerations involved) which have never been considered. Second, this is the dominating mechanism in the free carrier's kinetics (at least in SiO_2), implying time scales that are of the order of or below the typical pulse durations used in high intensity physics, so that optical breakdown models which simply disregard it cannot lead to convincing conclusions.

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Received 20 April 1999
PACS numbers: 78.47.+p, 42.50.Ct, 42.62.Cf

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 0031-9007/99/83(24)/5182(1)$15.00