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Phenomenological theory of laser damage in insulators

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A phenomenological theory of laser damage is presented. The physical mechanism for laser damage in insulators is presumed to be similar to the one for breakdown in semiconductors. A few starter electrons in the conduction band are excited by the external field to the point where an electron avalanche occurs. Heat is generated by electron relaxation by phonon emission, and damage occurs when the temperature of the irradiated volume reaches the melting point. The phenomenological theory contains two parameters: an average cross section for excited-state absorption, which for NaCl is 1.8×10^{-17} cm^2 and an average relaxation rate by phonon emission, which is about 10^{14} sec^{-1} for a 200-cm^{-1} phonon in NaCl. These values overcome problems with previous models, which require very large electron relaxation rates ($> 10^{15}$ sec^{-1}). The parameters of the theory are determined by fitting the results of calculations to experiment. With this theory, subthreshold properties such as the hot-electron distribution, rate of electron avalanche, and rate of heat generation can also be calculated. Ways of verifying the theory are discussed in detail.

I. INTRODUCTION

The phenomenon of laser damage in insulators is of considerable scientific interest as a manifestation of the properties of very hot electrons. The subject is also of technological importance since it places a limit on laser beam intensities in optical materials. We present here a phenomenological theory of laser damage in insulators. In this theory, both the electromagnetic absorption and the electron-phonon interaction are treated phenomenologically. No attempt is made to perform first-principles calculations of the interaction of hot electrons with radiation. We assume that the photon energy is at least 1 eV and that the band gap energy is more than three times the photon energy. While this somewhat limits the applicability of our theory, it is in just this range that present models of laser damage are inadequate. Reviews of the experimental aspects of laser damage and of theories are given by Smith,[1] Bloembergen,[2] and Fradin.[3]

The phenomenological theory discussed here contains two parameters: an average cross section for excited-state absorption by whatever hot excitations (electrons or holes) exist; and an average relaxation rate for those excitations by phonon emission. The parameters are determined by fitting the results of calculations to experiment. With this theory not only damage thresholds can be calculated but also such subthreshold properties as the hot-electron distribution, rate of electron avalanche, and rate of heat generation. These quantities can perhaps be estimated by fluorescence emission, excited-state absorption, photoconductivity, and photoacoustic experiments. In Sec. IX we present a detailed discussion of the predicted results of such experiments based on the theory developed here.

The purposes of this theory are twofold: (i) to provide a framework for interpreting damage data; and (ii) to provide a model useful enough to stimulate additional experiments to evaluate the model's validity and to ascertain details of very-hot-electron phenomena.

Measuring reliable laser-damage thresholds requires care in stabilizing, characterizing, and focusing the laser beam. Discussions of the required techniques and precautions are given in Refs. 2 and 4–7. For our purposes, a well-controlled experiment proceeds as follows. A laser beam of smoothly varying spatial and temporal intensity is focused to a 1–10 μm spot in the bulk of a reasonably pure, transparent, optically perfect insulator. Self-focusing is avoided. The material is said to be damaged when a small number of spots (for psec pulses) or larger damaged regions (nsec pulses) are observed at the focal point with an optical microscope. These damage sites are only apparent when the laser intensity exceeds a certain critical value, called the *threshold intensity*. The magnitude of the threshold intensity depends on the duration of the pulse. The threshold intensity is reproducible to ± 30% in controlled experiments. Usually, but not always, damage is accompanied by visible emission. If the damaged material is an optical component, the damage sites represent a degradation of the local optical properties of the component. Increasing the beam intensity above threshold increases the density and size of the damage sites until large damaged regions are obtained. Damage morphology is discussed by Milam,[4] Smith *et al.*,[8] and the reviews.[1-3]

Laser wavelengths from 10.6–0.3 μm have been used. A large amount of data exist for 1 and

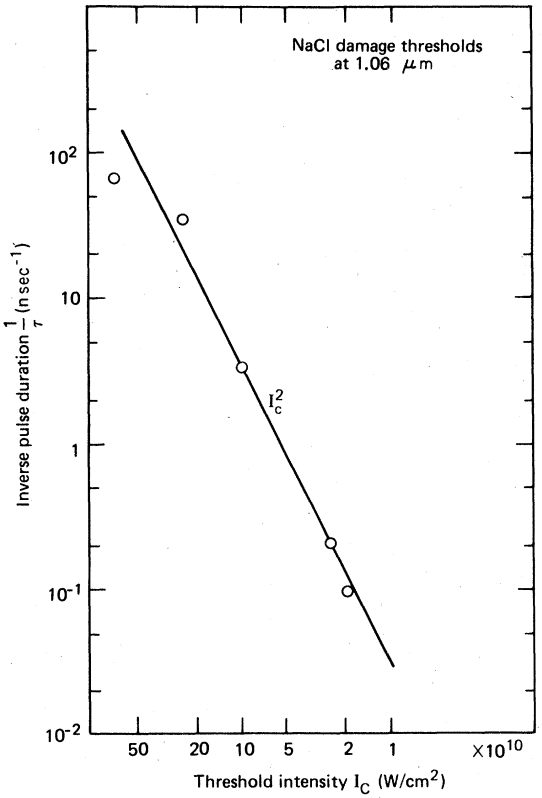

FIG. 1. Bulk damage thresholds for NaCl at 1.06 μ. I_c is the threshold intensity and τ is the pulse duration, full width at half-maximum. The data can be fit by a line $1/\tau \sim I_c^2$, although there is not theoretical support for this behavior. The points are from the tabulation in Ref. 1.

0.5 μm wavelengths. Solids include materials with very large band gaps such as Lif, NaCl, BeF_2, and SiO_2.

We are interested here in bulk intrinsic laser damage of materials of purity typical of high-quality optical materials. Polished surfaces have lower damage thresholds than the bulk material by as much as a factor of 5 in intensity. Damage is believed to occur at intensities below intrinsic thresholds at the sites of metallic inclusions (which readily absorb the incident radiation) or sharp edges (which enhance the local field). We assume such defects are not present.

Intrinsic threshold intensities at 1.06 μm for NaCl are shown in Fig. 1. The data were taken from Ref. 1, to which the reader is referred for the original references. In Fig. 1, I_c is the threshold intensity, defined as the maximum intensity of the pulse, and τ is the time duration, full width at half-maximum, of the pulse. The shorter the pulse the greater the intensity needed to cause damage. We see from Fig. 1 that the data follow the relation $1/\tau \sim I_c^2$.

II. PRELIMINARY REMARKS

It seems to be generally accepted that the sites which are the first signs of laser damage are due to local melting of the solid, followed by resolidification in such a manner that a variation of dielectric constant results. We will accept this explanation here.

Therefore, in order to explain laser damage, we must explain how a laser beam of photon energy near 1 eV can raise a local region of a transparent insulator (bandgap in excess of 8 eV and absorption coefficient of about 10^{-3}–10^{-4} cm^{-1} at the laser frequency) to the melting point.

Not enough heat can be absorbed by linear absorption to effect this temperature rise. The rate of absorption per cm^3 of energy from the beam is αI, where α is the absorption coefficient and I is the beam intensity. Neglecting thermal losses, the temperature rise produced by a square pulse of duration τ is

$$\Delta T = \alpha I \tau / C \ ,$$

where C is the specific heat of the solid. Using threshold values for NaCl (cf. Fig. 1) $I = 2\times10^{10}$ W/cm^2, $\tau = 10$ nsec and also using $C = 2\times 10^7$ erg/cm^3 °C, $\alpha = 10^{-3}$/cm, we find a temperature rise of

$$\Delta T = 0.1\,^\circ\mathrm{C}.$$

Moreover, the relation between the critical intensity and pulse width for damage due to linear absorption would yield the relation $\tau I_c = \text{const.}$, in contradiction to Fig. 1.

An additional, stronger absorption must be induced somehow by the incident laser beam. One way for this to occur is for electrons to be excited into the conduction band (CB) of the insulator. These electrons can strongly absorb the incident radiation. The phenomenon of *excited-state absorption* has been measured for a number of insulators,[9-12] and for electrons near the CB bottom appears to proceed with a large absorption coefficient.

An estimate of the number of CB electrons needed to produce laser damage by heating can be obtained as follows. At relatively low fields and long pulses (>1 nsec) the rate of heat generation by CB electrons equals the rate of energy absorption from the incident radiation. That is, energy is dissipated as rapidly as it is absorbed. The rate of absorption is $N_0 I \sigma$, where N_0 is the number of CB electrons, I is the beam intensity, and σ is the cross section for absorption by a CB electron [and not the cross section for absorption from donor levels or from the valence band (VB) to the CB]. Then at threshold the amount of absorbed energy required to raise the solid through temperature ΔT

to its melting point obeys the relation

$$N_0 \sigma I_c \tau = C\Delta T .$$

Let us use the values $C = 2\times10^7$ erg/cm^3 °C, $\Delta T = 10^3$°C. Then for threshold values for NaCl (cf. Fig. 1) we find

$$N_0 > \frac{5}{\sigma}\Big/\text{cm}^3 \quad (\tau = 10 \text{ nsec}),$$

$$N_0 > \frac{150}{\sigma}\Big/\text{cm}^3 \quad (\tau = 0.03 \text{ nsec}) .$$

For $\sigma = 10^{-15}$ cm^2, which results from an oscillator strength of unity and a natural linewidth of about 1000 cm^{-1}, we find

$$N_0 > 0.5\times10^{16}/\text{cm}^3 \quad (\tau = 10 \text{ nsec}) ,$$

$$N_0 > 1.5\times 10^{17}/\text{cm}^3 \quad (\tau = 0.03 \text{ nsec}) .$$

We use the inequalities above because, first, the cross section is probably one or two orders of magnitude smaller than 10^{-15} cm^2 and, secondly, because at high fields (smallest τ) the rate of heat generation is less than $N_0 I\sigma$ as will be seen later.

III. DIRECT EXCITATION OF ELECTRONS

There are a number of theories describing the manner in which a sufficient number of electrons are excited to the CB.[1] One way this can occur is to excite electrons by multiphoton absorption directly from the valence band to the conduction band.[13] However, for insulators, five or more photons are usually required, and damage occurs long before the radiation intensity is sufficient for such high-order processes. Moreover, in NaCl the τ vs I_c relation for multiphoton absorption at 1.06 μm and subsequent heating is

$$\tau I_c^8 \sim \text{const.}$$

which is in severe disagreement with the data of Fig. 1.

