Avalanche ionization and dielectric breakdown in silicon with ultrafast laser pulses

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Experimental evidence is presented demonstrating avalanche ionization as the dominant mechanism for dielectric breakdown in silicon with ultrafast laser pulses at above-gap photon energies. Data are presented for pulses between 80 fs and 9 ns at 786 nm and 1.06 μm. Associated electric fields range from 0.3 to 40 MV/cm. Avalanche ionization coefficients range from 10^{10} to 10^{14} s^{-1} and are discussed in relation to semiempirical dc ionization theory and recent ac Monte Carlo calculations. Correlation is obtained between electron collision times and associated ionization rates. [S0163-1829(98)03129-4]

In earlier work on laser-induced dielectric breakdown of SiO_2, it was observed that avalanche ionization was a major factor in interpreting the results of the experiment.[1] In that work, a set of ionization coefficients was experimentally determined and compared to the theory of Thornber for a dc-like response of the material. In the present paper we continue that line of investigation by applying the technique to the semiconductor silicon, where a considerable body of knowledge exists on its ac and dc breakdown behavior. These properties are examined in relation to dielectric breakdown at optical laser frequencies in the near infrared. An important distinction is that, in the SiO_2 case, multiphoton absorption processes seed the avalanche process, whereas, in the present case with Si, a single-photon absorption predominates. In a previous publication,[2] we reported on the observation of a plasmalike absorption phenomena occurring at the damage threshold in silicon for laser pulses ranging from 80 fs to 7 ns with 800-nm optical radiation. The process was interpretated, based on measured absorption depths and observed breakdown electric-field strengths, as being associated with the absorption of the laser pulse in an ac avalanche-initiated plasma. It was further determined in that work, based on a thermophysical plasma heating model, that the optical radiation was essentially captured in an averaged 1/*e* depth of 75 nm as a consequence of the operative plasma density.

As explained by Vaidyanathan *et al.*,[3] the damage threshold (fluence and field strength) will decrease with increasing laser wavelength for avalanche breakdown, whereas it rapidly increases for the multiphoton process. Therefore, the behavior of the breakdown threshold, as a function of laser wavelength, is a definitive demonstration of whether avalanche or multiphoton processes dominate. Since our optical radiation (1.55 eV) was above the bandgap of silicon (1.12 eV) in the work of Ref. 2, it is conceivable that band-to-band transitions could have played a role in developing the plasma. In the present work we provide a demonstration, through examination of breakdown as a function of laser wavelength, that avalanche ionization rather than band-to-band transitions, is responsible for the plasma condition and the observed damage threshold behavior.

By way of background, it had been shown in earlier work by Reitze *et al.*[4] that laser absorption processes, resulting from above-gap radiation in silicon (620 nm, 90 fs), can be initially explained by a one- and then two-photon absorption mechanism as a function of moderate laser intensity. At higher intensities they found that higher-order multiphoton absorption processes could no longer be invoked and that a new, and at that time, unexplained nonlinear process needed to be considered. We present in this paper an explanation of that observation in terms of the onset of an avalanche ionization process and provide experimental results with comparison to theory for justification of our interpretation. It is important to note, in comparing our work with Reitze *et al.*, that they held the pulse duration constant at 90 fs and varied the laser intensity, whereas in our work the pulse duration is varied and the laser intensity adjusted to provide a threshold for surface breakdown. In addition we use somewhat longer wavelength radiation. The essential comparison however remains valid, since results are presented and compared as a function of laser electric fields at the surface of the sample.

