The Relative Roles of Avalanche Multiplication and Multiphoton Absorption in Laser-Induced Damage of Dielectrics

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Abstract—The optical electric field strengths associated with pulsed laser exposures needed to produce conduction electron densities of $10^{18}$/cm$^3$ in several direct-gap alkali halides are calculated using three different models: a simplified avalanche model, the Keldysh formulation of multiphoton ionization, and a combination of the two. Numerical calculations are performed for crystalline NaCl, KCl, KBr, NaF, LiF, and CaF$_2$ at wavelengths of 1.064, 0.694, 0.532, and 0.355 μm, for nanosecond and picosecond pulse durations. The results are compared with available experimental data resulting in the following observations: the damage field strengths predicted by the avalanche model scatter around the experimentally measured values, but they always agree within a factor of approximately four. The electric field strengths required for breakdown solely from the simultaneous absorption of four or more photons are significantly larger than the experimental values or the predictions of the avalanche model. However, in NaCl, KCl, and KBr the electric fields necessary for damage due to four-photon absorption are only slightly smaller than those needed for catastrophic avalanche multiplication, and are in significantly closer agreement with the experimentally measured damage thresholds. When the avalanche and multiphoton models are combined in a direct manner the resulting thresholds are close to the smaller of the two previously calculated thresholds, and are in reasonable agreement with the experimental data with respect to their dependence on laser frequency and pulse duration.

I. INTRODUCTION

FOR MORE than a decade there has been continual interest in the theoretical and experimental investigation of laser-induced damage in transparent optical materials [1]–[7]. This interest has been stimulated, in large measure, by the limitations imposed on the transmission of high-power laser pulses through optical materials as well as by a desire to understand the fundamental physics of the interaction of intense coherent electromagnetic radiation with this class of materials. Despite an abundance of published experimental and theoretical papers on this subject, quantitative understanding of the fundamental damage mechanisms involved is far from satisfactory [1]–[7]. The avalanche multiplication process and multiphoton ionization are generally believed to be two of the more important mechanisms involved in laser-induced damage. However, the relative importance of these two mechanisms in determining the actual damage threshold in specific materials as a function of material properties, wavelength, and pulse duration is not clear. Bloembergen [3] has hypothesized that for photon energies greater than one-half of the bandgap energy, multiphoton absorption will dominate over avalanche multiphoton as the principal damaging process. In an effort to investigate this situation and compare the roles of these two competing mechanisms, we have calculated, by three different theoretical approaches, the electric field strengths needed for intrinsic laser-induced damage in several crystalline dielectrics, at laser wavelengths and pulse durations of practical interest, for which experimental data are readily available. In the first case, a simplified model of avalanche multiplication [4] that assumes constant collision frequencies and energy loss is used. Next, the general expression derived by Keldysh [5] for the multiphoton ionization rate in direct-gap crystals is employed. Finally, following the approach of Vinogradov and Faizullov [6], the avalanche and multiphoton mechanisms are treated together in combined form.

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The optoelectric field strengths associated with nanosecond and picosecond laser pulses needed to produce conduction electron densities of the order of $10^{18}$/cm$^3$ are calculated by means of the above three models. It is known that, when the conduction electron density reaches the above value, the absorption of the residual laser beam becomes appreciable, leading to localized heating and usually irreversible damage of the optical material. Hence, the electric field strengths obtained with the above criterion are taken to be the damage field strengths, and will be compared with the appropriate available experimental damage thresholds. From this comparison, the relative importance of avalanche multiplication and multiphoton absorption in causing laser-induced damage in dielectrics can be analyzed.

II. THEORY

A. Avalanche Model

The avalanche model has often been successfully used to qualitatively explain some of the observed features of laser-induced breakdown. In an effort to obtain more quantitative results, we first calculate the intrinsic damage electric field strengths in several alkali halides by means of a model of avalanche multiplication outlined by Sparks.

This model assumes the rate of energy gain of a conduction electron from an alternating electric field $F = F_0 \cos \omega t$ to be given by

$$\frac{dE}{dt} = \frac{e^2 F_0^2 \tau}{m^* (1 + \omega^2 \tau_k^2)}$$

(1)

where $\tau_k$ is the electron-phonon relaxation time when only large angle scattering is considered, and $m^*$ is the conduction electron effective mass. The rate of energy loss to the lattice phonons is given by

$$\frac{dE}{dt} = \frac{\hbar \omega_p}{\tau_L}$$

(2)

where $\omega_p$ is the average phonon frequency and $\tau_L$ is the relaxation time when both large- and small-angle scatterings are considered. Once the conduction electron has attained an energy equal to the bandgap energy $\Delta$, it can excite a valence electron across the bandgap, and this process repeats itself. Thus, the conduction electron density $n_e$ at the end of the pulse duration $t_p$ will be

$$n_e = n_{e0} \exp(\omega_1 t_p)$$

(3)

where $n_{e0}$ is the initial density of conduction electrons and $t_p$ is the time interval during which a conduction electron starting with zero energy gains an energy equal to the bandgap energy $\Delta$. The value of $t_p$ is obtained from (1) and (2), assuming for this calculation that $\tau_L$, $\tau_k$, and $\omega_p$ are constants.