It is possible to generate CB electrons in insulators by exciting them by one- or two-photon absorption to the CB bottom from shallow donor levels which are associated with defects and/or impurities and whose energies lie a few eV below the CB bottom. (A familiar example is the excitation of electrons from color centers in alkali halides.) Once in the CB, heat is generated by linear excited-state absorption and subsequent phonon generation.[13,14]

There are several experiments which indicate that this mechanism cannot furnish enough electrons in transparent solids of excellent optical quality, although of course in solids with large concentrations of impurities or color centers this mechanism is likely to be important for laser damage. One of these experiments involves some observations during laser gain measurements.[15] In these experiments, 1.06-μm laser beams are routinely propagated down Nd-doped glass rods of approximately 1-m length with very little attenuation. These beams have an intensity of as much as 3×10^9 W/cm^2, about a factor of 3 or 4 lower than threshold intensity, and pulse durations of a few nsec. Now we argue that if damage is due to heat generated by electrons excited from shallow donor levels, then the electrons can also be excited by this saturating beam. Since the saturating beam is of subthreshold intensity, fewer electrons are excited than at threshold and less heat is generated. Since the beam is not attenuated, the electrons excited into the CB by this saturating beam must have a total absorption coefficient α such that at the laser frequency $\alpha < 10^{-4}$ cm^{-1}.

Let us suppose, to be generous, that electrons must be raised from donor levels by a three-photon process. (One possibility is a two-photon absorption to a second trap level and a subsequent absorption to the CB.) Then the rate of excitation of electrons to the CB will be proportional to I^3. Let N_0 be the number of electrons produced by a beam of threshold intensity I_c. (In Sec. II we showed that $N_0\sigma > 5$ is required to produce damage by heating from long pulses.) Then the number of electrons generated by a subthreshold field of intensity I is

$$(I/I_c)^3 N_0.$$

The absorption coefficient of CB electrons excited by the laser beam is

$$(I/I_c)^3 N_0\sigma < 10^{-4}$$

and therefore we find the bound $N_0\sigma < 6.4\times10^{-3}$. This is far too small to produce damage by heating.

IV. ELECTRON AVALANCHE

Since it does not appear that a sufficient number of CB electrons can be generated either directly from the VB or from donor levels in pure, undoped materials, one is led to consider the possibility of electron avalanche. It is well known that dielectric breakdown in gases and some semiconductors is due to electron avalanche. However, avalanche has not been explicitly observed in insulators.

The steps in a mechanism of laser damage by electron avalanche have been developed by previous work and are as follows. (Complete references are given in the reviews.[1-3] See also Refs. 16–19 and the discussion below.) Initially some *primer electrons* are raised from shallow donor states to the

CB bottom. The number of these electrons is small, possibly of the order of $10^{10}/cm^3$, and not enough to produce damage by themselves. A lower bound of the number of primer electrons of about $10^{10}/cm^3$ has been presented by Smith.[8] The CB electrons absorb energy from the incident radiation and at the same time they lose energy by emitting phonons. If the rate of absorption is sufficiently great, some fraction of the electrons can gain an amount of energy greater than the band-gap energy. These electrons, by an Auger process, can excite an additional electron from the VB to the CB bottom. This process continues until a sufficient number of electrons are raised from the VB to the CB to absorb enough energy to cause damage by heating.

It seems probable that hole absorption will play a significant role in this process, so that it is more realistic to talk about both CB electrons and VB holes as absorbing the incident radiation. Usually we will just mention CB electrons, but the possibility of hole absorption is also presumed.

This physical picture of the damage process is the one that we assume. Our new contribution is to fill in some of the details of this picture by specifying the appropriate absorption mechanism and then by computing not only damage thresholds but also predictions of the results of a number of subthreshold experiments such as photoconductivity or photoacoustic spectroscopy.

The main characteristic of an avalanche mechanism for laser-induced breakdown is the requirement that the CB electrons (and/or the VB holes) gain large amounts of energy. For large-gap insulators the energies required are over 8 eV. These are extremely hot electrons, much hotter than the so-called hot electrons usually dealt with in semiconductors (see, for instance, Refs. 20–24). To create such hot electrons, one requires that the rate absorption of energy from the applied field be comparable to the rate of energy loss by phonon emission. Such processes have not been studied either theoretically or experimentally except for dc fields, where there is a large body of literature. In fact, it has not yet been demonstrated that irradiation at a wavelength of 1 μ can excite CB electrons to 8-eV energy. Such a demonstration is an important experimental problem.

Experimental evidence for laser-induced electron avalanche in solids is indirect at best. (Even for the case of dc breakdown in insulators, the contribution of impact ionization is still controversial.[20,25] Several authors have argued against impact ionization in insulators.[26–28]) A number of investigators[29,30] have observed that, when damage occurs, the transmission of the damaging laser beam through the sample was sharply cut off. Alyassini and Parks[31] in a surface damage study with nsec pulses observed the same thing and also observed a change in the reflected intensity of a probe beam which occurred 1 nsec before damage occurred. These effects were attributed to a strongly absorbing electron plasma produced by electron avalanche. Alyassini and Parks estimate that 10^{18} electrons/cm^3 are present in the CB when damage occurs.

Yasojima *et al.*[32] measured the integrated photocurrent of CB electrons which were excited during irradiation of LiF and KCl with intense ~ 50-nsec laser pulses at 1.06 and 0.69 μm. In all materials the slow growth of a population of CB electrons at intensities more than a factor of 5 below threshold was observed. Presumably these were the primer electrons. In LiF the authors aver that no electron multiplication occurred at the damage threshold. (The LiF data have considerable scatter.) On the other hand, for KCl a strong electron multiplication was observed over a narrow range of intensities just below threshold. There was little scatter in the KCl data. Thus the two alkali halides appear to display the opposite behavior.

Finally, in an article on transient excited state absorption in KCl, Williams, Bradford, and Faust[9] report the following observations, which seem an argument against the existence of an electron avalanche. At the fourth harmonic of YAlG:Nd, F bands were easily produced in KCl at intensities much below damage thresholds due to direct VB-CB excitation. These bands produce an intense coloration in KCl crystals. However, for the first three harmonics of YAlG:Nd no coloration in KCl could be produced at fields up to and exceeding damage thresholds. Intensities great enough to produce a "line of fractures" were used. (Pulse duration was ~ 20 psec.) This experiment possibly indicates that damage was not caused by avalanche, since direct excitation of VB electrons to the CB produces coloration.

It seems clear that the *unequivocal demonstration of the existence or absence of laser-induced electron avalanche is the most pressing experimental problem in laser damage in solids.*

V. CLASSICAL THEORY

One theory which incorporates electron avalanche in the breakdown mechanism assumes that the electric field of the incident radiation accelerates the electron as if the electron were a classical particle; that is, the electron-radiation interaction is classical and not quantum mechanical. The electron is taken to be a free particle with some effective mass and which is scattered by the phonons. The scattering is a perturbation. Hole ab-

sorption is ignored. (Holes in insulators generally have very low mobilities compared with electrons.) The avalanche arises from impact ionization, a phenomenon that is well known in dc breakdown in semiconductors and gases. We refer to this theory as the *classical theory*. The classical theory was sketched by several people[1-3,16,33] and was formalized by Holway[34] and Holway and Fradin.[35] The classical theory of laser damage was motivated by the fact that the damage threshold intensity increases slightly with frequency so there is a similarity between the threshold electric fields for dc dielectric breakdown and for long-pulse-duration (>1 nsec) laser damage at 10.6, 1, and 0.5 μm.[1-3,16] (Since the threshold intensity for laser damage depends on the pulse duration, it is only reasonable to compare dc thresholds with ac thresholds for long pulses.) Moreover, for both dc and long-pulse laser-induced breakdown, the threshold field decreases with decreasing bandgap.

Arguments against the classical theory are summarized below.

(i) An important prediction of the classical theory is that the avalanche rate for a dc field is greater than the avalanche rate for an rms ac field of the same magnitude. This is contradicted by experiment.

dc dielectric breakdown in insulators probably does not result from a destructive runaway electron avalanche as envisioned in older theories[42-44] and as certainly takes place in gases and narrow-gap semiconductors. The evidence for this seems conclusive in thermal SiO_2 films[36-40] and O'Dwyer[20,41] has theorized a nondestructive avalanche as a prerequisite for dc breakdown in many insulators including alkali halides. Therefore, for given electric field strength, dc avalanche rates are expected to be much smaller than rates required to explain the ac damage thresholds, where runaway, destructive avalanche is presumably the cause of damage. This has been demonstrated quantitatively for thermal SiO_2 films.[25] While impact ionization does occur during dc breakdown, its rate in SiO_2 films is orders of magnitude too small at dc fields below 5×10^6 V/cm to explain the laser induced breakdown. The argument was originally given by Hughes.[25]

(ii) The classical theory does not appear to be self-consistent. In the classical theory, breakdown occurs when the average rate of energy absorption equals, approximately, the average rate of energy dissipation by phonon emission.[42,44] (This is not altered much by considering a distribution of electron energies, since near threshold this function is such a rapidly varying function of the field that the avalanche rate changes by many orders of magnitude for a field change of less than 50%.[25,35,36,45,46]) Thus we find at threshold, for the classical theory,

$$\hbar\Omega_0 r = (q^2F^2/m^*)\, r/(r^2+\omega^2)^{1/2}\,,$$

where m^* is the electron effective mass, Ω_0 is the frequency of a dominant phonon, F is the dc field or rms ac field, ω is the angular frequency ($\omega=0$ for dc), and r is the electron relaxation rate. The quantity r is the average rate for emitting an optical phonon. For NaCl we use $\Omega_0 = 0.021$ eV, $m^* = m$, and $F_{dc} = 1.5\times10^6$ V/cm. In order to fit the dc threshold data, we find $r = 2.5\times10^{14}$ sec^{-1}. For a frequency of 1 μm we find, using this value of r, that $F_{ac} = 7F_{dc}$. Experimentally, $F_{ac} < F_{dc}$.

(iii) In order to agree with measured ac damage thresholds, the classical theory of laser-induced breakdown requires an electron-phonon relaxation rate r of *at least* 3×10^{15} sec^{-1}, or more than 1 eV.[1-3] Such a rapid relaxation rate means that the classical theory is not self-consistent and that the electron-radiation interaction must be treated quantum mechanically.

(iv) The formal theory of laser-induced breakdown given by Holway and Fradin[35] is in severe disagreement with experiment, except perhaps at the highest laser intensities. The reason is the rapid variation of the electron avalanche rate for fields required for average energy balance [see (ii) above].

VI. EXCITED-STATE ABSORPTION

We will adopt the general description of the steps leading to breakdown discussed in Sec. IV. That is, initially a few primer electrons are excited to the CB from shallow donors, these electrons gain energy from the field to the point where they can excite additional VB electrons to the CB. This process of avalanche continues and when the sample temperature is raised to the melting point, damage occurs. Heat is generated during this process due to the relaxation by phonon emission of hot electrons.