Presented in Fig. 1(a) are the observed laser fluence breakdown values for silicon as a function of the laser pulse duration for 786-nm and 1.06-μm radiation with comparison to thermophysical model calculations for 786 nm. These data are obtained by varying the laser pulse duration and in each case adjusting the intensity to achieve single-shot breakdown and surface damage.[2] In Fig. 1(b) are presented the associated electric-field values for breakdown as a function of pulse duration. Under these circumstances the breakdown state of the material defines the common condition for comparing field strengths between different pulse durations. The electric field is obtained from the laser intensity through the (time-averaged) relationship $I=(c/8\pi)(\varepsilon/\mu)^{1/2}E^2$ (in cgs). Inverting results in the electric field as $E=(27.4)I^{1/2}$, where I is the laser energy density, in J/cm^2, divided by the pulse duration in seconds. The breakdown event, as defined here, is always associated with the appearance of a faint optical emission spark at the center of the laser spot on the surface of the material. Atomic-force microscopy confirmed the presence of a damaged surface precisely at the point when such an optical emission spark occurs. Ellipsometry and cross-sectional TEM demonstrated no observable material changes for intensities at levels immediately below that which provided the emission spark. This condition was deemed an appropriate manifestation of irreversible surface

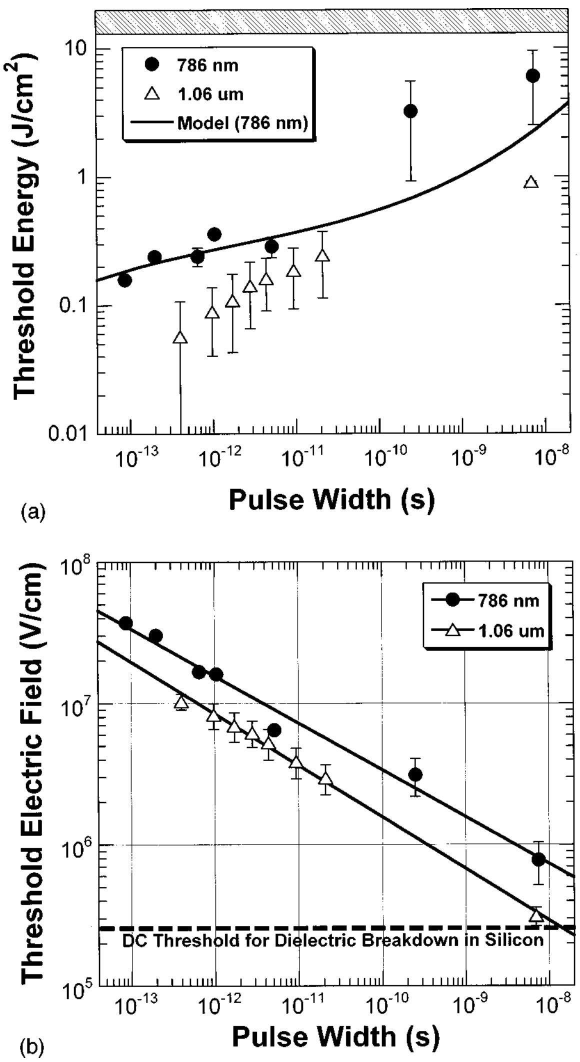

FIG. 1. (a) Energy fluence dependence of damage threshold as a function of pulse duration. The shaded region at the top of the figure indicates the zone in which damage threshold would occur for deep (7 μm) linear absorption. The model calculation is from Ref. 2 and represents a 75-nm absorption depth which is fitted to the 786-nm data. (b) Breakdown electric field as a function of laser pulse duration for 786-nm and 1.06-μm laser radiation.

change associated with plasma breakdown and initiation of surface evaporation damage from electron-lattice heating.

It is seen in Figs. 1(a) and 1(b) that the breakdown fluence and field strengths for 1.06-μm radiation lie consistently below that of 786 nm and that all breakdown field strengths are well above the value for dc breakdown of silicon, 0.25 MV/cm.[5] Error bars are shown relative to measurements made by different individuals in our research team. The threshold values for breakdown fluence are of course critically dependent on the beam spot size and associated irradiated area. These spot sizes were measured using both a calibrated line out from a CCD camera image and an integrated beam intensity measurement using a micropositioned blade edge that is moved across the focal spot. In both cases, the beam diameter is determined from the $1/e^2$ intensity value and the average fluence calculated as $E/\pi r^2$ where r is the $1/e^2$ value and E the laser energy.