The avalanche growth rate is normally described by the equation

$$n_e = n_{e0} \exp(\omega_1 t_p)$$

(4)

where $\omega_1$ is the avalanche ionization frequency. This avalanche ionization frequency $\omega_1$ is related to the Townsend first ionization coefficient $\alpha$ by the equation

$$\omega_1(F) = \alpha(F) \nu_{\text{drift}}(F)$$

(5)

where $\nu_{\text{drift}}$ is the drift velocity of the electrons. From (1)-(4) one obtains

$$\omega_1 = \frac{0.693 e^2 F_0^2 \tau_k}{m^* (1 + \omega^2 \tau_k^2)} - \frac{0.693 \hbar \omega_p}{\Delta \tau_L}$$

(6)

As stated in the Introduction, the appearance of $10^{18}$ electrons/cm$^3$ in the conduction band leads to appreciable heating and concurrent damage to the dielectrics, and hence, this can be taken as a justifiable criterion for laser-induced damage. It is further assumed that the ionization of weakly bound sites such as $F$-centers, impurities, defects, etc., gives rise to an initial conduction electron density of $10^9$/cm$^3$. With these assumptions, we obtain the following expression for the damage field strength due to avalanche multiplication:

$$F_0 = \left[ \frac{m^* (1 + \omega^2 \tau_k^2)}{e^2 \tau_k} \left( \frac{\Delta \ln 10^9 + \hbar \omega_p}{0.693 t_p \tau_L} \right) \right]^{1/2} \Delta$$

(7)

Equation (7) is an expression for the electric field amplitude within the dielectric at the onset of damage in pure materials, and it is valid in the MKS system of units. We will later use this equation to calculate the numerical values of the damage field strengths in several crystalline dielectrics.

B. Multiphoton Absorption

Keldysh has theoretically derived an expression for the probability of electronic transitions from the highest valence band to the lowest conduction band of a direct-gap crystalline solid when a strong electromagnetic radiation is incident on the crystal. In his analysis, the energy of the photon was assumed to be smaller than that of the bandgap, and the transitions were presumed to take place between Stark shifted energy bands. Keldysh presented the following expression for the transition rate per unit volume:

$$W = \frac{2\omega}{9\pi} \left[ \frac{\sqrt{1 + \gamma^2} - \mu^* \omega}{\gamma} \right] \frac{\Delta}{\hbar \omega} \left[ Q \left( \gamma, \frac{\Delta}{\hbar \omega} \right) - \gamma \right] \left[ \frac{1}{\sqrt{1 + \gamma^2}} - E \left( \frac{1}{\sqrt{1 + \gamma^2}} \right) \right] \exp \left( \frac{1}{\sqrt{1 + \gamma^2}} - E \right)$$

(8)

In (8), $\mu^*$ is the reduced effective mass of the conduction electron and valence hole, $\Delta$ is the effective bandgap in the radiation field given by

$$\Delta = \frac{2\Delta}{\pi} \sqrt{1 + \gamma^2} E \left( \frac{1}{\sqrt{1 + \gamma^2}} \right)$$

(9)

where $E$ and $K$ are complete elliptic integrals of the first and second kind, while $\gamma$ is the ratio of the radiation frequency to the tunneling frequency given by

$$\gamma = \frac{\omega \sqrt{\mu^* \Delta}}{eF_0}$$

(10)

The symbol $\langle \cdots \rangle$ in (8) denotes the integer part of the argu-
ment. The function \( Q \) occurring in (8) is given by [5]

\[
Q(y, y^*) = \left[ \frac{\pi}{2K \left( \sqrt{1 + y^2} \right)} \right]^{1/2} \sum_{n=0}^{\infty} \left\{ \phi \left\{ \left( \frac{\pi^2 (2y + 1 - 2y + n)}{2K \left( \frac{1}{\sqrt{1 + y^2}} \right)} \right)^{1/2} \right\} \times \exp \left\{ -\frac{n\pi \left( K \left( \frac{\gamma}{\sqrt{1 + \gamma^2}} \right) - E \left( \frac{\gamma}{\sqrt{1 + \gamma^2}} \right) \right)}{E \left( \frac{1}{\sqrt{1 + \gamma^2}} \right)} \right\} \right\}
\]

(11)

where the function \( \phi \) is the Dawson integral.