The problem of quantifying these ideas is one of formulating the mechanism by which a CB electron gains energy from the applied field. The discussion of the classical theory in Sec. V indicates that the electron must be treated quantum mechanically. Therefore, it is reasonable to expect that the electron gains energy by the absorption of discrete photons and that the energy gained in each absorption process equals the photon energy, apart from phonons created in the absorption. The absorption may be phonon assisted, as discussed below, but there are mechanisms whereby intra-

band transitions may become allowed even without the participation of phonons.

The phenomenon of excited-state absorption (absorption by CB electrons) in alkali halides and other ionic crystals has been documented by Williams and Kabler and co-workers.[9-12] For cool electrons this absorption seems to have oscillator strengths exceeding 10^{-1}.

There are a number of mechanisms by which a CB electron may absorb photons. First there are direct, interband transitions, which are allowed. Phonon-assisted intraband and interband absorption can occur. Furthermore, the presence of a random distribution of relatively immobile holes effectively breaks electron momentum selection rules and can have a strong influence on absorption strength. This is apparently observed in semiconductors (Ref. 47 and references therein). Cool electrons may become self-trapped, and hence localized, and will have strong absorption strengths.[9-12] The excitation of VB electrons to the CB may rapidly (< 10 psec times) make precusor sites for various defects.[9,58] These sites involve lattice distortion, which then destroy crystalline order in the vicinity of the hot electron, and this in turn enables intraband transitions. For amorphous solids, or crystalline solids with defects, where there is no translational invariance and where the electron may be localized to some degree in the Anderson sense,[48] all intraband CB transitions will be allowed.

The relative contribution of most of these mechanisms to intraband absorptions of CB electrons in insulators is at present completely unknown.

Theories of phonon-assisted excited-state electronic absorption, with applications to laser damage, have been given previously by Sparks[14,15] and by Wasserman.[49] Sparks has calculated damage levels and reports excellent agreement with experiment. In both Sparks's and Wasserman's theories, perfect crystals are assumed, the electron-phonon interaction is treated by second-order perturbation theory and momentum is conserved in the absorption process by the participation of a single phonon.

Sparks found that the electron relaxation rate required to explain the damage data is very large. For electron energies of about 1 eV Sparks calculates an electron relaxation rate of 1.14×10^{15} sec^{-1}, which is about 0.6 eV. These large relaxation rates make it unlikely that the perturbation approach is valid. It also may indicate the participation of several phonons in the absorption process.

It is well known that the electron-phonon interaction is strong for electron energies extending from about 0.05 eV to at least a few eV. The strength of this interaction for lower-energy electrons is revealed by the well-known saturation of electron mobilities with increasing dc fields.[22-24] For the case of higher energies, above a few tenths of an eV, it has been observed experimentally that electrons accelerated by strong fields through thin-film oxides lose energy at the rate of 0.03–0.1 eV/Å, or at least one phonon is created every time the electron travels the distance between two atoms.[50,51]

The problem of the large relaxation rate for hot electrons and the inability of perturbation theory to deal accurately with such rates is well known. Seitz[42] took pains to call attention to this problem several times, and the problem has been discussed by many others. The calculation of Thornber and Feynman[52] show that such energy losses can be obtained theoretically from a proper calculation. This calculation was not by perturbation theory but by a path integral method and is valid for arbitrary values of the electron-phonon coupling constant. Thornber and Feynman used Frohlich's Hamiltonian[53] and so did not include deformation potential scattering in their calculation. (The use of deformation potential[42] scattering will lead to strong scattering when the electron wavelength is of the order of an interatomic spacing of the material.) It is interesting to quote from this work on the nature of hot-electrons. "Even for very low temperature there is a range of velocities... over which the electron-lattice scattering is so severe that... a quasiparticle picture of the electron is not possible. The lifetime is so short and catastrophic that the electronic state cannot be viewed as decaying exponentially in time. (In fact the broadening is roughly two orders of magnitude larger than the energy.) Mott pointed out to us that under such conditions electrons and holes in this energy region in highly ionic materials would behave quite differently than a rigid-lattice band theory calculation would predict... ."

For the Frohlich Hamiltonian, "the troublesome region is confined to a few tenths of an eV." If deformation potential scattering were included, however, these comments would probably be true for a larger range of electron energies. This strong scattering would be expected to produce a reasonably large ($\sigma > 10^{-18}$ cm^2) phonon-assisted intraband absorption.

VII. RATE EQUATIONS

In this section we develop the rate equations describing laser-induced electron avalanche and heating. The hot electrons are characterized by two parameters, an absorption cross section and a relaxation rate, both of which are taken to be in-

TABLE I. Numerical values of avalanche rate β and of ϕ, where $1-\phi$ is the rate of heat generation. These values are computed from Eqs. (19) and (20) as discussed in the text, by integrating the Eqs. (16)–(18) and taking the appropriate long-time behavior.

	$\delta=4$			$\delta=6$	
f	β	ϕ	f	β	ϕ
10	1.44 (−1)	0.091 64	4	1.231 (−1)	0.1151
8	1.13 (−1)	0.091 64	2.75	8.136 (−2)	0.1151
6.5	8.983 (−2)	0.091 64	2	5.632 (−2)	0.1151
5	6.667 (−2)	0.091 69	1.5	3.969 (−2)	0.1185
4	5.125 (−2)	0.092 82	1	2.325 (−2)	0.128
3	3.589 (−2)	0.095 51	0.9	2.0 (−2)	0.1316
2	2.068 (−2)	0.102 5	0.8	1.679 (−2)	0.1364
1.5	1.326 (−2)	0.111 1	0.7	1.362 (−2)	0.1429
1	6.28 (−3)	0.131 6	0.6	1.052 (−2)	0.152
0.9	5.001 (−3)	0.139 3	0.5	7.535 (−3)	0.1657
0.8	3.795 (−3)	0.149 6	0.4	4.743 (−3)	0.1881
0.7	2.688 (−3)	0.163 6	0.3	2.32 (−3)	0.2290
0.6	1.716 (−3)	0.183 4	0.25	1.348 (−3)	0.2636
0.5	9.280 (−4)	0.212 6	0.2	6.175 (−4)	0.316
0.45	6.2 (−4)	0.232 6	0.166 667	2.929 (−4)	0.3664
0.4	3.773 (−4)	0.257 6	0.125	7.315 (−5)	0.4556
0.35	2.012 (−4)	0.289 4	0.1	2.099 (−5)	0.5278
0.3	8.89 (−5)	0.330 2	0.085	7.758 (−6)	0.5793
0.25	2.962 (−5)	0.383 0	0.07	2.177 (−6)	0.6377
0.2	6.335 (−6)	0.451 6	0.06	7.523 (−7)	0.6805
0.175	2.272 (−6)	0.493 6	0.05	2.039 (−7)	0.7266
0.1	1.501 (−8)	0.660 4	0.04	3.907 (−8)	0.7759
0.085	2.931 (−9)	0.702 5	0.03	4.38 (−9)	0.8282
0.07	3.873 (−10)	0.747 8	0.015	2.026 (−11)	0.9117
0.05	1.014 (−11)	0.813 3			
0.03	3.36 (−14)	0.884 3			

	$\delta=8$			$\delta=10$	
f	β	ϕ	f	β	ϕ
1.88	9.502 (−2)	0.127 7	2	1.574 (−1)	0.1531
1	4.608 (−2)	0.136 1	1	7.356 (−2)	0.1498
0.8	3.503 (−2)	0.142 9	0.8	5.7 (−2)	0.1538
0.7	2.952 (−2)	0.147 8	0.65	4.46 (−2)	0.16
0.6	2.404 (−2)	0.154 6	0.5	3.228 (−2)	0.1711
0.55	2.132 (−2)	0.158 9	0.4	2.413 (−2)	0.1836
0.5	1.860 (−2)	0.164 2	0.3	1.607 (−2)	0.2044
0.4	1.325 (−2)	0.179 0	0.25	1.203 (−2)	0.2211
0.35	1.063 (−2)	0.189 9	0.2	8.305 (−3)	0.2464
0.3	8.091 (−3)	0.204 9	0.15	4.746 (−3)	0.2895
0.25	5.67 (−3)	0.226 7	0.1	1.792 (−3)	0.3759
0.2	3.456 (−3)	0.260 9	0.08	9.34 (−4)	0.4364
0.175	2.474 (−3)	0.286 1	0.07	6.063 (−4)	0.4759
0.15	1.614 (−3)	0.319 8	0.06	3.538 (−4)	0.5233
0.125	9.131 (−4)	0.366 2	0.05	1.778 (−4)	0.5796
0.1	4.105 (−4)	0.431 2	0.04	7.158 (−5)	0.6459
0.085	2.131 (−4)	0.482 2	0.03	2.033 (−5)	0.7223
0.07	9.013 (−5)	0.544 4	0.025	8.822 (−6)	0.7641
0.055	2.788 (−5)	0.619 3	0.02	3.091 (−6)	0.8081
0.04	5.146 (−6)	0.707 6	0.01	1.082 (−7)	0.9015
0.03	1.016 (−6)	0.773 8	0.007	1.902 (−8)	0.9307
0.02	9.460 (−8)	0.845 3	0.005	3.715 (−9)	0.9503
0.01	1.052 (−9)	0.921 1	0.002 5	1.351 (−10)	0.9751
0.008	3.954 (−10)	0.936 6			
0.005	2.416 (−11)	0.960 2			

TABLE I. *(Continued)*

	$\delta = 20$	
f	β	ϕ
0.5	1.3 (−1)	0.2827
0.4	1.018 (−1)	0.2827
0.3	7.382 (−2)	0.2834
0.2	4.619 (−2)	0.2969
0.1	1.933 (−2)	0.3581
0.075	1.292 (−2)	0.3980
0.05	6.875 (−3)	0.4687
0.03	2.72 (−3)	0.5801
0.01	2.369 (−4)	0.8148
0.005	4.516 (−5)	0.9028
0.003	1.363 (−5)	0.9408
0.001	1.182 (−6)	0.9801

dependent of the electron energy. (The density of electronic states is part of the cross section and relaxation rate.) We do not mean to imply that the electron loses energy by emitting one phonon at a time. Rather we assume some average rate for emitting an amount of energy equal to the energy of some typical optical phonon. The manner in which this quantity is related to the details of phonon emission is not specified.

Electron avalanche will occur at a significant rate when the rate of energy absorption is comparable to the rate of energy emission, as discussed in Sec. IV.

Therefore, we have a two-parameter theory whose parameters may be obtained by fitting the predictions of the theory to experiment. It will be seen that this theory leads to good agreement with laser-damage experiments.