In our earlier work on silicon,[2] we noted that the breakdown field strengths are associated with very short absorption depths in the material, 75 nm on average. Linear absorption in silicon at these wavelengths will occur over much larger depths, typically 7 to 8 μm or more, and two-photon absorption can be shown to occur over a 3-μm depth.[4] The observed shallow absorption depths are therefore interpreted as the result of the laser pulses interacting with a dense breakdown plasma followed by a subsequent thermophysical response of the underlying material through electron-ion and plasma heating. Electron-lattice heating is a well-established process in dielectric breakdown and shown to be an essential part of solid-state avalanche processes.[6–8]

In further support of the avalanche process, if linear absorption were operating, the longer wavelength radiation (1.06 μm) would have a larger optical-absorption depth[5,9] than the shorter (786 nm) and therefore would require a higher damaging threshold fluence, the opposite of what is shown in Fig. 1(a). The reversal of surface damage threshold as a function of wavelength is compelling proof that the process is a nonlinear avalanchelike mechanism rather than a linear or multiphoton[10] band-to-band transition mechanism. It will be shown that this conclusion is further supported by recent Monte Carlo calculations[11] showing longer wavelength radiation, in the near IR, as being more efficient in producing an avalanche process than shorter wavelengths. Lower breakdown fields will therefore result at longer wavelengths as is demonstrated in Fig. 1(b). Thus, the results of Fig. 1(b) establish credence to an avalanche process, both in terms of field strengths and their dependence on wavelength as predicted by theory.[3]

The effective ionization coefficient for the avalanche process can be computed from the simple electron multiplication equation $N=N_0\exp(\alpha t)$ where N_0 is the conduction-electron concentration at the start of the process (i.e., the extrinsic doping level in silicon), t is the time over which effective and sustained multiplication occurs, and α is the ionization (rate) coefficient. This is a simplification of the more complete form given by Niemz:[12] $N(t)=[\exp(\alpha t)]/[(1/N_0)+(\gamma/\alpha)\{\exp(\alpha t)-1\}]$, where α is the effective avalanche generation rate and γ is the inelastic collision loss rate. For the sake of accuracy, α is further given as $\beta-\delta$, where β is the actual ionization rate and δ the rate at which electrons diffuse out of the active zone. Experimentally we extract the effective generation rate α. An important electron density is the plasma critical density: $N_c=\omega m/4\pi e^2$, where ω is the laser angular frequency, m is the electron mass, and e is the electron charge. At critical density, the plasma frequency equals the laser frequency which gives $N_c=1.7\times10^{21}\ \mathrm{cm}^{-3}$ for 780-nm radiation. Higher electron concentrations introduce an out-of-phase condition that inhibits further absorption, and lower electron densities dilute the plasma allowing the laser radiation to penetrate more deeply with less efficient absorption. In both cases absorption will still occur but with greatest efficiency near this critical density. For simplicity then, we can set the electron concentration to the critical value and extract ionization parameters from the above equations, and compare them to predicted values from ac and dc theories.

In support of using the simplified form of the electron multiplication equation, it is noted that when using the full form of the avalanche equation to extract effective ionization

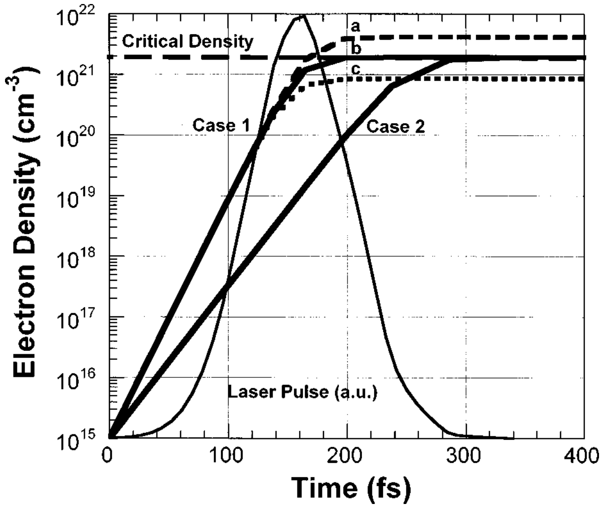

FIG. 2. Example of avalanche-induced electron number density as a function of time, showing the effect of the inelastic collisional loss coefficient on the achievement of plasma critical density (per discussion in text). The laser pulse is 100 fs fwhm.