One limiting case of (8), where \( \gamma \gg 1 \), has been extensively studied in the past [8]. In that case it has been often stated that the Keldysh formula is inaccurate or even invalid when \( \Delta / \hbar \omega_p \) is small, especially when this ratio is less than four. Recently it has been shown [9] that the above notion is ill founded and that the Keldysh formula can be employed even when \( \hbar \omega_p \) is a significant fraction of \( \Delta \). In order to further improve the predictive accuracy of the Keldysh formula in the large \( \gamma \) limit, as well as to treat the more general case of arbitrary values of \( \gamma \), we employed (8), instead of the more simplified expression for multiphoton absorption which has been used in the past, to calculate the electronic transition rates. The laser electric field strengths necessary to damage the dielectrics solely by multiphoton absorption will be later calculated with the aid of (8)–(11), for the condition

\[
W_{tp} \approx 10^{18} \text{ electrons/cm}^3.
\]

(12)

\section*{C. Combination of Avalanche and Multiphoton Models}

Vinogradov and Faizulllov [6] combined the effects of multiphoton and avalanche mechanisms by assuming that the two processes were independent of each other. They obtained the following expression for the conduction electron density:

\[
\eta_c = \left( \eta_{c0} + \frac{W}{\omega_L} \right) \exp \left( \frac{\omega_L t_p}{\omega_L} \right) - \frac{W}{\omega_L}
\]

(13)

where \( W \) and \( \omega_L \) are, respectively, the multiphoton and avalanche ionization frequencies. Equation (13) along with (6) and (8) will be used to calculate the damage electric field strengths in several dielectric crystals.

\section*{III. Numerical Calculations}

Equations (7)–(13) were used to calculate the laser damage electric fields in crystalline NaCl, KCl, KBr, NaF, LiF, and CaF\(_2\), for several wavelengths and pulse durations for which experimental damage thresholds are available. The selection of these materials was made on the basis of the availability of reliable values for the relevant effective masses. In employing (7) we assumed the following values of the relevant parameters [4] \( \tau_k = 1.36 \times 10^{-15} \text{ s}, \tau_L = 8.77 \times 10^{-16} \text{ s} \), and \( \hbar \omega_p = 1/40 \text{ eV} \). In addition, the values of the band structure parameters used in these computations are listed in Table I. The elliptic integrals and Dawson integrals encountered in the Keldysh formula were evaluated numerically. We included sufficient number of terms in the infinite sum in (11) to obtain convergency of the order of 0.01. Finally the calculated rms values of the electric field strengths in the dielectrics are listed in Table II along with a sampling of the published experimental data.

\section*{IV. Discussion}

Before making a comparison of the theoretical predictions with the experimental damage thresholds, the following points must be borne in mind. The theoretical predictions are for intrinsic breakdown. However, most of the experimental damage field strengths are governed largely by impurities and defects. Furthermore, the experimental data obtained by different researchers differ considerably from each other. These limitations make it very difficult to draw precise conclusions regarding the merits of different theoretical approaches. However, one can note the following general trends.

\subsection*{A. Electric Field Strengths at Damage}

The damage electric field strengths predicted by the avalanche model scatter around the experimentally measured values within a factor of four. The electric field strengths needed for dielectric breakdown caused solely by multiphoton absorption of order higher than four are much larger, and this mechanism plays a secondary role when \( \Delta \gg \hbar \omega \). It is also noted that in many cases when \( \Delta = 4 \hbar \omega \), the damage field strengths predicted by the Keldysh multiphoton absorption formula are smaller than those resulting from the avalanche model, and are in closer agreement with the published experimental values. These observations are in general agreement with Bloembergen’s [3] hypothesis, according to which, for photon energies greater than one-half of the bandgap energy, the multiphoton absorption will be the more dominant mechanism of laser-induced damage. When the avalanche and multiphoton mechanisms are combined as in (13), the resulting damage thresholds are close to the smaller of the two previous

\begin{table}[h]
\centering
\caption{Values of Band Structure Parameters Used}
\begin{tabular}{|c|c|c|}
\hline
Crystal & Energy gap \((\text{eV})\) & Conduction electron effective mass \((\text{eV})\) \\
& \text{[Ref. 13]} & Free electron mass \((\text{eV})\) \text{[Ref. 14]} \\
\hline
NaCl & 8.6 & 0.6 \\
KCl & 8.7 & 0.496 \\
KBr & 7.3 & 0.48 \\
NaF & 11.5 & - \\
LiF & 13.6 & - \\
CaF\(_2\) & 12.1 & - \text{[Ref. 15]} \\
\hline
\end{tabular}
\end{table}
B. Dependence of Damage Thresholds on Laser Frequency and Pulse Duration