It is useful here to state our final results. We derive an electron multiplication rate $\gamma\beta$ which is a function only of the rate of absorption Γ and a rate γ of emitting phonons. We also derive a quantity ϕ such that $1-\phi$ is proportional to the rate of energy dissipation per electron. The quantity ϕ is also a function only of Γ and γ. Both β and ϕ are tabulated in Table I. The equations for laser damage can then be written in terms of β and ϕ as well as rates for excitation from donors and direct VB-CB excitation. These equations are Eqs. (21)–(24). The condition for damage is given in Eq. (10). These equations, together with the results of Table I, are all that are required for this phenomenological theory.

The rate equations quantify the physical processes involved in laser damage that have been discussed in Secs. IV and VI. They incorporate the following features: (i) absorption and stimulated emission of photons by CB electrons; (ii) electron relaxation by phonon emission; (iii) electron multiplication by an Auger process; and (iv) excitation of primer electrons from donor levels.

For simplicity of analysis we replace the continuum of CB levels by a sequence of equally spaced energy steps. (We will refer to the discrete energy points as *steps* rather than levels since we do not want to imply that the CB actually is composed of a series of discrete levels equally spaced in energy.) The resulting equations are much simpler to integrate than equations based on the true case of a continuum of CB levels. Compared to the generalizations of using an energy-independent absorption cross section and electron-phonon relaxation rate, little error is entailed in this approximation.

We can now envision the electron as climbing the "ladder" of CB steps during absorption and cascading down the ladder during phonon emission. A schematic diagram of this model is given in Fig. 2. (The model, formulated in this manner, is similar to one of laser-induced dissociation of molecules.) When the electron arrives at the top of this ladder (the Nth step) it excites an additional electron from the VB to the bottom of the CB, and it also loses its energy and falls to the CB bottom. This model incorporates all the physical steps for an avalanche theory of laser damage discussed in previous sections.

Let there be $N+1$ CB steps (cf. Fig. 2) and let $p_i(t)$ be the number of CB electrons in level i; $i = 0, 1, 2, \ldots, N$. (We mean, of course, electrons and holes both.) Let η be the number of electrons in shallow donor levels. The equations for p_i and η are

$$\frac{d}{dt}P_i = \Gamma[P_{i+\delta}\Theta(N-i-\delta)-P_i]+\Gamma(P_{i-\delta}-P_i)\Theta(i-\delta) + \gamma[P_{i+1}\Theta(N-i-1)-P_i] \quad \text{for } \frac{\delta}{2} < i \leq N , \tag{1}$$

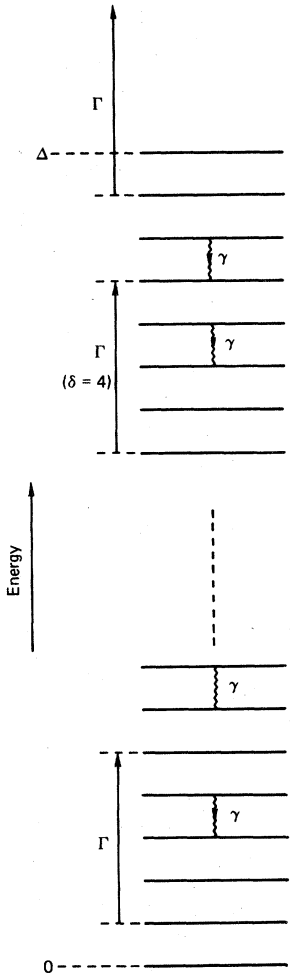

FIG. 2. Model used for the energies of conduction band (CB) electrons. The continuum of CB levels is replaced by a series of $N+1$ discrete steps in order to facilitate integration of the Eqs. (1)–(4) for the hot electron distribution. The photons cause transitions of δ steps, at rate Γ [cf. Eq. (5)], while the electrons can cascade down the steps (by phonon emission) at rate γ. The density of states is convoluted into Γ and γ. The energy Δ is approximately the band-gap energy. In this figure, $\delta = 4$.

$$\frac{d}{dt} P_i = \Gamma (P_{i+\delta} - P_i) + \gamma(P_{i+1} - P_i) + 2\Gamma P_{N+2i-\delta} \quad \text{for } 0 < i \leq \frac{\delta}{2} , \tag{2}$$

$$\frac{d}{dt} P_0 = \Gamma(P_\delta - P_0) + \gamma P_1 + 2\Gamma P_{N-\delta} + \rho\eta - K(N_0 - \eta)P_0 , \tag{3}$$

$$\frac{d}{dt} \eta = K(N_0 - \eta)P_0 - \rho\eta . \tag{4}$$

In these equations, Γ is the rate of absorption of photons by the CB electrons. We have

$$\Gamma(t) = \sigma I(t)/h\nu , \tag{5}$$

where σ is the absorption cross section, taken to be energy independent, I the intensity of the applied electric field, a function of time, and ν the radiation frequency. The quantity Γ given by Eq. (5) is an explicit function of time through the dependence of the intensity I on time. (Generally the laser pulses have a Gaussian temporal dependence.) The quantity Δ is the band-gap energy, taken to be the energy of the top level N of the ladder. Thus the energy between steps in the ladder is Δ/N. The electron advances δ steps at each absorption, so that ignoring the energy of the phonons in a phonon-assisted absorption step, we have

$$\delta = h\nu/(\Delta/N) . \tag{6}$$

The quantity γ is an average rate at which the electron loses the amount of energy Δ/N by phonon emission. More than one phonon may be emitted in this relaxation, depending on the number of steps used and the system being modeled. If it is desired to take into account the change of the phonon distribution due to heating, γ can be a function of temperature. Therefore, γ may be an implicit function of time.

The step function $\Theta(x)$ is 1 unless x is strictly less than zero, in which case it vanishes. Thus $\Theta(0) = 1$.

The rate ρ is the rate at which electrons are excited from donor levels to the CB bottom (i.e., to the step p_0), and $K(N_0 - \eta)$ is the recombination rate of the CB electrons with the donor levels. There are a total of N_0 donor sites. We expect $\rho = cI^n$, where c is some constant and an n-photon process is required to excite the donors.

We have ignored the trapping of electrons at hole sites (the holes having been created in the avalanche). The trap levels will be near the CB bottom and at the high excitation intensities of interest the difference in energy between these levels and the CB bottom can be ignored.

We now discuss several terms in Eqs. (1)–(4). The term $\Gamma P_{i-\delta}$ describes the absorption and the term $\Gamma P_{i+\delta}$ describes the stimulated emission. It is the inclusion of this latter term which distinguishes the form of the rate equations above from the equations of the classical theory in which the electron-radiation interaction is classical. The presence of the stimulated emission term insures that there will be no "current runaway" such as appears in the classical analysis.[20, 22, 46, 54, 55] It is therefore responsible for the fact that the electron multiplication rates have a much weaker dependence on the intensity of the field than in the classical theory.

The term $2\Gamma P_{N+2i-\delta}$ gives the rate of electron multiplication and so is the term responsible for the avalanche. An electron which has been excited to an energy greater than the Nth step is not permitted to fall below the Nth step by stimulated emission or phonon emission but rather excites an VB-CB electron transition and is deexcited to near the CB bottom. The $\Theta(N - i - \delta)$ and $\Theta(N - i - 1)$

step functions make sure of this process. This Auger process is presumed to take place instantaneously as soon as the electron gains sufficient energy.

The electron gas in the CB is assumed nondegenerate so Fermi statistics in the excitation from the donor or from the VB are neglected. Moreover, the change in dielectric constant due to the electron plasma resulting from the avalanche is neglected.

We have also neglected the electron-electron interaction, and we will indicate that for the range of parameters Γ, γ of interest this interaction is probably unimportant. This is done at the end of this section.

To calculate damage thresholds, we need to compute dQ/dt, the rate of heat generation. The heat comes from the phonon emission by the electrons as they relax. We ignore the energy released by recombination with donor sites, since the number of donor electrons is presumed to be too small to cause a significant temperature rise (cf. Sec. II). Thus for the rate of heat generation we have

$$\frac{dQ}{dt}=\frac{\gamma\Delta}{N}\sum_{i=1}^{N} P_i \ . \tag{7}$$

This can be rewritten

$$\frac{dQ}{dt}=\frac{\gamma\Delta}{N} Z\left(1-\frac{P_0}{Z}\right) , \tag{8}$$

where Z is the total number of CB electrons

$$Z=\sum_{i=0}^{N} P_i . \tag{9}$$

The condition for threshold damage is

$$\int_{-\infty}^{\infty} dt\ \frac{dQ}{dt}=C\Delta T \ , \tag{10}$$

where ΔT is the temperature difference between the melting point and ambient, and C is the specific heat of the solid.

If the rate γ is a function of the instantaneous temperature T, we require the subsidiary relation for the temperature

$$T(t)=T_0+\frac{1}{c}\int_{-\infty}^{t} dy\ \frac{dQ}{dy} \ , \tag{11}$$

where T_0 is the ambient temperature.

It is possible to coarse grain Eqs. (1)–(4) if $I(t)$ varies slowly enough. In this way, we can derive a simple equation for Z and P_0/Z directly. We now proceed to derive the conditions for this coarse graining.

Let us suppose that a characteristic time t_c exists such that

$$\frac{d}{dt}\ln I(t)\ll\frac{1}{t_c} \ , \tag{12}$$

i.e., $I(t)$ changes very little during a time t_c. We also assume that ρ, $K\ll t_c^{-1}$. The time t_c is really the time required for the Eqs. (1)–(4) to come to a steady state starting from any initial values of the set of p_i. Thus when I and γ are time independent, then for times larger than t_c the quantity $d\ln Z/dt$, which is the electron multiplication rate, will hardly vary, as will p_0/Z. That is, for I, γ time independent and when $t>t_c$, we have

$$\frac{d}{dt}\ln\frac{P_0}{Z}\cong 0 \ , \tag{13a}$$

$$\frac{d}{dt}\ln Z\cong\text{const.} \tag{13b}$$

For I time dependent the time scale for these quantities to vary significantly is

$$\left(\frac{d}{dt}\ln I\right)^{-1} .$$

Therefore, if Eqs. (12) and (13) are valid, the electron multiplication rate $d\ln Z/dt$ and the quantity p_0/Z will depend on the instantaneous value of $I(t)$ and will not depend on the history of the system or on the total number of CB electrons.

We will show later that Eqs. (12) and (13) are satisfied for the values of the parameters of interest.