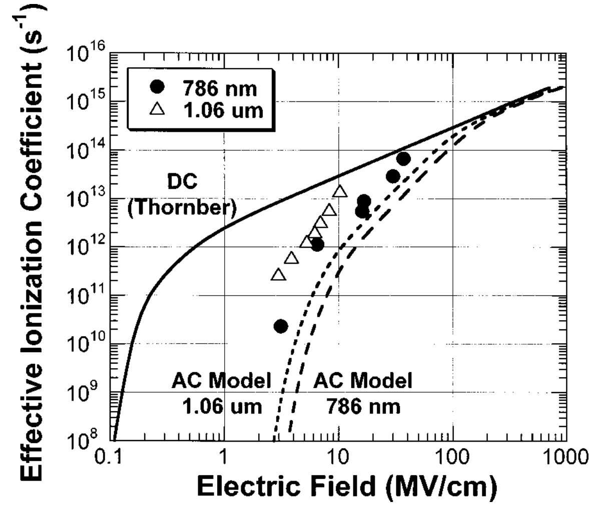

FIG. 3. Experimental and theoretical (Monte Carlo) ac ionization coefficients as a function of electric field and comparison to the semiempirical dc results of Thornber for silicon.

coefficients it is found that the inelastic collision term γ is a dominating factor only in determining at what level the maximum electron concentration will occur. This is shown by way of example in Fig. 2 where the electron concentration is plotted as a function of time for a 100 fs full width at half maximum (fwhm) pulse using the complete form of the avalanche equation. Two cases are shown. In case 1, the integration was carried out for a constant β value of 9.1×10^{13} s^{-1} and for three values of γ (2.2×10^{-8} $\mathrm{s}^{-1}\,\mathrm{cm}^3$, 4.7×10^{-8} $\mathrm{s}^{-1}\,\mathrm{cm}^3$, and 1.1×10^{-7} $\mathrm{s}^{-1}\,\mathrm{cm}^3$ for subcases a, b, and c, respectively) resulting in saturation at the pulse peak. Case 2 uses $\beta=5.8\times10^{13}$ s^{-1} where saturation occurs near the end of the pulse (2.5 fwhm). It is determined that, in order to achieve critical density, the value of γ must be at most 4.7×10^{-8} $\mathrm{s}^{-1}\,\mathrm{cm}^3$ in case 1 and 3.0×10^{-8} $\mathrm{s}^{-1}\,\mathrm{cm}^3$ in case 2. The primary role played by this inelastic collision term is in determining the electron saturation level, whereas the buildup and saturation time depends almost solely on α. Since we are using critical density as our defined absorption level, the γ term in the avalanche equation can effectively be dropped since its contribution is predetermined by the choice of critical electron density as the electron saturation level.

The electron diffusion term contained in $\alpha=\beta-\delta$ likewise makes a minimal contribution to the effective ionization coefficient, since typically $1/\delta$ is on the order of 500 ps and δ will therefore be on the order of 2×10^{9} s^{-1}. Ionization coefficients β will generally be much larger than this (for relevant electric fields) and so the quantity α, which is the effective value extracted from experiment, will be very close to the actual value of β for pulse durations less than approximately 300 ps. Further, in regard to the origin of electrons, the samples we are using in the present experiments have an extrinsic conductivity of 10–30 $\Omega\,\mathrm{cm}$ representing an initial electron reservoir N_0 of 1×10^{15} cm^{-3}. The presence of these extrinsic carriers eliminates the need for identifying the origin of the initial avalanche electrons.