According to the avalanche model of breakdown, the damage field strength predicted by (7) should increase with increasing frequency, approximately as \((1 + \omega^2 \tau_e^2)^{1/2}\). The multiphoton model, on the other hand, predicts a rapid decrease in damage thresholds with increasing frequency. The agreement of some of the earlier experimental results with the predictions of the avalanche model has been frequently used [2], [3] to advocate in favor of this model. However, the recent experiments of Manenkov [10] reveal that in many samples of NaCl, KCl, and CsI the damage threshold increases with increasing frequency up to that of ruby laser and decreases on further increasing the frequency to that of the second harmonic of Nd laser (see Fig. 1). (Newnam and Gill [11] have noted a similar behavior in thin film coatings of several oxides. However, we will not consider these data for comparison with theoretical predictions, since here we are concerned with damage in the bulk of crystalline materials.)

In Fig. 1 we have plotted the variation of the optoelectronic damage fields in NaCl as a function of the laser frequency. From a comparison of the theoretical predictions with the experimental data, we conclude that a combination of the multiphoton and avalanche mechanisms is closer to the real situation than either of these two models considered alone. We also believe that including the effect of multiphoton transitions on the avalanche ionization frequency will bring the theoretical predictions closer to the experimental results.

In order to more completely describe the frequency dependence of the damaging optoelectric field strengths, we repeated our calculations by adopting the following procedure. The electric field necessary to produce the critical number density of conduction electrons at the plasma frequency was obtained by setting the value of the critical number density varied from approximately \(10^{21}/m^3\) at 1.064 \(\mu\)m to about \(10^{22}/m^3\) at 0.355 \(\mu\)m. This variation, however, did not significantly affect the damage fields in the avalanche model, apparently due to the exponential nature of this process. When the above procedure was applied to the Keldysh formula, it resulted in large Stark shifts of the conduction electron energy states, rendering the model inappropriate.

In Fig. 2 we have plotted the pulswidth dependence of the optoelectric damage fields in NaCl at 1.06 \(\mu\)m. It is noted that the avalanche model predicts a weak inverse dependence of the approximate form \(F_0 \sim t^{-0.155}\), while the multiphoton model predicts a somewhat stronger variation of the form \(F_0 \sim t^{-0.355}\). The experimental dependence of the damage fields on the pulse duration is stronger than either of these

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Wavelength ((\mu)m)</th>
<th>Pulse duration ((\mu)sec)</th>
<th>R.M.S. electric field strength (V/cm)</th>
<th>Avalanche model</th>
<th>Multiphoton model</th>
<th>Multiphoton assisted avalanche</th>
<th>Experimental</th>
<th>Reference to experiments</th>
<th>Photon energy gap (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaF</td>
<td>1.064</td>
<td>0.03</td>
<td>5.21</td>
<td>2.05</td>
<td>3.24</td>
<td>1.98</td>
<td>2.05</td>
<td>10.12</td>
<td>1.12</td>
</tr>
<tr>
<td>KCl</td>
<td>1.064</td>
<td>0.03</td>
<td>5.21</td>
<td>2.05</td>
<td>3.24</td>
<td>1.98</td>
<td>2.05</td>
<td>10.12</td>
<td>1.12</td>
</tr>
<tr>
<td>CaF(_2)</td>
<td>1.064</td>
<td>0.03</td>
<td>5.21</td>
<td>2.05</td>
<td>3.24</td>
<td>1.98</td>
<td>2.05</td>
<td>10.12</td>
<td>1.12</td>
</tr>
</tbody>
</table>

The table above represents a comparison of calculated and experimental R.M.S. electric field strengths at damage.
V. CONCLUSION

It is shown that neither the avalanche nor the multiphoton model alone can explain all of the experimentally observed features of laser induced damage. It is suggested that it is more appropriate to combine these two mechanisms into a single formulation. This should be done in a more detailed and appropriate manner through treatment of their interdependency. Specifically, the effect of multiphoton transitions on the avalanche growth rate constant should be included. The energy dependence of the collision frequencies as well as the effects of surface states, impurities, color centers, etc. should also be considered. Calculations incorporating the above are currently under active pursuit.

REFERENCES