This observation allows us to coarse grain Eqs. (1)–(4) to obtain a much simpler equation. It will be useful to define a reduced time t^* and a reduced transition rate f such that

$$f\equiv\Gamma/\gamma\equiv\sigma I/h\nu\gamma \ , \tag{14}$$

$$t^*=\gamma t \ . \tag{15}$$

We consider the set of *reduced equations*:

$$\begin{aligned}\frac{d}{dt^*}P_i(t^*)=&f[P_{i+\delta}\Theta(N-i-\delta)-P_i]\\&+f(P_{i-\delta}-P_i)\Theta(i-\delta)\\&+[P_{i+1}\Theta(N-i-1)-P_i]\quad\text{for } \tfrac{1}{2}\delta<i\leq N \ ,\end{aligned} \tag{16}$$

$$\frac{d}{dt^*}P_i=f(P_{i+\delta}-P_i)+(P_{i+1}-P_i)+2fP_{N+2i-\delta}\quad\text{for } 0<i\leq\tfrac{1}{2}\delta \ , \tag{17}$$

$$\frac{d}{dt^*}P_0=f(P_\delta-P_0)+P_1+2fP_{N-\delta} \ . \tag{18}$$

These equations are the same as Eqs. (1)–(4) but without the terms in η. We may solve Eqs. (16)–(18) when I and γ are time independent, and in this way we may determine the rates β, ϕ, defined as

$$\beta\{f\} \equiv \lim_{t^*\to\infty} \frac{d}{dt^*} \ln \sum_{i=0}^{N} P_i(t^*) , \tag{19}$$

$$\phi\{f\} \equiv \lim_{t^*\to\infty} P_0(t^*) \Big/ \sum_{i=0}^{N} P_i(t^*) . \tag{20}$$

The limit $t^* \to \infty$ actually means t^* much larger than the characteristic time t_c discussed previously.

The rates β, ϕ are central to our theory. The quantity β is the rate of electron multiplication, the central quantity in our theory. The notation $\beta\{f\}$ means that β depends on the instantaneous value of f and not on the previous history or initial conditions of the system. (Recall that I and γ are time independent for this initial calculation.)

The quantity $1-\phi$ is proportional to the rate of heat generation per electron, as shown in Eq. (8). The notation $\phi\{f\}$ means the same as for $\beta\{f\}$.

The quantities β, ϕ for the case $N=40$ and for several values of f, δ are given in Table I. The nature of these results and their application to laser damage and other hot-electron experiments is discussed in Sec. VIII.

Let us now show that the conditions (12) and (13) are satisfied for values of the parameters of interest.

In Fig. 3 we plot the approach of $(d/dt^*)\ln Z(t^*)$ to its steady-state value for the case $\delta=6, N=40$, and a few values of the rate f. To anticipate some results of Sec. VIII, these curves show that a time t_c can be found which fulfills the relations (12) and (13). For example, for the case $f=0.17$, the time t_c is such that $\gamma t_c = 50$. Now in NaCl the threshold intensity is such that $f=0.17$ for a 10-nsec pulse. We find that $\gamma \sim 10^{13}$ sec^{-1} to fit the NaCl damage data at 1.06 μm. Then $(d/dt)\ln I \sim 10^9$ sec^{-1}, while $1/t_c = 2\times10^{11}$ sec^{-1}, so that the relations (12) and (13) are both satisfied.

These results allow us to calculate damage levels for any known temporal beam profile $I(t)$ and for any dependence of γ on temperature. This comes about as follows. The equation for Z, the total number of CB electrons [cf. Eq. (9)] can now be written

$$\frac{dZ}{dt} = \gamma\beta\{f(t)\}Z + [\rho - K(N_0-\eta)]\eta , \tag{21}$$

where $f(t)$ depends on time through its dependence on the intensity $I(t)$ [cf. Eq. (19)]. We also have

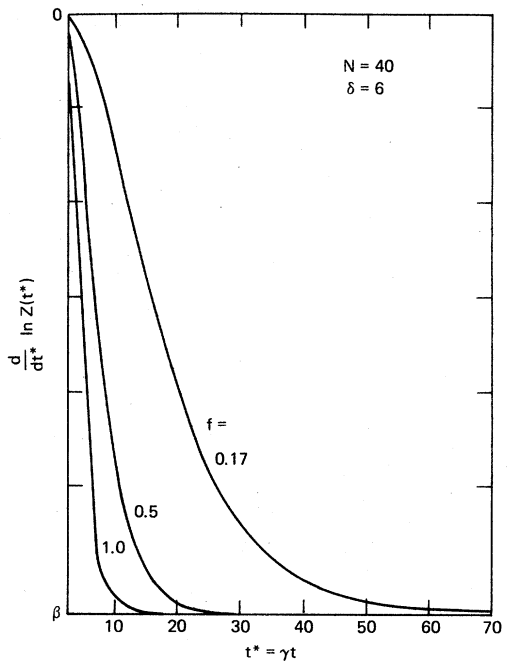

FIG. 3. Approach of $\ln Z(d/dt^*)\ln Z(t^*)$ to its equilibrium value β according to Eqs. (16)–(18). The quantity Z is the total number of CB electrons and β is the equilibrium electron multiplication rate, given by Eq. (19). Values of β are tabulated in Table I.

the additional equations

$$P_0/Z = \phi\{f(t)\} , \tag{22}$$

$$T = T_0 + \frac{1}{c}\int_{-\infty}^{t} dy\, \frac{dQ(y)}{dy} , \tag{23}$$

$$\frac{dQ}{dt} = \frac{\gamma\Delta}{N} Z\,[1-\phi\{f(t)\}] . \tag{24}$$

Equations (21)–(24) together with the condition (10) and the subsidiary relations (5), (6), and (14) and the results of Table I are all that are required to calculate damage thresholds from this model. One can integrate Eq. (21) to find the values of Z and Q, interpolating the results of Table I to find the appropriate values of β and ϕ for any f.

Finally, we indicate that the electron-electron interaction will probably not have much effect on our results. Of course, some model of the electron-electron interaction must be adopted since a rigorous calculation is out of the question. Therefore, we model the electron-electron interaction by assuming that it drives the system to a Boltzmann distribution.[23,56,57] We also assume a single relaxation time approximation. Thus we alter Eqs. (16)–(18) by addition of the term

$$(\psi/\gamma)[p_i(t^*)-y_i] \tag{25}$$

to each equation for dp_i/dt^*, where ψ is a constant rate for the relaxation due to the electron-electron interaction. The quantity y_i describes a Boltzmann equilibrium distribution, i.e.,

$$y_i = A a^i ,$$

where A and a are determined from the relations

$$\sum_{i=0}^{\infty} y_i = \sum_{i=0}^{N} P_i = Z ,$$

$$\sum_{i=0}^{\infty} i y_i = \sum_{i=0}^{N} i P_i = E .$$

The quantities Z and E are the total number of CB electrons and the total energy of these electrons, respectively. These conditions mean that the system is forced to a Boltzmann distribution with an electron temperature T determined by the energy E. The temperature of the electron gas may be obtained simply by solving the above equations for A and a, and noting that

$$a = e^{-\Delta/k_B T N} ,$$

k_B being Boltzmann's constant.

In addition, the avalanche rate is affected because electrons are transferred to energies greater than Δ by the electron-electron interaction. The rate of transfer of electrons to energies greater than Δ by the electron-electron interaction is

$$\psi \sum_{i=N+1}^{\infty} y_i ,$$

and therefore, Eq. (18) is modified by the addition of the term

$$\frac{2\psi}{\gamma} \sum_{i=N+1}^{\infty} y_i .$$

The quantities β, ϕ are now computed from these modified equations for the case $\delta = 6$, $N = 40$, and $\psi/\gamma = 0.6$. (This means that the rate of electron-electron relaxation is comparable to the rate of an electron cascading down two steps by phonon emission.) The revised values for β are compared with the values determined from the unmodified Eqs. (16)–(18) (i.e., the values of Table I) in Fig. 4(a). It is seen that the addition of a strong electron-electron interaction makes a very small effect on the avalanche rate. In particular, the qualitative behavior of β as a function of intensity I is not altered. It is also found that ϕ is unaffected. Therefore, in view of the uncertainties in calculating the electron-electron interaction and because of the small effect caused by the addition of another unknown parameter, it is probably best to neglect this interaction.

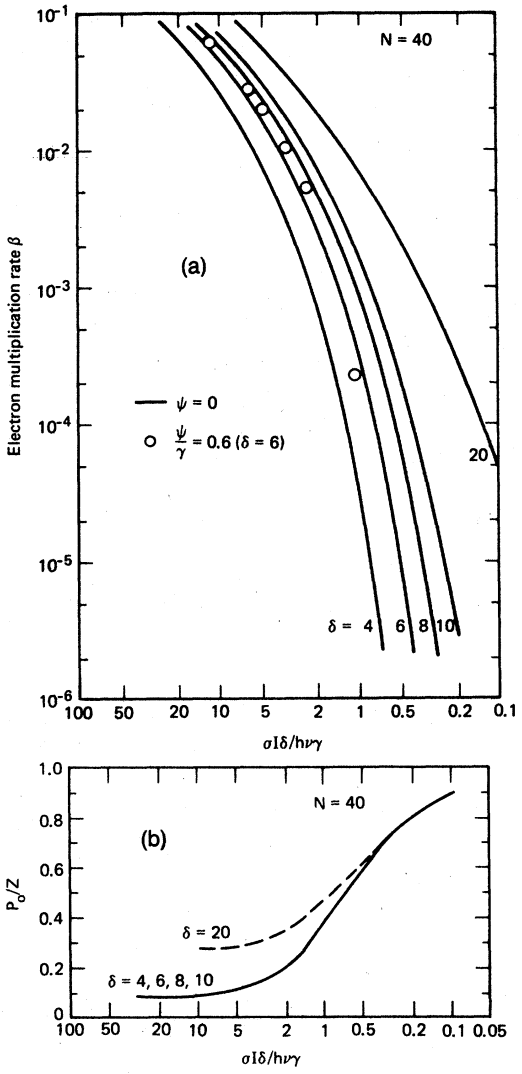

FIG. 4. Electron multiplication rate β, defined by Eq. (19), as a function of reduced intensity $f\delta$, for various values of δ, for the case $N = 40$. Note that the curves for $\delta \leq 10$ can nearly be superimposed. Note also the change in slope at $f\delta \sim 2$. The points (○) are the rates for $\delta = 6$ calculated for a modification of Eqs. (16)–(18) by the addition of an electron-electron interaction, as discussed at the end of Sec. VII. (b) The quantity $\phi = p_0/Z$, as defined by Eq. (20), as a function of $f\delta$. The curves for $\delta = 4$, 6, 8, 10 are nearly the same.

VIII. RESULTS AND COMPARISON WITH EXPERIMENT

The electron multiplication rate β is plotted as a function of $f = \sigma I\delta/h\nu\gamma$ in Fig. 4(a) for various

values of δ. The quantity $\phi = p_0/Z$ is plotted in Fig. 4(b) on the same horizontal scale. These quantities were evaluated by numerical integration of Eqs. (16)–(18), with β, ϕ defined by Eqs. (19) and (20), respectively. We now discuss these results.