Application of the simplified avalanche equation to the data of Fig. 1(b) yields a computed ionization coefficient α that is given by $\alpha=(I/t_{\mathrm{crit}})\ln(N_c/N_0)$, where t_{crit} is that portion of the pulse duration needed to achieve a critical plasma density at a specific breakdown field. The value of t_{crit} was chosen as the time over which the laser electric field is high enough to produce ac breakdown, with $t_{\mathrm{crit}}=2.5\tau_{\mathrm{fwhm}}$ being an average for the pulses analyzed here. Using this criteria for t_{crit} produces the results in Fig. 3 for pulses ranging from 80 fs to 300 ps. The computed ionization coefficients are compared with the Thornber dc semiempirical theory[13] and to ac Monte Carlo results[11] for the conditions of our experiment. It is seen in Fig. 3 that the experimental ionization coefficients range from 10^{10} to 10^{14} s^{-1}, depending on field strength, and are in general agreement with the Monte Carlo theory for ac avalanche at these optical frequencies and that they tend towards the dc Thornber value at the highest fields associated with the shortest laser pulses.

Bloembergen and Yablanovich have used the Drude theory of ac conductivity in metals to arrive at a simplified expression for the ac electric field breakdown strength of a material in terms of its dc breakdown field.[14] This is given as $E(\omega)=E_0(1+\omega^2\tau_{\mathrm{eff}}^2)^{1/2}$, where ω is the laser angular frequency and τ_{eff} is the mean time between collisions of the electrons with the atoms of the material. For a fixed value of α in Fig. 3, we can extract a dc and ac breakdown field, and then calculate τ_{eff} using this equation. Doing this for the two experimental wavelengths results in extracted collision times from 5.0 to 0.5 fs, as presented in Fig. 4. These are compared, in the same figure, to the experimental and Monte Carlo ionization coefficients as a function of field strength. It is seen that in all cases the collision times are less than the ionization times with the difference becoming less significant at shorter pulse durations and higher breakdown fields. Using simplified Drude concepts[15] the average energy transferred to the atoms in successive collisions is given as $\Delta Q=(eEt)^2/2m$, where E is the dc field, t is the collision time (τ_{eff}), and m is the electron mass. It is determined that the average energy transfer per collision is less than $\sim$1 eV, even at 50 to 60 MV/cm. Thus the ionizing collisions are statistically controlled through the high-energy tail of the

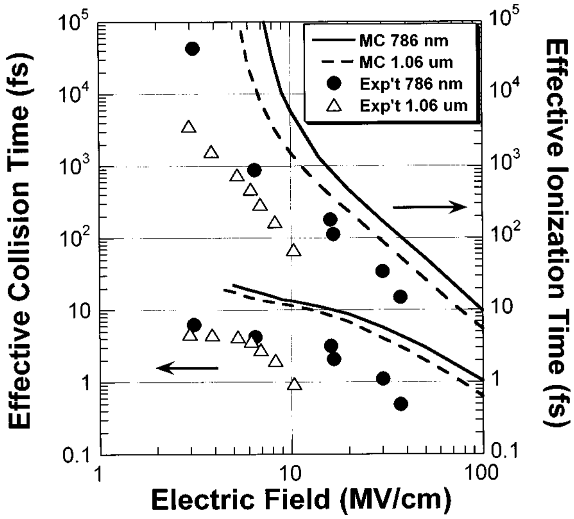

FIG. 4. Effectice electron lattice collision times as a function of electric field as obtained from experimental and theoretical (Monte Carlo) ionization coefficients.

electron distribution with the probability of ionization increasing with increasing field strength. It is observed that the ratio of scattering to ionizing collisions varies from several thousand to approximately 50 across the field strengths investigated. For very high fields the ionization collision times will converge to the dc rates while the scattering collision times become subfemtosecond.

In conclusion then, the present work confirms our earlier studies that a plasma condition is formed during the absorption of near-IR ultrafast laser pulses in silicon when the laser intensity exceeds a critical threshold and that this process is driven by avalanche ionization. In addition, intense electron heating occurs during the development of a critical plasma density with this energy being transferred to the lattice through strongly coupled plasma interactions. The energy density within the lattice is then responsible for the damage production that occurs at the surface of the sample through plasma expansion and associated thermodynamic processes resulting from plasma heating.

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