For given $\sigma I/h\nu\gamma$ the electron multiplication rate β increases with increasing δ. This is reasonable when we recall that δ is proportional to the laser frequency ($h\nu = \Delta\,\delta/N$) and therefore the smallest number of photons required to excite an electron to band-gap energy Δ is the smallest integer greater than or equal to N/δ.

The shape of β as a function of $f\delta$ is not a very sensitive function of δ in the range $10^{-1} \geq \beta \geq 10^{-5}$ and $\delta \leq 10$, which turns out to be the region of experimental interest. This universality in the functional form of β as a function of $\sigma I\delta/h\nu\gamma$ leads to a kind of scaling law for laser damage. If we have a set of materials whose differences (for experiments of interest here) can be described simply by differences in the parameters γ, σ, ν, δ, Δ, then the electron multiplication rates as a function of the log of the intensity will differ only by a change of scale. This seems to have been observed for the alkali halides, where the damage thresholds I_c vs $1/\tau$ have a similar form when normalized to the NaCl results.[1]

Note also that, as a function of intensity I, the electron multiplication rate β becomes very steep for small I but much more gradual for larger I. The change in the slope occurs when the quantity

$$f\delta = \frac{\sigma I}{h\nu}\,\frac{1}{(\gamma/\delta)} \tag{26}$$

lies between 1 and 2. The quantity $f\delta$ is the ratio of two rates: The rate of absorption of photons, $\sigma I/h\nu$, and the rate γ/δ, which is the rate of emission of a total number of phonons whose energies equal $h\nu$, the photon energy. When these two rates become comparable, it means that the absorption rate is comparable with the energy loss rate so that high-energy CB levels can become significantly populated.

An analytic approximation to β, which leads to a qualitative understanding of the trends discussed above, can be obtained as follows. We reduce the model shown in Fig. 2 to its simplest form. We imagine the CB "steps" separated by exactly the photon energy $h\nu$. The rate of absorption Γ is given by Eq. (5), and is the rate to go from one step to the one above (absorption) or the one below (stimulated emission) under the influence of the applied field. The rate of phonon emission W is the rate of emission of a total number of phonons of energy equal to $h\nu$. Therefore we have $W = \gamma/\delta$.

Let us ignore the avalanche process for a moment and imagine an infinite number of such steps. Then the equations for the population of the steps are

$$\frac{d}{dt}P_i = \Gamma(P_{i+1} + P_{i-1} - 2P_i) + (\gamma/\delta)(P_{i+1} - P_i)$$

$$\text{for } i = 1, 2, \ldots, \tag{27}$$

$$\frac{d}{dt}P_0 = \Gamma(P_1 - P_0) + (\gamma/\delta)P_1\,, \tag{28}$$

$$\sum_{i=0}^{\infty} P_i = 1\,. \tag{29}$$

At equilibrium, which is attained rapidly, we have $dp_i/dt = 0$. The equilibrium solution to Eqs. (27)–(29) is

$$P_i = \frac{\gamma/\delta}{\Gamma + \gamma/\delta}\left(\frac{\Gamma}{\Gamma + \gamma/\delta}\right)^i\,. \tag{30}$$

Now for $\Gamma\delta/\gamma$ not too large, the avalanche process will not greatly distort the equilibrium distribution. Therefore, the rate of avalanche will be given by the rate at which electrons cross the energy Δ. This gives

$$\beta \cong \Gamma\frac{\gamma/\delta}{\Gamma + \gamma/\delta}\left(\frac{\Gamma}{\Gamma + \gamma/\delta}\right)^{\Delta/h\nu} \quad \text{for } (\Gamma\delta/\gamma \ll 1)\,. \tag{31}$$

The result (31) will become inaccurate for $\Gamma\delta/\gamma$ large because the equilibrium distribution will be considerably distorted by the avalanche. For large $\Gamma\delta/\gamma$ and an avalanche occurring we must have instead of Eq. (30) the relation

$$P_i \cong (h\nu/\Delta)\Theta(\Delta/h\nu - i) \quad \text{for } (\Gamma\delta/\gamma \gg 1)\,, \tag{32}$$

and therefore for large $\Gamma\delta/\gamma$

$$\beta \cong \Gamma h\nu/\Delta \quad \text{for } (\Gamma\delta/\gamma \gg 1)\,. \tag{33}$$

Consider now our numerical results in the light of these approximate formulas. For $\Gamma\delta/\gamma$ small, we have

$$\beta \sim \Gamma^{(\Delta/h\nu + 1)}\,,$$

and so β has a very sharp dependence on $\sigma I\delta/h\nu\Gamma$, as seen in Fig. 4(a). The values of $f\delta$ required for this exponential dependence are smaller than those in the scale of Fig. 4(a). On the other hand, as $\Gamma\delta/\gamma$ becomes large, $d\beta/d\Gamma$ tapers off so that β grows less steeply with intensity. The changeover in behavior will occur when $\Gamma\delta/\gamma \sim 1$, which is obvious from the expression (31) and which is

found numerically in Fig. 4(a).

For very large $\Gamma\delta/\gamma$, Eq. (33) predicts that β is proportional to $\sigma I\delta/h\nu\gamma$. This is seen for large values of $f\delta$ in Fig. 4(a).

Consider now the quantity P_0/Z in Fig. 4(b), which is related to the rate of heat generation per electron by Eq. (24). It is easily seen from Eq. (30) that for $\Gamma\delta/\gamma \ll 1$ we have

$$P_0/Z \cong 1 - \delta\Gamma/\gamma = 1 - f\delta \ . \tag{34}$$

This relation is satisfied by the values in Table I for the smallest values of f. Therefore, as the intensity I becomes small, the rate of heat generation per electron [Eq. (24)] beomes $\Gamma h\nu$, which is equal to the rate of absorption of energy, per electron, from the external field. We have used this result in Sec. II to estimate the number of CB electrons required to absorb enough energy to cause laser-induced breakdown.

On the other hand, as the intensity I becomes large, so that $\Gamma\delta/\gamma \gg 1$, we find from Eq. (32) that $p_0/Z \sim \delta/N$ (cf. Table I) so that the rate of heat dissipation per electron now becomes approximately $\gamma h\nu/\delta$, which is the fastest rate per electron that heat can possibly be generated by phonon emission. This rate will be smaller than $\Gamma h\nu$. Most of the absorbed energy is used up in the avalanche, which requires that energy Δ be expended in order to excite an electron from the VB to the CB. The saturation of heat generation rate was alluded to in Sec. II to infer that the bound on N_0 for short pulses (large intensities) is too small.

We now calculate the damage thresholds and rate of heat generation and electron multiplication for this model. We will assume here that at $t = 0$ a number of electrons N_0 already exist in the CB, so we will ignore the rate of generation of the primer electrons from donor levels. (We assume they are generated very rapidly.) This makes the calculation easier to interpret. The finite rate of generation of primer electrons will be discussed later. We also let γ be independent of the temperature. These approximations mean that in Eqs. (21)–(24) we set $R = \rho = K = \eta = 0$ and that Eq. (23) is needed.

The applied field has Gaussian time dependence

$$I = I_0 e^{-(t/\tau)^2} \ , \tag{35}$$

so that 2τ is the full pulse width at the $1/e$ point. We find from Eq. (21) that the number of CB electrons is

$$Z(t^*) = N_0 \exp\left(\int_{-\infty}^{t^*} \beta\{I(y)\}\, dy\right). \tag{36}$$

The criterion for damage is then

$$\int_{-\infty}^{\infty} dt^* \frac{\Delta}{N} Z(t^*)[1 - \phi\{I(t)\}] = C\Delta T \ . \tag{37}$$

We integrate Eq. (37) using Eqs. (35) and (36). The value of I_0 which satisfies Eq. (37) is then the threshold intensity I_c. We use the values $C = 2\times10^7$ erg/cm^3, $\Delta T = 10^3$ °C, $\Delta = 8$ eV, $N = 40$, and $\delta = 6$.

In Fig. 5 we plot the reduced threshold intensity $\sigma I_c/h\nu\gamma$ vs $1/\gamma\tau$ for the value $N_0 = 6.25\times10^{10}$ e/cm^3. The data for NaCl are shown as points in Fig. 5. The values of σ and γ required to fit the data are

$$\gamma = 9.71\times10^{12} \text{ sec}^{-1} \ ,$$

$$\sigma = 1.81\times10^{-17} \text{ cm}^2 \ .$$

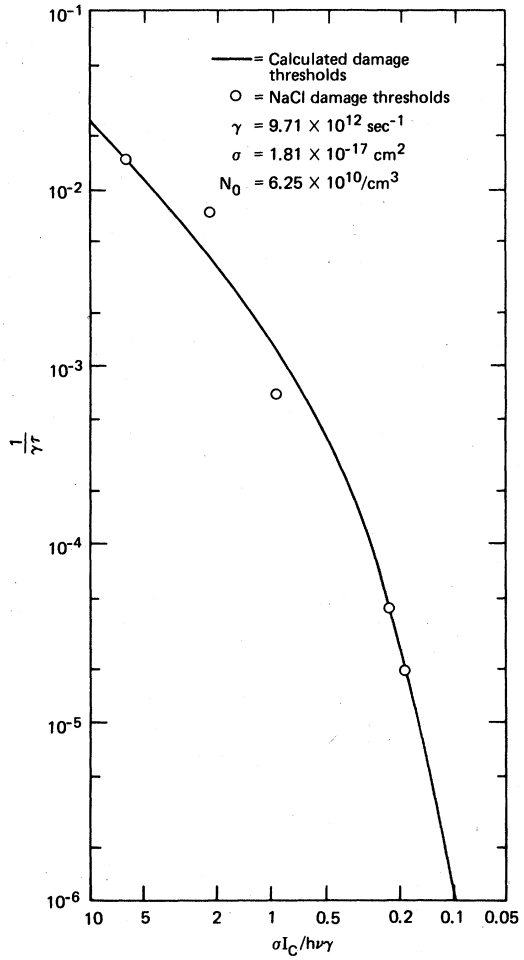

FIG. 5. Calculated damage threshold intensity I_c vs the inverse pulse duration τ in reduced units (solid line). The threshold intensity I_c is the value of I_0 from Eq. (35) such that Eq. (37) is satisfied. This means that the temperature of the irradiated volume is raised to the melting point. The points are damage thresholds for NaCl at 1.06 μm (cf. Fig. 1). The points are fitted by the theory for the values of the parameters listed on the figure.

Assuming a dominant phonon frequency in NaCl of 200 cm^{-1}, γ is the rate of generating energy equal to that of eight phonons. Then the average rate of emission of energy equal to that of a *single* 200-cm^{-1} phonon is 7.8×10^{13} sec^{-1}.

In Fig. 6 we give the calculated number of CB electrons excited for each primer electron for an avalanche occurring at the threshold intensity. It is seen, for the case $\delta=6$, $N_0=6.25\times10^{10}$ cm^{-3}, and γ and σ with the values above, that for all values of the intensity greater than 10^{10} W/cm^2 the electron multiplication is greater than a factor of 10^6, and therefore considerable avalanche is required for NaCl damage.

In Fig. 7 we present the computed damage thresholds as a function of the number of primer electrons for both $\delta=6$ and $\delta=10$ (band-gap energy = 6.7 and 4 photons, respectively). We see that over six orders of magnitude of primer electron concentration, the damage threshold intensities vary by less than a factor of 2. For real systems, this would indicate that damage thresholds are very insensitive to impurity content. The

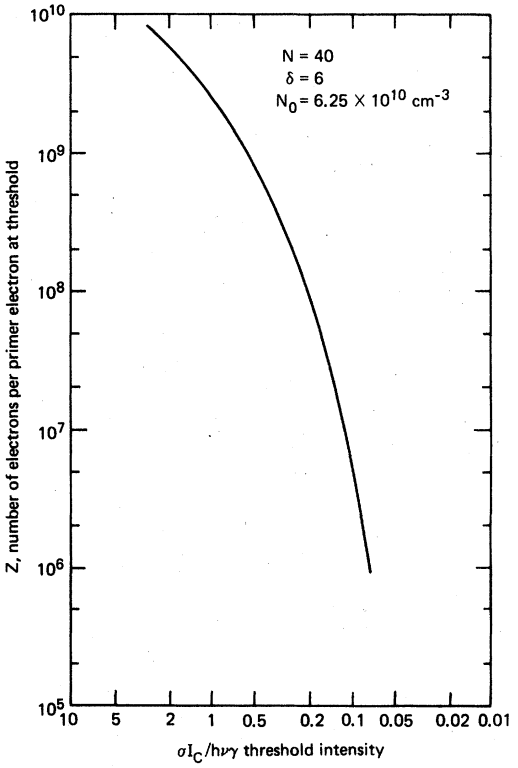

FIG. 6. Number of electrons Z excited in the CB for each starter electron, for a Gaussian pulse of threshold intensity. The same values of the parameters are used in Fig. 5. Therefore, this is our estimate of the number of CB electrons at the damage threshold in NaCl.

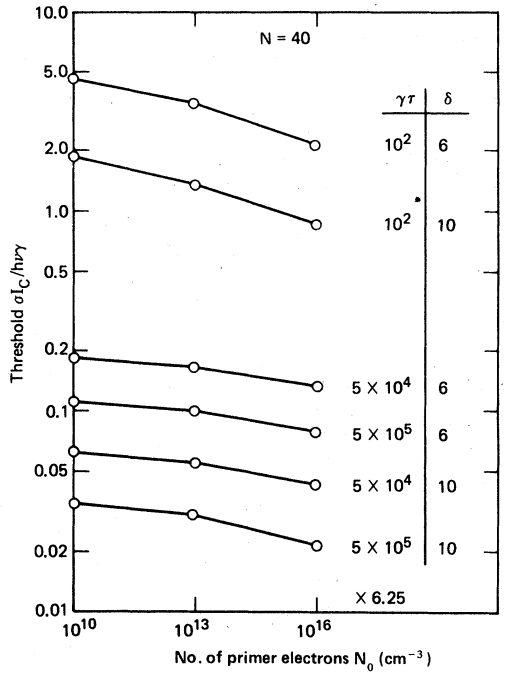

FIG. 7. Computed damage threshold intensities as a function of N_0, the number of primer electrons in the CB before the applied field is turned on. The threshold intensity is determined from Eq. (37). Curves are drawn for various values of pulse duration and δ. For NaCl and $\delta=6$, $\gamma\tau=10^2$ is about a 20-$psec^{-1}$ pulse.

reason is that, once an avalanche is begun, only a very small change in intensity changes the number of excited electrons by a huge amount. [This will be shown in more detail later (cf. Fig. 9).]

An interesting feature of Fig. 7 is that the threshold intensity I_c is somewhat more sensitive to N_0 for short pulses than for long ones. The reason for this is that the avalanche rate varies less rapidly with intensity at large intensities than at smaller ones, as seen in Fig. 4(a).

There are some experimental results which have a bearing on the curves of Fig. 7. Generally in a long-pulse damage experiment, it is observed that the threshold intensities measured at different places in the same sample are statistically distributed about some mean, and that this distribution is greater than the uncertainty in beam intensities.[59] Moreover, at short pulses such fluctuations are not observed.[1,2]

The fluctuation of thresholds for long pulse durations is probably not due to the fact that the source of primer electrons varies, so that in some parts of the sample there are less primer electrons available than in others. (According to Fig. 7, fluctuations in N_0 should be more apparent at shorter pulses than at larger ones, which is opposite to what is

observed.) Rather, it is probable[59] that the rate of excitation of primer electrons will fluctuate more at small intensities then at large intensities, especially if these electrons must be excited from donors by a multiphoton process. Thus the apparent fluctuations in N_0 will reflect the fluctuations in the rate of ionization of donors[59] and will be greater at longer pulses than at shorter ones, as I_c is smaller for longer pulses.

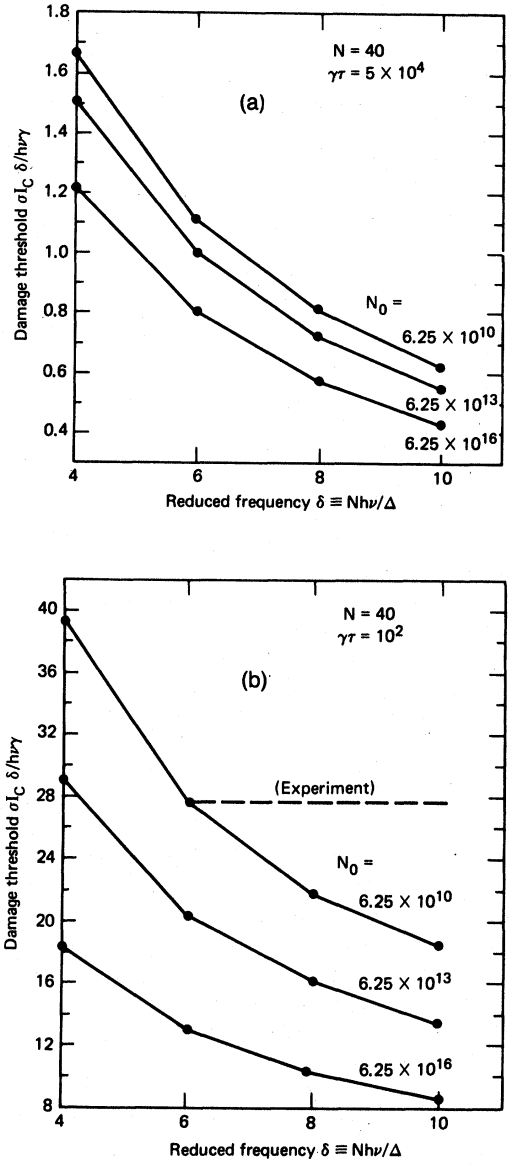

FIG. 8. Variation of damage threshold intensities as a function of δ for various values of N_0, the number of primer electrons. The threshold intensity is determined from Eq. (37). For NaCl, $\gamma\tau = 10^2$ is about a 20-psec^{-1} pulse. The dotted line in (b) is the experimental variation of I_c for NaCl as taken from the tabulation of Ref. 1.

Another possibility that must be kept in mind is that if σ is large enough then at long pulses damage can occur without avalanche. Recall that in Sec. II we showed that if $\sigma = 10^{-15}$ cm^2, then for $\tau = 10$ nsec damage requires no more than 10^{16} e/cm^3 in the CB. This damage mechanism may be expected to occur in materials with large numbers of color centers, or with trapping sites which lead to large values of σ.

Consider now the variation of I_c with frequency in Fig. 8. The horizontal axis is δ, which is proportional to the laser frequency [cf. Eq. (6)], while the vertical axis is $\sigma I_c \delta/h\nu\gamma$, so that the frequency dependence is eliminated. Note that according to the calculation there is a decrease in I_c with increasing frequency. One would expect this behavior on the basis of the model (cf. Fig. 2), since the larger the frequency the greater the energy imparted to the electron at each absorption step.

The experimental observations for NaCl are shown in Fig. 8(b), and it is apparent that there is some disagreement between theory and experiment, in that observed damage thresholds are either independent of frequency or actually increase with increasing frequency.[1] If we were to assume that primer electrons are excited by multiphoton absorption, the disparity between theory and experiment would be somewhat greater than is shown in Fig. 8 because the effective N_0 would increase with frequency.

There seems to be no way to rationalize the disagreement of Fig. 8 unless we were to let the cross section σ decrease with increasing frequency. If σ were inversely proportional to frequency over the optical range, the model would fit the data within experimental error. This is a rather *ad hoc* assumption (although no less *ad hoc* than allowing σ to be constant or even to increase with frequency).

Therefore, we feel that the disparity of Fig. 8 cannot be resolved at present. The best we can say is that our model does predict that the thresholds vary slowly with frequency, which is indeed the case, and which is a nontrivial conclusion.

IX. SUBTHRESHOLD EXPERIMENTS

Let us now consider predictions of the results of experiments done at subthreshold intensities, so that other quantities besides damage thresholds are measured. One quantity of interest is the total number of electrons generated per starter electron by a Gaussian pulse of intensity I_0 and duration 2τ [cf. Eq. (35)]. This quantity,

which can be determined in a photoconductivity experiment, is calculated to be

$$Z = \exp\left(\int_{-\infty}^{\infty} dt^* \beta\{I(t)\} \right) . \tag{38}$$

In addition, the total heat generated per starter electron during a pulse can be estimated from photoacoustic experiments and is calculated to be the quantity

$$Q = \int_{-\infty}^{\infty} \frac{\Delta}{N} Z(t^*)[1 - \phi\{I(t)\}] \, dt^* . \tag{39}$$

The quantity Z and $NQ/N_0\Delta$ are plotted in Fig. 9. For reference we note that damage occurs when

$$NQ/N_0\Delta \sim (6.25\times10^{22})/N_0 ,$$

which is 10^9 for $N_0 = 6.25\times10^{13}/\text{cm}^3$.

In order to apply the results of Fig. 9 to real solids, we must account for at least two aspects of solids that are not included in the calculation of Fig. 9. These are the finite rate of excitation

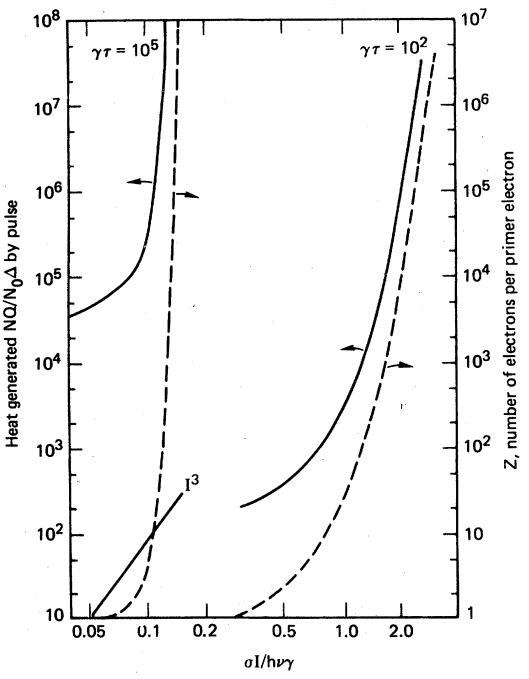

FIG. 9. Values of Q, the heat generated, and Z, the number of CB electrons per primer electron, as a function of subthreshold intensity I for a Gaussian pulse. The pulse is given in Eq. (35), where I_c is to be identified with I in the figure. The quantities Q and Z are calculated from Eqs. (38) and (39) of the text. Note that the rate of change of Q and Z with I decreases with decreasing τ. The experimental consequences of this behavior are discussed in Sec. IX of the text.

from donors, which depends on intensity, and also the rate at which CB electrons can be trapped at hole centers. This latter rate can be very large (of the order of psec^{-1}) when there is an appreciable concentration of CB electrons.[55]

Consider first a photoconductivity experiment. In such an experiment, electrons are excited to the CB and are swept toward an anode by a large dc field, of the order of kV/cm. Assume first long pulse durations and small intensities. For NaCl, these would be pulse durations greater than 1 nsec and intensities less than threshold, which is 1.8×10^{10} W/cm^2 at 10 nsec. Thus $\sigma I_c/h\nu\gamma \sim 0.2$ for a pulse such that $\gamma\tau = 10^5$. This pulse is about 10 nsec. As the field is raised from subthreshold values (10^9 W/cm^2 say) one initially records only electrons raised from donors. The number of electrons will go as I^n for n-photon excitation from donors to a CB energy for which the oscillator strength is large. (The exponent n may be nonintegral if a stepwise excitation occurs.) As seen in Fig. 9, no electron avalanche occurs until $\sigma I/h\nu\gamma \sim 0.1$ or an intensity about half that of threshold. Then the number of electrons begins to multiply quite rapidly. In order to observe this avalanche experimentally, however, several conditions must be met. First, the avalanche rate must be competitive with the rate of excitation of donors. If this latter rate goes as I^3, say, it would be of the form of the straight line in Fig. 9, and would be expected to overwhelm the initial stages of electron multiplication. Thus the avalanche may not be apparent until intensities are within 20% or 30% of threshold.

The electrons resulting from avalanche will be created largely near the peak of the laser pulse. It is these electrons that Alyassini and Parks[28] claimed to see in their experiments, since they observed a plasma created 1 nsec before the cutoff of the laser beam itself.

As the number of CB electrons increases, the lifetime against trapping becomes smaller, possibly of the order[55] psec at electron concentrations greater than 10^{18}/cm^3. Trapping does not influence Eqs. (1)–(4) as the trap level can be incorporated into the lowest step p_0. It is not likely that much trapping occurs during the course of the pulse itself, because the strong intensities used keep the electrons at energies above the trap level, and this transition will have a large cross section. However, after the pulse is ended recombination should be nearly instantaneous compared with the nsec time duration of the pulse, and so recombination will have a considerable effect on the measured photocurrent, decreasing it a great deal from what would be predicted on the basis of Fig. 9 alone.

In fact, for such a situation it may be impossible to observe an avalanche in the photocurrent even though one were occurring. The reason is that the photocurrent will behave something like

$$J \sim cI^n[1+aZ/(b+rZ)],$$

where cI^n gives the rate of generation of primer electrons from donors, Z is the number of electrons created per primer electron in the avalanche (and is the quantity plotted in Fig. 9) and rZ is the rate of recombination which we have set proportional to Z itself, which is apparently what is observed in insulators. The constants a and b are independent of the intensity. The effect of avalanche is only observed at intensities such that

$$\frac{aZ}{b+rZ} \geq 0.1 ,$$

depending on the resolution. Thus if $rZ > b$, the photocurrent is nearly independent of field and no electron multiplication is observable in the photocurrent.

There are two ways of getting around this problem. For one thing, one can increase the dc voltage on the sample to draw the CB electrons out of the traps. One can also apply a weaker second laser beam or uv irradiation which continues to excite electrons from the trapped states, making them available as a photocurrent. (This intensity should be too small to cause avalanche.) Then the effective trapping rate of electrons may be decreased to the point where an electron multiplication is measurable in the photocurrent.

It is tempting to apply this reasoning to the measurements of the laser-induced (~ 50-nsec pulse) photocurrent in LiF by Yasijima *et al.*,[29] where no charge multiplication was observed prior to breakdown. However, these workers did measure charge multiplication in KCl, so that the results are contradictory.

Consider now the behavior of the photocurrent in the short-pulse regime. For NaCl $\gamma\tau = 10^2$ implies a pulse of about 10 psec. Damage occurs at $\sigma I_c/h\nu\gamma \sim 5$ (when $N_0 = 6.25\times 10^{10}/\mathrm{cm}^3$). In this case the number of electrons created is a much more gradual function of pulse intensity and significant subthreshold avalanche occurs over nearly an order of magnitude of intensity. The avalanche should be competitive with excitation from donors at intensities from at least a factor of 3 or 4 below threshold. However, the remarks on trapping made for long pulses are equally applicable to short pulses.

Less unambiguous results will be obtained from measurement of electron multiplication at short pulses than at long pulses. At short pulses there is a broad range of fields over which avalanche occurs at an appreciable rate. At longer pulses a small fluctuation in intensity of a nominally subthreshold pulse will either cause damage or will decrease the avalanche rate by orders of magnitude. This should not be as much of a problem at shorter pulses.

Consider now the heat generated by a subthreshold pulse, shown by the solid curve of Fig. 9. Concerning the rate of change of Q with intensity, the previous remarks about Z are pertinent. In contradistinction to the case for Z, however, trapping should have a small effect on heat generated during the course of the pulse. Since Q can be determined by a calibrated photoacoustic experiment, such an experiment may well be a good way to measure the existence of an electron avalanche. In this case, in order to obtain unambiguous results it is preferable to use short pulses. The reason is that during damage a great deal of heat is generated, and one wants to be certain the intensity is subthreshold so that one is measuring a nondestructive electron multiplication. As explained previously, this is better done with short pulses, where dQ/dI is more gradual, than for long pulses.

Another experiment which can measure the presence of a subthreshold electron avalanche is a measurement of excited-state absorption. In cases where laser damage occurs, various experiments[26-28] show that a strong absorption is induced, presumably by the electron plasma created in the avalanche. However, as far as we are aware, no cutoff of the laser beam has been observed for subthreshold fields. The reason is probably because the total absorption is small. Suppose the focal region of the beam is 10-μm long. Then for the strong laser beams used, a change of intensity of at least 1% must occur in order to be measurable. Thus $\alpha d = 0.01$, where α is the absorption coefficient of the region where the avalanche occurs and $d = 10^{-3}$ cm. Therefore, $\alpha = 10\ \mathrm{cm}^{-1}$. Such absorption coefficients are obtained for long pulses only very near the damage threshold, when a very large number of electrons are excited. In the case of short pulses, an absorption coefficient of this magnitude can probably be obtained without causing damage. However, in order to detect subthreshold excited-state absorption, it would be useful to be able to detect changes in the intensity of a probe pulse of the order of 0.01%.

X. CONCLUSIONS

A phenomenological theory of laser damage has been presented and the results compared with ex-

periment. This theory is based on the existence of an electron avalanche whose rate β was derived. Laser damage thresholds for NaCl at 1.06 μm can be fit by assuming an average absorption cross section for hot electrons of about 1.8×10^{-17} cm^2, and an average rate of emission of an energy of 200 cm^{-1} of about 10^{14} sec^{-1}.

A central prediction of this theory is the saturation of the electron multiplication rate β as the intensity of the applied field becomes large, as shown in Fig. 4(a). The experimental consequences of this is that nondestructive avalanche should be observable for psec pulses at intensities at least between $\frac{1}{2}$ of threshold and threshold. That is, for psec pulses it should be possible to produce an electron avalanche whose extent is too small to produce damage but which should be detectable by a number of different experiments, such as photoconductivity, photoacoustic spectroscopy, or excited-state absorption. At present, the detection of a nondestructive avalanche is an important experimental problem. Unless it is possible to measure this phenomenon, no theory of laser damage due to electron avalanche can be considered as established.

It is possible to derive dc damage thresholds from our theory. As noted, the electron relaxation rate is 7.8×10^{13} sec^{-1} for 200-cm^{-1} energy. This quantity is to be used for τ in the classical expression of Sec. V. Then we predict a dc threshold intensity a factor of 3 smaller than observed.

There are several drawbacks to the present theory which should be brought out here. The most obvious is that the parameters—cross section and electron relaxation rate γ—are not derived from a microscopic model. Thus even though the values of these parameters are reasonable, the theory is still incomplete. Unfortunately, it is not likely that realistic calculations will be possible for some time.

A theory of the type presented here is not unique, in the sense that σ and γ probably can have different dependences on electron energy and laser frequency and still lead to substantially the same curves of Figs. 3–9. Thus as we noted, Sparks's theory also leads to good agreement with experiment. Sparks predicts the correct variation of threshold intensity with frequency, since σ used by him decreases with increasing laser frequency.

Another failure of this theory is the inability to correctly predict the variation of NaCl damage thresholds with frequency, as shown in Fig. 8. Thus we predict that damage thresholds decrease with increasing frequency while the observed thresholds are independent of frequency, or may increase slightly. If we let the cross section decrease with increasing frequency, we could fit the data, but all we can say at this point is that the true variation of cross section with frequency will have to await a microscopic approach. In any event, this lack of agreement is not significant enough to argue conclusively against the validity of the present theory.

In conclusion, we would like to stress the very reasonable values of σ and γ required to fit the NaCl damage data, which demonstrates that a physically plausible theory of laser damage can be constructed. It is hoped that this theory will provide an impetus to experimentalists to document the existence of a nondestructive avalanche for psec pulses, whose existence is central to the present theory.

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